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POLARIZED TARGETS

Gilbert Shapiro

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Berkeley, California

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1. Introduction
2. Methods of Obtaining Nuclear Polarization
   - A. Brute Force
   - B. Dynamic Nuclear Polarization
   - C. Optical Pumping
   - D. Static Methods
   - E. Other Methods
3. Description of the Berkeley Polarized Target
   - A. Choice and Preparation of Target Material
   - B. Cryogenics
   - C. Microwave System
   - D. Magnetic Field Requirements
   - E. Measurement of Polarization
     1. Thermal Equilibrium Signal
     2. Uniformity of Polarization Throughout Sample
     3. Non linearity of the Q meter
     4. Change in Line Shape
     5. Measurement of Polarization by Shift in Local Fields
     6. Nuclear Scattering to Measure Polarization
     7. Spin-Echo Technique
4. Further Discussion of Polarized Proton Targets
   - A. Other Materials Containing Hydrogen
     1. Solid Hydrogen

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1. Introduction.

The successful use of targets containing polarized nuclei in several types of scattering* experiments has encouraged many nuclear physics laboratories to consider the use of such targets in connection with their own facilities. This article is intended to serve as a guide to those who are interested in the construction or use of polarized nuclear targets.

Attention will be concentrated on the polarization techniques applicable to, and special problems arising from, nuclear scattering targets. Many other uses of polarized and oriented nuclei, particularly the study of the decay of oriented radioactive nuclides, have received much attention in recent years, but they are not considered within the scope of the present writing. For example, methods that apply only to microscopic samples, or to nuclei that are minor impurities in the material samples used, are clearly not useful for producing polarized targets. Emphasis here will be given to systems that have already proved themselves by being used in actual nuclear scattering experiments. Since nuclear reactions involving protons are considered (particularly in high energy physics) the simplest to interpret in terms of fundamental interactions, especial attention will be given to targets in which the polarized nuclei are those of hydrogen.

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* The term "scattering" as used in this article is to be understood to include nuclear reactions as well as elastic and inelastic scattering.

Review articles dealing with more general applications of polarized nuclei have appeared by AMBLER 1960 , ROBERTS and DABBS 1961 , STEENLAND and TOLHOEK 1957 and HUISKAMP and TOLHOEK 1961 . For the most part these articles discuss static methods of producing polarization. DANIELS and GOLDEMBERG 1962 discuss specifically targets of polarized nuclei, and treat in some detail the problem of the heat generated in the target by the incident beam. Dynamic nuclear orientation has been covered, with much attention to the solid-state aspects, in a recent monograph by JEFFRIES 1963 and a review article by ABRAGAM and BORGHINI 1964 . These are both excellent sources, containing much original material, and no attempt will be made to duplicate them here. Older reviews on dynamic methods include one by BARKER 1962 and by JEFFRIES 1961 .

The existence of polarized targets makes possible measurements of many nuclear scattering parameters that were hitherto impossible, or extremely difficult. Others can be measured with much improved accuracy. Without pretending to be exhaustive, since no doubt many more applications will arise, we list here some of the suggested uses of polarized targets.

a. GOLDFARB and BROMLEY 1962 have given a general formalism for analyzing the spins and parities of nuclear states (or, by extension, particle resonances) by scattering from polarized targets.

b. Double and triple-scattering parameters of nuclear collisions can be measured with one less scattering when a polarized target is used. In addition to providing the obvious advantages of higher rates and less dependence on precise geometrical alignment

of detectors, polarized targets make possible measurements in situations where second scatterings are inconvenient (e.g., when secondary particle has too low energy).

c. Time reversal invariance of the scattering interaction can be checked, in the case of elastic scattering, by comparing the target-polarization-dependent rate of certain experiments with the results of similar experiments using unpolarized targets in which the polarization of the final particles is measured. (See Appendix A)

d. In the scattering of unstable spinless particles (which can neither be polarized in a beam, nor used as targets themselves), certain spin-rotation parameters (KIM 1963) can only be measured by performing double-scattering experiments with a polarized target.

e. The relative intrinsic parities of strange particles, or nuclear states, can be measured directly by comparing the asymmetry in their production from a polarized target with the final-state polarization of these particles in the same reaction using an unpolarized target (BILENKY 1958).

f. ZICKENDRAHT et al. 1961 have analyzed the information to be gained from photodisintegration of polarized and aligned deuterons.

g. The limits of validity of the first Born approximation in analyzing electron-proton scattering (or electron scattering from any nucleus) can be explored by the use of polarized targets. Spin-dependent scattering arises from the exchange of two or more protons. (see BIZOT et al 1963)

h. The combination of polarized beams with polarized targets opens many more possibilities and combinations.

i. DANIELS and GOLDEMBERG 1962 review many nuclear physics applications involving reactions of low energy neutrons and gamma rays with oriented nuclei.

This brief listing is intended to give an indication of the versatility and wide applicability of polarized targets. There are no doubt other uses that will suggest themselves in the future, or that perhaps already have appeared in the literature. The multiplicity of applications may indicate some of the motives for construction of polarized targets by many laboratories (the author knows of projects underway or contemplated at, among other places, Harwell, Argonne, Los Alamos, and Rochester). While it may not be strictly true that every scattering experiment ever performed can be profitably repeated with a polarized target, there is still enough work to be done with them to keep many laboratories busy for many years.

Section 2 of this article deals with the various methods that have been used to produce working polarized targets. Section 3 concentrates on the polarized proton target that has been operating at the University of California in Berkeley. Some of the problems that arise in the operation of such a target (for example, the question of precise measurement of the amount of polarization obtained) are discussed. Section 4 deals with general problems arising in the experimental use of polarized proton targets.

2. Methods of Obtaining Nuclear Polarization.

A). Brute Force

In an external magnetic field, H, and in contact with a thermal bath at temperature T (but otherwise weakly coupled to its surroundings), a system of nuclei of spin I ($I \neq 0$) will be somewhat polarized. The polarization of the nuclear spins is defined as

$$P = \frac{\langle I_z \rangle}{I} = \frac{\sum_i m_i p_i}{I \sum_i p_i} \qquad (2.1)$$

where m_i is the magnetic quantum number of the i^{th} level, namely: the eigenvalue of the component of I (I_z) along the direction H · P_i is the relative population of the i^{th} level. In thermal equilibrium at temperature T, p_i obeys the Boltzmann distribution law.

$$p_i = \exp(-m_i \mu H / I k T) \qquad (2.2)$$

where is the magnetic moment of the nuclear species in question, and k is Boltzmann's constant. In particular, when $I = \frac{1}{2}$, one can readily deduce that

$$P(I=\tfrac{1}{2}) = \tanh(\mu H / 2 k T) \qquad (2.3)$$

When the nuclei in question are protons, the magnetic moment is such that when $T = 1^{\circ}$ K and H = 10 kOe, one calculates P = .001.

The magnetic moments of all other nuclei are of the same order of magnitude as that of the proton. Therefore, very low temperatures combined with rather strong magnetic fields are required to obtain sizable polarizations by this direct method. SCHERMER, et al., 1961 have actually obtained proton polarizations in the neighborhood of .02 by cooling palladium hydride to a temperature of

.07° K in a field of 17.5 kOe. This polarization was large enough for them to detect and measure the difference in transmission of a beam of polarized low-energy neutrons through the sample when the neutron spin direction was alternately parallel and anti-parallel to that of the protons (see Section 3E for further discussion of this effect). Presumably one can obtain higher polarizations simply by using stronger magnetic fields or going to lower temperatures.

This method of polarization does not seem to be applicable to solid hydrogen. The ground state of the hydrogen molecule is parahydrogen in which the proton spins are required, by the Pauli exclusion principle, to be anti-aligned. Orthohydrogen molecules with proton spins parallel, can be incorporated into solid hydrogen by rapid freezing from high temperatures. A considerable refrigeration problem is presented by the spontaneous ortho- to para-hydrogen conversions, which release 180 calories per gram of ortho-hydrogen converted. Even if this is spread over many hours or days, the heat load is far beyond the capacity of most adiabatic demagnetization systems, which are needed to maintain the low temperatures required.

The "brute force" method is of interest even if it is not the primary means of obtaining polarization. Many of the methods of measuring the polarization involve using the thermal equilibrium polarization as a calibration point. Much of the literature on dynamic nuclear orientation quotes "enhancements," the ratio of polarization obtained to that of thermal equilibrium, as the result. The polarization is then obtained by multiplying this

enhancement by the thermal equilibrium value given by equation (2.3).

B). Dynamic Nuclear Polarization.

The method of dynamic nuclear orientation has been the most successful way of producing high proton polarizations. It has also been applied to many other nuclear species. Excellent review articles have recently been published by ABRAGAM and BORGHINI 1964 and by JEFFRIES 1963 , so there will be no attempt in this article to make an exhaustive discussion of the essentially solid-state physics aspects of the technique. Nevertheless, a brief discussion of the important features of this method must be given here.

One begins with recognition of the essential fact that a free electron has a magnetic moment 660 times that of the proton. Therefore, at convenient temperatures and fields (say, 1° K and 10 kOe) the electrons will have a thermal equilibrium polarization in excess of 0.50. OVERHAUSER 1963 suggested one way of transferring this polarization from the conduction electrons in a metal to the nuclei. Radiation was to be supplied at the Larmor frequency of the electrons of sufficient power to saturate the transition. The electrons, now depolarized, would regain their polarization through mutual-spin-flip collisions with the nuclei of the lattice. If the other mechanisms by which nuclear spins couple to the thermal bath are relatively weak, this process can continue until the nuclear polarization is equal in magnitude to the thermal equilibrium polarization of the electrons. The existence of the Overhauser effect was soon verified by CARBER and SLICHTER 1956 .

To date no nucleus has been polarized in bulk samples to more than .005 by the simple Overhauser effect; references may be found in JEFFRIES' book 1963 .

With the success of the Carver-Slichter experiments, a number of authors pointed out that the method was not restricted to the saturation of electron spin resonance but was a technique which should be quite widely applicable to paramagnetic materials having the right kinds of coupling. The names of ABRAGAM 1955 and JEFFRIES 1957 have been associated early and often with developments along this line. In principle one may use as a starting point any paramagnetic center that can be treated as if it were an isolated magnetic dipole with magnetic moment comparable to that of a free electron. Among the paramagnetic centers that have been successfully used in obtaining dynamic orientation are: (a) free radicals, in solution or incorporated into solids; (b) F-centers and other damage centers created by radiation bombardment; (c) unfilled inner shells in rare-earth and transition elements.

JEFFRIES 1957 showed that nuclear polarization could be produced through saturation of the partially forbidden transitions in which electron and nuclear spins simultaneously undergo a change in projection quantum number. In contrast to the direct Overhauser effect, one here forces the nuclear reorientation by the application of microwave power. The paramagnetic centers (briefly referred to as "electrons") are then repolarized by means of some other mechanism that couples them strongly to the thermal bath. The nuclei, being otherwise weakly coupled, retain the polarization

transferred to them. Jeffries used the salt lanthanum magnesium double nitrate-- $La_2Mg_3(NO_3)_{12} \cdot 24\ H_2O$, in which part of the magnesium was replaced by cobalt. The cobalt nuclei were then polarized, using the unfilled d shell of the cobalt atom as the paramagnetic center.

ABRAGAM and PROCTOR 1958 , and independently ERB, MOTCHANE and UEBERSFELD 1958 , made the next important step by showing that the paramagnetic center and the nucleus being polarized need not belong to the same atom. The coupling between the center and the nucleus need be no more complicated than magnetic dipole magnetic dipole, sufficient to provide a mechanism for the partially forbidden mutual-spin-flip transitions to take place. The recognition of this fact, which has been named the "solid effect", made it possible at once to consider the possible polarizaton of any nucleus at all by dynamic methods. The paramagnetic centers, even though introduced as minor impurities in the solid, can be used to polarize nuclei that outnumber the centers by factors of 10^3 or more. This can happen because of a secondary spin-diffusion process among the polarized nuclei. Because mutual spin-flips between identical nuclei can take place without interchange of magnetic energy with the lattice, these occur rapidly. The polarization diffuses outward from the nuclei most closely coupled to the paramagnetic centers to the more remote nuclei. When the relevant relaxation rates are in the right ratio, all the nuclei of a given species in a certain solid can be highly polarized by relatively few paramagnetic centers.

The "solid effect" has proved the most useful method for obtaining high proton polarizations. Important papers in the development of

this method include those by ABRAGAM, MC CAUSLAND and ROBINSON 1959 HWANG and SANDERS 1960 , BORGHINI and ABRAGAM 1960 , LEIFSON and JEFFRIES 1961 , and SCHMUGGE and JEFFRIES 1962 . The reviews by JEFFRIES 1963 and BORGHINI and ABRAGAM 1964 include many new data arising from more recent work.

Figure 1 illustrates the energy level diagram appropriate to a system consisting of one paramagnetic center coupled weakly to a single neighboring nucleus in an external magnetic field. The frequencies indicated refer particularly to the system of a neodymium impurity center in a lanthanum nitrate crystal, coupled to a nearby proton, in an external field of 9.1 kOe. These are the conditions used in the report of CHAMBERLAIN et al. 1963 , with the Berkeley polarized proton target. The argument applies to a much more general situation.

The magnetic dipole-dipole coupling is assumed to be so weak that it does not appreciably effect the energy of any of the levels. The magnetic energy, E_n, of any level is given by

$$E_n = m_e \Delta - m_p \delta \qquad (2.4)$$

where m_e and m_p are the spin projection eigenvalues of the paramagnetic center and of the polarizable nucleus, respectively,

$$\Delta = g_e \mu_o H \qquad (2.5a)$$

$$\text{and} \quad \delta = g_p \mu_n H \qquad (2.5b)$$

where μ_o and μ_n are the Bohr magneton and the nuclear magneton, H is the external magnetic field, and g_e and g_p are numerical factors describing the magnetic properties of the particular elements involved. The dipole-dipole coupling does ensure that the energy eigenstates are not true eigenstates of m_e and m_p, but that there is

a small admixture of $m_e = + \frac{1}{2}$ in the nominal eigenstates of $m_e = - \frac{1}{2}$ and vice versa. This admixture makes possible the "forbidden" transitions shown by the dotted lines in Fig. 1.

The "allowed" transitions, in which m_e reverses and m_p remains the same, are assumed to be closely coupled to the lattice. Relaxation times may be measured typically in milliseconds. This means that for our purposes we may consider two levels connected by an allowed transition to be always in thermal equilibrium with the bath at temperature T. The relative populations of two such levels will be given by the Boltzman distribution.

$$\frac{P(m_e = + \frac{1}{2})}{P(m_e = - \frac{1}{2})} = e^{-\Delta/kT} \qquad (2.6)$$

Whatever disturbance in the populations that may take place, these level pairs will quickly readjust themselves by interaction with the lattice to the distribution of equation (2.6).

Microwave power is applied at the frequency of one of the forbidden transitions, of sufficient power to saturate this transition. Then the populations of the levels connected by the forbidden transition will be equalized. Let the relative populations of these two levels be unity. Then the populations of the remaining levels can be determined from equation (2.6), since each is connected to one of the saturated levels by an allowed transition. Having all the relative populations, one can then compute the nuclear polarization from equation (2.1). The results are shown in Fig. 1 for each of the two forbidden transitions. One notes that

1. The ideal nuclear polarization is given by $\pm \tanh (\Delta/2kT)$, the magnitude of the thermal equilibrium polarization of the

paramagnetic centers alone. (i.e., the high polarization of the latter is transferred to the nuclei.

2. Either sign of polarization can be obtained by proper choice of which forbidden transition to saturate. This involves only a small change in microwave frequency. All other factors-- magnetic field intensity, geometrical arrangement, etc.--remain unchanged. This is of considerable benefit when the polarized nuclei are being used as a target, since many experimental systematic errors can be cancelled out by making use of this feature.

Certain limitations on the use of this method of polarization must be mentioned here.

(a) The question of how strongly the ions must be coupled to the lattice to guarantee that equation (2.6) holds is answered by the requirement

$$\frac{T_p}{T_e} \gg \frac{N_p}{N_e} \qquad (2.7)$$

where T_p and T_e are the spin-relaxation times of the polarized nuclei and the paramagnetic centers respectively, and N_p/N_e is their relative abundance in the target. Equation (2.7) sets a lower limit on the concentration of paramagnetic centers required. Since the relative relaxation rates may be temperature-dependent, there may be an optimum temperature for polarization. The relaxation rates may also depend on the impurity concentration.

(b) Microwave power at the allowed transition frequency depolarizes the paramagnetic centers, and it is to be avoided. The allowed line has a finite width, and so there will be an appreciable tail to it existing at the frequency of the forbidden transitions. To minimize

loss of polarization due to this effect, one requires that the separation between allowed and forbidden lines, namely δ, be as large as possible compared with the line width of the allowed transition. The line width is generally only slightly dependent on the external field strength, whereas δ is linearly proportional to H. Therefore, high magnetic fields are desirable, for this as well as other reasons. Line broadening can occur when paramagnetic centers interact with each other, and this factor tends to set the upper limit on desirable paramagnetic center concentration.

The first use of a dynamically polarized proton target in a nuclear scattering experiment was by ABRAGAM et al. 1962 . Using both a polarized beam of protons and the polarized target, this group measured the spin-correlation parameter C_{nn} in proton-proton scattering at 20 MeV. The work of the Berkeley group with a similar target will be described in Section 3.

C. Optical Pumping.

A recent series of papers by workers in Texas-- WALTERS et al. 1962 , and SCHEARER et al. 1963 -- reports on high polarizations induced in He^3 nuclei by a method employing optical pumping. This system has the advantages of operating at room temperature, and employing an isotopically pure sample. So far, however, the method has been successfully applied only to a gaseous sample at pressures near 1 mm Hg.

Metastable 2 $^3S_1$ He^3 atoms are formed in the He^3 cell by a weak electrodeless discharge. The level of the discharge, maintained by a 50 Mc/sec oscillator, is kept as low as possible to still be self-sustaining, since this situation produces the best

polarizations. A He^4 lamp is used to provide light at the 1.08-μ wavelength corresponding to the $2\,^3S_1 - 2\,^3P_o$ transition. The light is circularly polarized along the direction of a weak magnetic field. The direction of the induced He^3 polarization is either parallel or anti-parallel to the direction of the pumping light, depending on whether the light is right or left-hand circularly polarized.

The optical pumping depopulates one of the m_J levels of the $2\,^3S_1$ state, leaving the metastable atoms partially polarized. The absolute polarizaton values depend greatly on such factors as the experimental geometry, lamp intensity, and cell discharge level. The metastable atom polarization is then transferred essentially completely to the He^3 nuclei of ground-state atoms via the metastability-exchange collisions:

$$He^3 + He^{3*} \longrightarrow He^{3*} + He^3$$

where the asterisk indicates the metastable atom. These transitions often occur in such a way that the incident and emerging ground-state He^3 atoms have magnetic quantum numbers differing by ± 1, while the corresponding metastables differ in their magnetic quantum numbers by ∓ 1. By means of such mutual-spin-flip processes, it is argued by WALTERS et al. 1962 , the ground-state He^3 nuclei are driven to the same degree of polarization as that of the metastable atoms.

SCHEARER et al. 1963 , report nuclear polarizations as high as $40 \pm 5\%$. The cell volume is 65 cm^3. The polarization is measured by three methods, which yield mutually consistent results.

(a) The amplitude of the magnetic resonance signal of the He^3 nuclei is compared with that of protons in benzene.

(b) The polarization of the $^3S_1$ levels is determined by the transmission of the 1.08- μ light through the cell.

(c) In the experiment by PHILLIPS et al. 1962 , the nuclear scattering of He^4 by He^3 is tested for polarization dependence at energies and angles at which the relevant nuclear scattering parameters have been determined by other experiments.

Maximum polarization is obtained at pressures of 1 mm Hg and below. At pressures above 10 mm Hg the polarization obtained is negligible. Nevertheless, the experiment of PHILLIPS et al. demonstrates that, in situations where intense beams are available and cross-sections are large, it is feasible to use even this tenuous gas as a nuclear scattering target. SCHEARER et al. 1963 , suggest that, inasmuch as the He^3 relaxation time in this target is of the order of 10 minutes, attempts might be made to compress the gas after it has been polarized.

This group intends (G. C. Phillips, private communication) to pursue experiments with He^3 targets using beams of protons, deuterons, alpha particles, and polarized neutrons.

D. Static Methods.

The most widely used methods of producing nuclear polarization have been those employing the static equilibrium polarization that occurs at low temperatures in properly chosen materials. An external constant magnetic field may also be employed. The brute-force method described in Section 3A falls

into this category. More sophisticated techniques make use of the local fields present at the nuclear sites in many solids.

(a) Pound alignment (POUND 1949) arises from coupling of the nuclear electric quadrupole moment with local electric field gradients.

(b) Bleaney alignment (BLEANEY 1951) arises from magnetic hyperfine coupling with local magnetic fields.

(c) Rose-Gorter polarization (ROSE 1949 ; GORTER 1948) arises from magnetic hyperfine structure plus an externally applied magnetic field. The nucleus to be polarized is coupled to a paramagnetic center with a coupling strength that is large compared with kT. The external field is such that the magnetic energy of the electron in this field is much larger than the hyperfine coupling, which in turn is much stronger than the direct coupling of the nucleus to the external field. Under these circumstances the electrons are highly polarized. The strong effective local field at the nuclear site due to the electrons leads to high nuclear polarization.

(d) More recently many workers have made use of the strong local fields existing in ferromagnets and anti-ferromagnets to obtain nuclear alignments and polarization.

One may note that some of these methods require no external magnetic field, which may be a useful experimental consideration. However, in such circumstances the expectation value of any component of the nuclear spin (an axial vector) must vanish, so the polarization as defined in equation (2.1) is zero. For nuclei with spin greater than $\frac{1}{2}$,

however, one can define P_2, the second moment of polarization, or "alignment", as

$$P_2 = \frac{1}{I(2I-1)} \left[3 \langle I_z^2 \rangle - I(I+1) \right]. \quad (2.8)$$

When $P_2 \neq 0$, one may expect anisotropies in the rates of certain nuclear processes with respect to the axis of alignment.

AMBLER et al. 1962 have performed an experiment with aligned Ho^{165} nuclei in holmium metal, a ferromagnet. The anisotropies in the (γ, n) nuclear reaction in the region of the giant dipole resonance were investigated.

Polarized and aligned nuclei have been used experimentally to study radioactive decay processes, to investigate properties of solids, to obtain very low temperatures, and in other applications that are not within the scope of a review of scattering from polarized targets. The reader is referred to the articles of AMBLER 1960 and ROBERTS and DABBS 1961 for information about the many uses of nuclear polarization.

Experiments involving as targets nuclei polarized by static methods have largely involved the transmission of polarized neutrons through the targets. Early work was done by the Oak Ridge group (DABBS and ROBERTS 1954 ; ROBERTS et al. 1954 ; BERNSTEIN et al. DABBS et al. 1955 , some of which was extended by STOLOVY 1960). Recently an experimental program at Brookhaven (POSTMA et al. 1962 ; SAILOR et al. 1962 ; MARSHAK et al. 1962 ; SCHERMER 1963 POSTMA et al. 1964) has been devoted to systematic study of the transmission of polarized neutrons through polarized targets. The spins of nuclear levels have been determined, as well as other properties of the

structure of resonances in these systems. In some cases hyperfine coupling constants in the solids used were measured in the course of these experiments. The methods used to obtain polarization usually fell under categories (c) and (d) above. Among the nuclei that have been used as polarized targets are Mn^{55}, In^{115}, Sm^{149}, Eu^{151}, $Gd^{155,157}$, Ho^{165}, Co^{59} and Tb^{159}.

The amounts of polarization obtained in these targets are sometimes quite large. For example SAILOR et al. 1962 obtain polarization of 0.476 in holmium nuclei, in an 874-mg sample of holmium-indium alloy, at 0.071° K in an external field of 17 kOe. Polarizations in the range from 0.10 to 0.15 were obtained in cobalt metal (23 grams, 0.1°K), samarium salts (double nitrate and ethylsulfate, 0.15°K, 10 kOe) and in terbium metal (0.95°K, 17 kOe). In both holmium metal and holmium ethylsulfate crystals at 0.95°K, nuclear polarizations in the range from 0.15 to 0.25 were obtained in external fields of 17 kOe.

The polarizations obtained at 0.95°K are significant, since this temperature can be maintained with a He^4 cooling system, with its considerable heat capacity. The lower temperatures were obtained using paramagnetic salts, usually iron ammonium alum, as the cooling agents. The latter system has limited heat capacity. The heat load arising just from the ionization loss by a charged particle beam traversing the target would strain the capabilities of such a system. The static methods seem to have been mostly applied to rare-earth and transition-metal elements, because these elements are the ones with strong hyperfine couplings in the solids they form. The methods can probably be extended to the actinide elements. So far no application seems to

have been made to the elements at the beginning of the periodic table (but see the result of SCHERMER et al. 1961 discussed in Section 2A).

E. Other Methods.

CLARK and FEHER 1963 have proposed a method, which has been discussed theoretically by WEGER 1963 to obtain nuclear polarization by application of a direct current to certain semi conductors. This method has the attractive feature that no microwaves are required to produce the nuclear polarization. In a sample of InSb, T = 4.2°K, H = 12.7 kOe, polarizations as much as 100 times thermal equilibrium were obtained (still less than $\frac{1}{2}\%$ polarization because of the low starting value). The basic idea is that the temperature characterizing the distribution of electron spins is made different from the temperature that characterizes their transitional motion. This may be done in several ways, the simplest of which is to "heat" the electrons by passing a direct current through the sample. The "hot" electrons constitute the thermal reservoir which supplies the energy for one of the mutual-spin-flip transitions between electrons and nuclei. The hot electrons fulfill the same function as one of the "forbidden" transitions in Fig. 1, tending to equalize the populations of the respective levels.

No experiments have yet been done using this technique to produce a polarized target. We await developments with interest, since this technique not only eliminates the necessity for microwave power, but also extends the possibilities for nuclear polarization to a new class of elements.

JEFFRIES 1963a and ABRAGAM 1963 have proposed a method that accomplishes the same objectives as dynamic nuclear polarization, but which eliminates the necessity for microwave power. The paramagnetic centers are to operate as the working substance of a refrigerator which pumps heat from the nuclear spin system to the lattice thermal bath. The thermal switches that alternately connect the centers to the lattice and to the nuclei are to be represented by relaxation rates which depend strongly on the orientation of the crystal. An appropriate sample is rotated with respect to an external magnetic field, and the refrigerator goes through its cycles, resulting in high nuclear polarization.

These methods are appealing for their originality, and have some advantages over existing methods of nuclear polarization, as well as some drawbacks. As yet, however, they do not seem to be ready for application to the construction of polarized targets.

3. Description of the Berkeley Polarized Target

A target of dynamically polarized protons has been in use at the Lawrence Radiation Laboratory since late 1962 (CHAMBERLAIN et al. 1963, 1964). This target consists of 26 grams of the lanthanum magnesium nitrate crystal, in which proton polarizations in the neighborhood of 60% can be regularly achieved. This target has been used to measure the double-scattering parameter P in pion-proton interactions at 246 MeV (SCHULTZ and CHAMBERLAIN et al. 1963) and in proton-proton scattering between 2 and 6 BeV (STEINER et al. 1964).

This section contains a general description of the construction and operation of this particular target. While other polarized targets that have been or will be built may differ in detail from the Berkeley target, the problems that have appeared are representative of those likely to be encountered with any such target, and particularly targets containing dynamically polarized hydrogen nuclei. When different approaches to some of the same problems have been employed by other groups, these are also included in the discussion.

A. Choice and Preparation of Target Material

The material used in the Berkeley target is lanthanum magnesium double nitrate -- $La_2Mg_3(NO_3)_{12} \cdot 24\ H_2O$ -- with a small neodymium doping. There were several reasons for this choice, not the least of which was that sizable proton polarizations had already been obtained in this and similar crystals by BORGHINI and ABRAGAM 1960 , and by SCHMUGGE and JEFFRIES 1962 . The crystal contains a relatively large amount of hydrogen. Large single crystals of this

material are easy to grow. The properties of the material have been much studied and are fairly well understood.

The small fraction of neodymium ions that replaced some of the lanthanum provide the localized paramagnetic centers required by the dynamic nuclear polarization method. Neodymium ions have an odd number (three) of unpaired inner-shell (4f) electrons, in the ground-state $i_{9/2}$ configuration. The different m_J levels within this configuration are split by the crystalline electric field. At the temperatures, near 1°K, that these crystals are used, only the $m_J = \pm \frac{1}{2}$ levels are populated appreciably. This pair of levels is known as a Kramers doublet; its behavior can be approximated as that of an immobile spin-$\frac{1}{2}$ object having magnetic moment comparable to that of a free electron.

Neodymium was chosen in preference to other rare earth elements with unpaired inner electrons (e.g., cerium) for several reasons. The ESR line of Nd is sharper in large crystals than that of other ions. The spin-lattice relaxation time of neodymium is more favorable for obtaining high polarizations. The rather high g factor of the neodymium ion -- as high as 2.70 -- makes it possible to use a lower magnetic field strength to obtain a given splitting of the doublet. This splitting is determined by the frequency of the microwave generator available, independently of the properties of the crystal.

Natural neodymium contains about 20% odd-A isotopes, i. e., nuclei with non-zero spin. When the dynamic polarization process is applied to ions containing such nuclei, the ESR line becomes split to such an extent that these atoms do not participate in the polarization process, but can serve as centers for the relaxation of proton spin polarization.

To avoid this possibility, enriched even-isotope neodymium salts (available from Stable Isotope Division, Oak Ridge National Laboratory) were used. The neodymium in the enriched salts consists 98.5% of even-A isotopes, principally Nd^{142}. Use of this preparation also helps reduce contamination of all other rare-earth elements.

The optimum concentration of neodymium is not known exactly. Many factors can control the performance of different individual crystals--such as unwanted impurities, lattice defects, alignment with respect to magnetic field, etc. Carson D. JEFFRIES (University of California private communication) reports 2-1/2% doping is not as good as 1%, and both 5% and 0.1% are definitely worse. Too high a concentration is to be avoided, lest the paramagnetic centers cease to be dilute, and interact appreciably with one another, resulting in undesirable line broadening. The crystals used in the Berkeley target were grown from a solution that was originally .01 Nd/La atomic concentration. The crystals do not necessarily have this ratio, since the kinetics of crystallization are such that the lanthanum ions incorporate more rapidly into the lattice structure, per unit concentration in solution, than do the neodymium ions, by a factor of perhaps two or three. The part of the crystal formed at the start is therefore reduced in neodymium concentration. As time goes on, however, the process of selective crystallization tends to increase the neodymium concentration in the liquor. When large single crystals are grown from a limited volume of solution, the part of the crystal grown last may have a local concentration of neodymium even greater than that of the original solution.

The crystals are grown from saturated solution in a desiccating jar, kept at 0°C to assure slow uniform growth. The seed crystal is placed on a narrow pedestal raised from the bottom of the crystal-growing dish so that the crystal grows freely in all directions (except straight down. There is an indentation in one face of each crystal because of this pedestal). Lanthanum magnesium nitrate crystals grow in hexagonal platelets (see Figure 2) with a thickness of about one-quarter the diameter. Crystals of the size shown take 2 or 3 weeks to grow.

In order to make a target of roughly a 1-inch cube, four of these crystals are stacked together. It is important that all four be aligned with their crystal planes parallel. The crystals are highly anisotropic; the g-factor of the neodymium ion depends upon the angle between the external magnetic field and the normal to the flat face of the crystals: $g = 2.70$ when this angle is 90°, but $g = 0.4$ at 0°. If one hopes to polarize all four crystals simultaneously, one must make sure that the field makes the same angle with all the crystals. Crystals are selected for having flawless flat faces. Kel-F grease (which contains no hydrogen) is used to make them adhere to each other, and pressure is applied to hold them together by means of the rf pickup windings wrapped around the crystals. The field is always aligned in the 90° position (i.e. parallel to the crystal faces) so that the g factor is least sensitive to slight misalignments -- this orientation is also the best for experimental purposes. The remaining dimensions of the crystal may be easily cut to any desired shape with a wet cotton thread, the crystals being highly soluble in water.

When not in use the crystals are stored in a jar containing concentrated sulfuric acid as a desicant. Precautions must be taken, when removing crystals from the cryostat, not to expose them to air until

they are fully warmed up to room temperature. Otherwise ice may condense on the crystals and subsequently cause damage to them by dissolving some of the material.

It is good practice not to allow any hydrogenous material in the neighborhood of the crystal, for experimental reasons. First, particle scattering by the extraneous hydrogen cannot be distinguished from scattering by the protons in the crystal, but the former will not yield any polarization-dependent effect. The result will be to reduce the magnitude of any measured effect from its true value. Second, if nuclear magnetic resonance is used to measure the polarization of the target, the extraneous hydrogen may contribute to the signal observed, particularly at thermal equilibrium, in such a way as to make the target polarization appear smaller than it actually is. These two effects are in directions that partially cancel, but the two methods of sampling the protons are far from uniform. Some error is almost surely introduced by the presence of extraneous hydrogen, and is to be avoided.

B. Cryogenics

The Berkeley polarized target is maintained at a temperature near $1.2^{\circ}K$ by being immersed in a bath of liquid He^4 at a vapor pressure below 1 mm Hg. The cryostat, a diagram of which is shown in Figure 3, has a capacity of between 15 and 20 liters. The vacuum insulating jacket and the liquid-nitrogen-temperature ($77^{\circ}K$) radiation shield are of standard design. A six-inch-diameter steel conduit connects the helium reservoir to a high-speed Kinney KC-310 pump. The pumping speed is 10 cubic meters (310 cubic feet) per minute.

The latent heat of liquid He^4 is such that a dissipation of 1 watt in the liquid will boil away 1 liter per hour. If there is negligible pressure drop in the pumping line, and one assumes that the gas has warmed up to near 273°K by the time it reaches the pump, one can calculate the increase in reservoir pressure caused by each additional watt of dissipation. One liter of liquid helium at a density of 0.15 g/cm^3 vaporizes into 840 liters of gas at STP. One liter liquid per hour therefore, becomes 14 liters per minute of gas at STP. When this is compared with the pumping speed, one obtains

$$\frac{P}{760 \text{ mm Ng}} \text{(per 1 watt dissipation)} = \frac{14 \text{ } \ell/\text{minute}}{10{,}000 \text{ } \ell/\text{ minute}}$$

or P = 1 mm Hg for each watt of dissipation. The temperature change, near 1.3°K; corresponding to 1 mm increase in vapor pressure is about 0.1°K.

When microwave power is not being applied to the target, the principal source of dissipation is heat conduction through the walls of the reservoir, the liquid helium transfer line, the brass waveguide, and rf leads that extend into the bath. The effect of these is minimized by making the first two of stainless steel, and the others as thin as possible. One section of the waveguide has been filed down from the outside to minimum thickness. Baffles are placed in the neck of the reservoir both to intercept radiant heat from the warm flange at the top, and to make efficient use of the boiled-off vapor to cool the upper parts of the reservoir. The amount of heat leaking through these sources depends considerably on the level of the liquid; almost half of the useful running time takes place after the level indicators show that the reservoir proper is empty and helium remains only in the long

section that extends downward into the magnet. (The level indicators are carbon resistance thermometers -- 1/4 watt, nominally 100Ω Allen Bradley resistors, whose resistance increases five-fold at liquid helium temperatures.) Under steady conditions the conduction heat loss is estimated to be between 300 and 500 milliwatts. This can no doubt be improved upon, but for this target it is somewhat less than the power dissipated when microwave power is applied, and so it is considered acceptable.

During the pumping-down from 4.2°K to 1.2° K, 45% of the liquid helium must be boiled to cool down the remaining fluid. With no microwaves a full reservoir can last more than 12 hours. When microwaves are on, the average time between fills is about four hours. A certain fraction of the liquid is inevitably lost during the initial transfer. The overall liquid helium consumption of the Berkeley target when it is running full-time can be as much as 100 to 150 liters per day. This is probably the largest single operating expense ina target this size. Liquid helium is available commercially in the United States at a delivered cost of $8.00 per liter. A laboratory considering the use of such a target had better consider carefully its sources of liquid helium. The helium can be recirculated to conserve natural resources. At present the major part of the expense, however, is the cost of liquefaction, and not the cost of helium gas.

The lower part of the cryostat is so designed that it presents negligible extraneous material in the path of any beam of particles to be used in a scattering experiment. The lower part of the helium reservoir and a beam window in the outer wall of the vacuum insulating jacket

are made of 0.1-mm-thick special-strength aluminum alloy. Materials containing hydrogen are to be avoided in the region of the beam, for reasons given in Section 3A. Magnetic materials are also to be avoided in this region lest the field homogeneity be disturbed.

In this target the liquid helium circulates through slits in the waveguide so that it comes directly in contact with the surface of the crystals. This presents no particular experimental problem when a thick target can be used; the amount of helium in the beam is small compared to the heavy elements already in the target. When energy loss in the target is an important consideration, however, the helium must be kept out of the beam's path. ABRAGAM and BORGHINI 1964 discuss their solution to the problem of cooling a thin crystal polarized target.

Heat dissipation due to the beam traversing the target is negligible in the Berkeley target. A minimum-ionizing particle deposits 10 MeV in passing through a 1-inch-thick crystal. If the time-averaged beam intensity is 10^8 particles per second, (which is larger than necessary for most applications) the heat generated is 0.16 milliwatt.

There is presumably an optimum temperature at which to operate a crystal of this nature to obtain maximum polarization. There are reasons (see JEFFRIES 1963 or ABRAGAM and BORGHINI 1964) to expect that at low enough temperatures the relaxation rate of the paramagnetic ions to the lattice falls off faster than the proton relaxation rate. When this happens the dynamic polarization process becomes inefficient, and the attainable polarization decreases. In the experience with the Berkeley target, this theoretical limit has not been reached. Every decrease in temperature, down to 1.2°K results in an increase in the polarization. NEGANOV et al. 1963 have carried the process down to 0.5° K using a liquid He^3 refrigeration system. They report proton polarizations of 8%, using a 6 times 6 times 2 mm lanthanum magnesium nitrate crystal doped with 0.8% cerium in magnetic fields of 3.5 kOe with microwaves of 9 GH_z. With similar equipment, but at 1.7°K, LEIFSON and JEFFRIES 1961 obtained 3% polarization. Clearly something can be gained by going to these lower temperatures.

A liquid He^4 cooling system can be operated below 1° K if the heat load is small, and a fast enough pump is available. AMBLER, DOVE, and

KAESER 1963 have constructed a cooling system employing liquid He^3. The lowest target temperature that can be obtained is $0.28^\circ K$ with no extraneous heat input and about $0.4^\circ K$ with a maximum heat of 2 mW. The apparatus requires only 1.5 STP liters of He^3 gas for its operation. These figures represent conditions during an actual experiment with a polarized target: measurement of the (γ,n) cross-section for aligned Ho^{165} nuclei (AMBLER et al. 1962).

To attain even lower temperatures, paramagnetic salts must be used as the cooling agent. The major difficulty (which applies to He^3 cooling systems to some extent also) is the limited cooling power. Most paramagnetic systems cannot hold their temperature against a heat input of as little as a microwatt. DANIELS and GOLDENBERG 1962 have made an analysis of the heat likely to be dissipated by the particle beam itself in various nuclear reactions. Experiments with charged particle beams seem to be ruled out with such cooling systems. One does not rule out the possibility that some systems may be adapted to pulsed operation, in which cooling and polarizing cycles alternate with short periods of particle-scattering measurements.

The Brookhaven Group (SAILOR et al. 1962 , MARSHAK et al. 1962) regularly obtain temperatures in the region 0.05° to 0.10°, using potassium ion alum as the cooling salt. Rare-earth and transition metals are polarized by static methods and used in transmission experiments with polarized neutrons. In particular, SCHERMER 1963 reports that a 23-gram sample of cobalt metal was maintained below $0.1^\circ K$ for five hours, after an initial cooling-down time of 1 hour, while the neutron beam was incident on the target. These results are quoted to indicate the sort of performance that can be expected from these cooling systems, under actual experimental conditions.

It is not expected that any of these cooling systems, except liquid He^4, can at present provide sufficient cooling power for targets that require substantial microwave input, or that are intended to be used with charged-particle beams. There are likely to be exceptions to this statement, however, particularly when very thin targets are to be used.

C. Microwave System.

The thermal equilibrium polarization of the paramagnetic ions is given by tanh ($h\gamma_e/2kT$), where γ_e is the frequency of the "allowed" ESR transitions. Once the frequency range of the microwave signal generator is selected, this factor--which represents the maximum attainable polarization--is determined, independent of magnetic field or g factor of the ions. In fact, it has been found at Berkeley that each increase in microwave frequency has led to a higher polarization of the target. At the present operating conditions γ_e = 70 GH_z and T = 1.2°K, yielding tanh ($h\gamma_e/2kT$) = 0.90, so perhaps little more remains to be gained in this direction.

There are two factors that in general set upper limits on the microwave frequency. One is the maximum available magnetic field intensity; the product of this H with the g value of the paramagnetic ions used determines the maximum microwave frequency. The other determining factor, important in the case of large crystal samples, is the amount of microwave power that can be attained with available signal generators. With improvements in the state of technology (e.g., superconducting magnets) one can expect both of these limits to be pushed higher. One may also note that there exist paramagnetic ions with very large g factors. Erbium impurities in lanthanum ethylsulfate have g = 8.8. Such an ion would have an ESR frequency of more than 200 GH_z in a field of 20 KO_e.

A diagram of the microwave circuitry used with the Berkeley target is shown in Figure 4. The signal generator is a "carcinotron" COE 40, manufactured by CSF, ORSAY, France. This generator puts out up to 40 watts of power, and is electronically tunable from about 69 to 71 GH_z. An isolator, or Y-circulator, serves to protect the generator from reflected power. A specially built attenuator, capable of dissipating large heat loads, controls the amount of power delivered to the target. Directional couplers permit monitoring of the frequency and relative power level of the input, and aid in locating the ESR signal from the spectrum of power reflected by the cavity. A switch (not shown in the diagram) which can reroute the power into a matched load is useful in the circuit at this point. A rotating joint can be placed where the waveguide bends into the cryostat, to permit rotation of the assembly

to obtain the optimum orientation of crystal with the field. As explained in 3A, the Berkeley target, consisting of four single crystals, worked well only in the 90° orientation. A vacuum seal must be placed in the waveguide at the last joint before the cryostat; a 0.1 mm mylar wafer, together with an O-ring, serves adequately for this purpose.

The waveguide used from this point on is of the 8 mm size. This makes it possible to accommodate lower frequency microwaves when desired. Also the attenuation is less at 70 GH_z in this size guide, if the waves propagate in the lowest mode, than in 4 mm waveguide. This section of guide is 2 meters long. Propagation in higher modes is possible, but does not appear to present any disadvantage in this case.

The large size of the crystal makes it necessary to use a high-mode cavity. In fact, no attempt was made to tune the cavity to any mode at any frequency. At the end of the waveguide a horn opens up into a rectangular box 2.5 times 2.5 times 10 cm in dimensions. Such a large cavity can be compared to an echo chamber in which the radiation bounces from wall to wall until it is either absorbed or escapes back up the waveguide. In practice less than one-tenth of the incident power at the top of the 2-m guide returns to that same point by reflection. Some is absorbed by the waveguide walls, but there is evidence that most of the power is absorbed in that region of the system that is below the liquid helium, i.e., crystals and cavity. One specific advantage of the "echo chamber" vs. tuned cavity is that in the former, one may expect that the power is distributed rather uniformly throughout the sample, so that all parts are polarized equally. If any nodes exist (and their existence may be questionable in the presence of strong absorption) the low-intensity regions occupy a rather small volume in a high-mode cavity, and are closely surrounded by points of high field intensity. The disadvantage of such a chamber is that it is "low-Q" and therefore does not make most efficient use of the power available.

Some discussion is necessary of how much power is required to obtain maximum polarization in a given size crystal at specified frequency,

temperature, etc. The heat that must be pumped can be determined from the rate at which the polarization returns to its thermal equilibrium value when the microwaves are turned off. This follows an exponential decay with a time constant of about 10 minutes. An energy of $h\gamma_e$ must be expended to repolarize each proton. In a 26-gram (3% hydrogen) sample, the energy required to maintain 60% polarization is

$$Q = (0.03 \times 26 \text{ grams}) \times (1/2 \times 0.60) \times (6 \times 10^{23}) \times h\gamma_e = 7 \text{ joules.}$$

The calculated power input, is

$$\frac{dW}{dt} = \frac{7 \text{ joules}}{600 \text{ sec}} = 11 \text{ milliwatts.}$$

It seems clear that the efficiency is far from ideal. One obvious loss mechanism stems from the fact that the allowed ESR transition has finite line width. At the position of the forbidden transition the "tail" of the allowed transition is still so great that allowed transitions far outnumber forbidden ones. The effect of this competition is simply that much microwave power is used to heat the crystal, and under unfavorable conditions to reduce the polarization of the paramagnetic ions. No doubt there is also some power dissipated by eddy currents in the cavity walls.

Some empirical estimates of the power requirements may be of use to the reader. One must keep in mind that absolute measurements of microwave power are often difficult or inconvenient to make at these frequencies. From the rate of helium boil-off it appears that the optimum power input for the Berkeley target lies between 1/2 watt and 2 watts. At one time a klystron signal generator was used whose factory-rated maximum output was 170 mW. Its actual output may have been closer to 100 mW. With this generator working full blast, the polarization achieved in the 26-gram sample, with untuned cavity, was about 20%. The same klystron was later used at maximum output by JEFFRIES 1963 to obtain 65% polarization in a 375-mg crystal, using a tuned cavity with a Q value of more than 1000. It is interesting to note that NEGANOV et al. 1963 obtained 8% polarization in a similar size crystal (6 x 6 x 2 mm) with only 1-mW of microwave power. The tuned cavity had $Q \sim 1000$.

An upper limit to the amount of power that can be used is set when the temperature of the crystal rises sufficiently to reduce the amount of proton polarization. There is a possible thermal runaway that can take place under these circumstances. The paramagnetic ion-lattice relaxation rate increases rapidly with temperature, so the absorption of microwaves increases, thus contributing to further heating of the crystal. Proton relaxation rates also increase with temperature. Onset of thermal runaway is signalled by the abrupt disappearance of all polarization, usually within seconds. Short of this catastrophe the immediate effect of an increase in microwave power is an increase in proton polarization. But in the long run a lower power level may produce better results than a higher one because of the lower crystal operating temperature it makes possible.

D. Magnetic Field Requirements

When the dimensions of the target are not dictated by experimental considerations, the principal limitation on the size of a dynamically polarized target is set by the homogeneity of the magnetic field. The useful volume for the target is that region of the magnet gap in which the field intensity differs from the central value by less than the linewidth of the microwave "forbidden" transition. The permissible deviation in the lanthanum magnesium nitrate crystal, is about 1-Oe, independent of total field intensity. It is usually an advantage, for the sake of obtaining high polarizations, to have the field as strong as possible. For conventional iron-core electromagnets this is about 20 kOe. One therefore requires a field homogeneity, over a volume of several cubic centimeters, of one part in 20,000. These are the specifications met by the magnet for the Berkeley polarized target.

One also requires uniformity with respect to time. The Berkeley magnet is provided with current regulation of better than one part in 10^5 for currents up to 1700 amp, with a power rating of 150 KW. One pitfall that should be noted arises when the target is to be used while located in the stray field of other pulsed magnets, in particular, that of a synchrotron. The effect of the pulsed stray field must be

negated either by shielding or by compensating currents initiated through the voltage (rather than the current) feedback loop of the regulator. This means that this loop must have sufficient gain to make this compensating at frequencies down to perhaps 0.1 cycle per second, depending on the behavior of the pulsed magnets. No particular problems are posed by D. C. stray fields.

The Berkeley magnet is mounted with respect to the cryostat as shown in Figure 5. The magnetic field direction is horizontal. The beam enters horizontally and perpendicular to the field direction. Scattered particles can be detected when they emerge in or near the plane perpendicular to the field.

Experimental considerations can dictate many of the properties of the magnet. If low-momentum charged particles are involved, one may require that the product of the field intensity by its lateral extent be small. One may desire to bring in particle beams parallel to the field direction, or to detect scattered particles that emerge directed toward the pole face. Beams may be available with particles polarized along a vertical axis, and require that the target polarization be also vertical. All of these considerations lead to design problems of considerable complexity.

It may be appropriate to remind the reader that some of the static methods of nuclear orientation mentioned in Section 2D (not applicable to protons, however) require no magnetic field at all.

E. Measurement of Polarization

The interpretation of any nuclear scattering experiment employing a polarized target usually requires accurate knowledge of the exact amount of nuclear polarization.* It is therefore important to be able to measure the polarization with a precision that has not usually been sought by the solid-state physicists who developed the technique. When the target

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* There are some experiments in which precise knowledge of the target polarization is not crucial. Such experiments might be those in which the effect of reversing polarization is nearly zero, or the counting statistical errors are large, or the experiment is one in which only the algebraic sign of the effect is sought.

is large, or the polarization high, special problems of measurement arise which require considerable corrections to the raw data to be applied, in order to deduce the true amount of polarization. In the case of the Berkeley target it is estimated that, given the uncertainties in these corrections, the true value of the polarization lies within 15% of the calculated value (i.e., a quoted polarization of 0.60 has a range of error of $\pm$ .09). This amount of precision has proved satisfactory in the analysis of experiments performed with this target until now, in which the counting errors have been quite a bit larger than 15% of the observed effect. When more accurate results are required, it is expected that improved methods of applying the corrections to the polarization measurement--involving straight-forward but tedious calculations--will yield more precise values.

The proton polarization is most commonly measured by determining the strength of a nuclear magnetic resonance signal produced by these nuclei. A small amount of radio-frequency power, at the frequency corresponding to spin transitions of free protons in the external magnetic field, is circulated through the crystal. The equality of transition probabilities for absorption and induced emission guarantees that the rate of power absorption in a nuclear resonance experiment performed with a fixed number of nuclei (other factors remaining the same) is proportional to the nuclear polarization, defined as

$$P = \frac{I_z}{I} = \frac{N_+ - N_-}{N_+ + N_-}$$

The apparatus used in this measurement is shown in Figure 6. This is a standard Q-meter circuit, which has the advantage over other nuclear magnetic resonance detectors of giving a response that, in first

approximation, is linear in the polarization. In general, some compromise must be made between linearity and sensitivity.

1. Thermal Equilibrium Signal

The nuclear magnetic resonance method, as well as most of the other methods to be mentioned in this section, of measuring nuclear polarization suffers from a lack of direct absolute calibration. Such calibration would require knowledge of matrix elements, geometrical factors, and the gain of various amplifiers, etc., that is difficult to obtain. This difficulty can be circumvented by comparing the size of the signal observed with the target highly polarized with that of a similar signal observed when the proton spin system is in thermal equilibrium at the temperature of the helium bath. The polarization at thermal equilibrium is given by the brute force value:

$$P_o = \tanh (h\nu_p/2kT)$$

One notices immediately that the measurement of the polarization is limited in accuracy by the uncertainty in the thermal equilibrium value. One needs a good signal-to-noise ratio, not merely to be able to see the thermal equilibrium signal, but to measure its magnitude to within a few per cent.

Thermal equilibrium is recognized when, with microwave power turned off, the signal does not change with time. Since proton spin lattice relaxation times are of the order of many minutes, verification that thermal equilibrium exists can be a time-consuming procedure. The fastest way to achieve equilibrium is to warm the sample up to 4.2° K (by stopping the pump and letting in Helium gas). Proton relaxation

times in lanthanum magnesium nitrate are only a few seconds at this temperature. When the sample is again cooled down to 1.2° K the polarization stays close to its equilibrium value down to about 2°. It reaches 90% of its final value within about five minutes of the bath attaining its final temperature.

The temperature of the bath is determined by measuring its vapor pressure with an oil manometer, and referring to standard tables (VANDIJK 1960). Sensitivity is such that, at 1.20° K, an increase of 1 mm of oil column (sp grav = 1.04) corresponds to .01° rise in temperature.

2. Uniformity of Polarization Throughout Sample

The target polarization measured by an electromagnetic technique will correspond to the average polarization of the nuclei actually struck by the beam if any two of the following three conditions is met. (a) All portions of the target are equally polarized. (b) The beam illuminates the target uniformly. (c) The measuring technique is equally sensitive to all parts of the target. In general, none of these conditions is completely satisfied.

The high-mode microwave cavity is likely to provide, on a coarse-grained level, uniform radiation of the sample. This need not be so, however, if power is absorbed near the surface of crystals so strongly that the amount reaching the interior is reduced. Magnetic field inhomogeneity can lead to reduced polarization in regions where the field differs appreciably from its central value.

The beam spot can be designed to cover the whole crystal if desired. However, the illumination will be strictly uniform throughout the sample only if the spot is made so large that a sizable fraction of it misses the target completely.

The nuclear magnetic resonance pickup coil used in the Berkeley target is shown in Figure 7. The coil is wound as two figure-eights in series, above and below the crystals, inside the microwave cavity. The thin copper septum extending across the middle of the cavity forces the magnetic field lines generated by the coil currents to circulate completely around the septum, looping all four turns of the coil and providing a fairly uniform rf field intensity throughout the crystals.

Tests were made with a small sample of water, at room temperature, that was moved about within the volume occupied by the coils. The signals generated with the water sample at the most and least sensitive spots differed by a factor of 2; intermediate sensitivities are evenly distributed through the volume. A calculation based on these data shows that under the most extreme circumstances (100% polarization in the most, or least, sensitive half of the voluem; zero elsewhere) the volume-averaged polarization differs from the sensitivity-averaged (i.e., measured) polariztion by less than 0.10. Under any real circumstances the discrepancy is likely to be much less.

The insulation about the pickup coils is made of teflon tubing. It is particularly important to avoid hydrogenous material close to the wires, since the nuclear magnetic resonance is most sensitive in this region.

3. Non linearity of the Q-Meter.

In a parallel-resonant circuit fed by a constant current, I, the amplitude of the rf voltage, E, across the resonating elements is given by:

$$E = I\,|Z_L|\,Q \qquad (3.1)$$

where Z_L is the impedance of the branch including the pickup coil. Q is defined in the usual way; it is equal to 2π times the ratio of the average energy stored in this branch to the energy dissipated per cycle. Since this branch includes a full-wavelength line of coaxial cable (from the pickup coil to the top of the cryostat) with certain losses, Q is rather low (~ 20). This is desirable in one sense, so that the circuit remains tuned when the frequency is swept slowly

over the full width ($\sim$100 kc) of the proton magnetic resonance. On the other hand, Q must be large enough to assure a good signal-to-noise ratio for the thermal equilibrium signal.

The Q-factor can be separated into a part corresponding to dissipation in the ohmic leads, Q_R, and a part corresponding to power absorbed or emitted, Q_C.

$$\frac{1}{Q} = \frac{1}{Q_R} + \frac{1}{Q_C} \tag{3.2}$$

Equation (3.1) can be multiplied by E, and rearranged to give

$$EI = \frac{1}{Q_R} \frac{E^2}{|Z_L|} + \frac{1}{Q_C} \frac{E^2}{|Z_L|} \tag{3.3}$$

The left-hand part represents the energy input to the circuit. E and I are considered to be in phase when the circuit is on tune. The two terms on the right exhibit the two types of energy absorption. Both terms are proportional to the square of the current circulating in the resonant loop. This circulating current is given by E/Z_L; Z_L is considered to be a constant.

The last term in (3.3) is proportional to the polarization of the target, P.

$$\frac{1}{Q_C} = \alpha P, \tag{3.4}$$

where α includes the various geometric factors that relate the circulating current to the amplitude of the oscillating magnetic field in the crystal, the filling factor, transition matrix elements, etc. It is a function of

$(\omega - \gamma H)$, where ω is the frequency, γ the gyromagnetic factor of the proton resonance, and H the applied external field. Solving for E

$$E = \frac{I|Z_L|}{\frac{1}{Q_R} + \alpha P} \qquad (3.5)$$

The magnitude of E can be observed directly. It is not possible to see the effect on E of P at thermal equilibrium, because of poor signal-to-noise ratio. More sensitivity is obtained in a standard manner by varying the magnetic field by a small fraction of the line width at 400 cps and using the lock-in detector to extract the corresponding variation in E while suppressing noise. The signal observed is proportional to the derivative of E with respect to H . E depends on H through the factor ,

$$\frac{dE}{d(\gamma H)} = \frac{I|Z_L|}{(1/Q_R + \alpha P)^2} \alpha' P \qquad (3.6)$$

where α' denotes the derivative of α with respect to its argument, $(\omega - \gamma H)$. Combining (3.5) and (3.6),

$$\frac{dE}{d(\gamma H)} = \frac{E^2}{I|Z_L|} \alpha' P \qquad (3.7)$$

I and Z_L may be regarded as constants. With a large crystal and high polarization, noticeable changes in E occur as one sweeps through resonance, as large as 30% in the case of the Berkeley target. The proper procedure is to monitor both E and $dE/d(\gamma H)$, and at each frequency divide the latter by E^2 to obtain the signal that is to be compared with the similar quantity at thermal equilibrium.

It has been observed that Q_R may change by about 10% due to temperature changes between the time of thermal equilibrium and that of high polarization. Equation (3.7) automatically corrects this effect also.

No correction has yet been made for the dispersive part of the nuclear magnetic resonance. This has the effect of making χ a complex number on either side of the center of the line. Equation (3.7) is still correct, but information about the phase of the rf signal is needed to extract the value of the polarization with complete accuracy.

4. Change in Line Shape

In the lanthanum magnesium nitrate crystal the protons occupy 8 inequivalent sites in the lattice (ZALKIN et al. 1961). The proton resonance signal is therefore a complicated superposition of many closely spaced single resonances. The relative position of each of these resonances is determined by local magnetic fields, which in turn depend in part upon the relative orientation of nearest-neighbor proton spins. The transition from near-zero polarization at thermal equilibrium to very high polarization brings about considerable rearrangement of these resonance lines. Consequently the shape of the compound "line" undergoes a drastic change. In order to make an accurate comparison between the thermal equilibrium and high polarization signal, it is not adequate to measure any single feature of the resonance, such as its peak amplitude. One must take the integrated area under the absorption curve, the curve of χP vs frequency, as the measure of the polarization.

In practice the frequency is swept slowly, with a linear sweep, through the resonance, and both E and its derivative are recorded on

a chart recorder. The function

$$\frac{1}{E_2} \quad \frac{dE}{d(\gamma H)}$$

is computed at each frequency. According to (3.7) this function is proportional to the derivative of the atsorption curve. This function is integrated once to yield the absorption curve itself. Then it is integrated again to yield the quantity that is compared with a similar one calculated at thermal equilibrium. This is a tedious procedure that must be performed many times during the experiment. Errors in estimating areas, truncation errors, etc., enter every calculation and propagate themselves strongly in such a double integration. At present methods are being sought to improve this step in the polarization measurement, which sets the present limit on the accuracy.

5. Measurement of Polarization by Shift in Local Fields

ABRAGAM, BORGHINI, and CHAPELLIER 1962 describe a method used during the experiment of ABRAGAM, et al. 1962 to measure the polarization. In this experiment the target crystal was so small that the thermal equilibrium signal could not be used. The method of measurement was based on the fact that a system of polarized proton spins distributed throughout a crystal constitutes a net magnetization of the sample and should give rise to measureable local magnetic fields. The measurement consists in observing the shift in position of the ESR resonance line due to the proton polarization. In principal, any nuclear resonance in the sample, such as that of nitrogen or lanthanum nuclei or of the proton line itself, might be used for this type of measurement. In the

small sample, of course, these lines were probably lost in noise, for the same reasons as for the thermal equilibrium proton signal.

The microwave power is reduced (so that the proton polarization is not destroyed prematurely) and the frequency tuned to the steepest-sloped position of the allowed ESR transition. The proton polarization is then destroyed by saturation with an intense rf field at the proton Larmor frequency. A shift is observed in the ESR signal. The external magnetic field is then changed to restore the initial condition of the ESR, and this field shift (about 1/2 oe.) is measured. The shift is proportional to the magnetization, which in turn is proportional to the proton polarization. The proportionality constants are calculable, so this represents an absolute measurement of the polarization.

Aside from the objection that the proton polarization must be destroyed in order to measure it, there are some difficulties with this method. For one thing, in a spherical sample of a crystal with cubic symmetry this effect vanishes completely. The sample used in the experiment described was in the shape of a thin slab, which gives the maximum sensitivity, but the results must always depend strongly on the exact geometry. The anisotropy of the crystal leads to another contribution which is difficult to assess accurately, but which may be a small (about 10%) fraction of the total in the flat-slab case. The change in the shape, if any, of the ESR line with proton polarization is another factor that had better be well investigated before this method can be applied.

The Saclay group has recognized these difficulties and resolves them by calibrating this method of measurement in a large sample of similar shape, using the size of the proton magnetic resonance signal as the calibrating measurement.

6. Nuclear Scattering to Measure Polarization

The use of some nuclear effect, that has been well measured by some other methods, as a calibration of the target polarization has some definite advantages. The problem of non uniform polarization of the target can be removed. Presumably the calibrating technique samples the different parts of the crystal with the same relative weighting as the beam used in the actaul measurement. The idea is straightforward, avoiding the problems of non linearity, line shape change, etc., associated with magnetic resonance measurements.

For example, the double-scattering parameter P can be measured in several ways. This parameter is usually considered to be equal to the average polarization of one of the final particles after scattering (at a given incident energy and scattering angle) of an unpolarized beam by an unpolarized target. This final-state polarization can be measured by a second scattering, by a target of known analyzing power. The same parameter may also be measured (if time-reversal invariance applies to the interaction) by observing the dependence of the rate of the same reaction upon the polarization of the particle before collision. Such a measurement may be made with a beam of known polarization incident on an unpolarized target. Or one may use an unpolarized beam with a polarized target. In either of the latter cases one measures the fractional change in rate when the polarization of the beam (or target) is reversed. This measured effect is equal to the product of the initial polarization and the parameter P. Thus, we can expect that measurement of such a parameter by two such methods (perhaps in the course of the same experiment) can be used to calibrate the target polarization with a minimum of systematic errors.

At high energies (above 150 MeV) there are few experiments in which the parameter P has been measured to greater precision than 10% of its own value, without using a polarized target. Until better measurements become available, this method of calibration does not yet compete for accuracy with magnetic resonance methods. At lower energies the P parameter for proton scattering from hydrogen and helium has been measured at many energies and angles, often with precision of a few per cent.

Another polarization-dependent effect that may be used as a calibration is the transmission of low-energy polarized neutrons through the target in question. The cross-section may depend strongly on the total angular momentum of neutron plus target. This is $(I + 1/2)$ when both beam and target are polarized in the same direction. When the beam polarization is reversed $(I \pm 1/2)$ states are present in known proportions. I is the spin of the target nucleus; orbital angualr momentum is zero at the energies considered. If there is a resonance in one of these states, whose cross section and other parameters are measured separately, the difference in transmission between the two beam polarization states can be related directly to the polarization of the target. As an example consider the total cross section of polarized low-energy neutrons on polarized protons. If the $J = 0$ cross section is σ_0, and the $J = 1$ cross section is σ_1, then when beam and target are polarized parallel to each other we have

$$\sigma\,(\text{parallel}) = \sigma_1$$

When beam and target are anti-parallel

$$\sigma\,(\text{anti-parallel}) = 1/2\ (\sigma_0 + \sigma_1).$$

If beam polarization is P_N, and target polarization is P_T, the difference in cross section when one of the polarizations is reversed is

$$\sigma(\text{difference}) = 1/2\ (\sigma_o - \sigma_1)\ P_N\ P_T.$$

The effect is large, since $1/2\ (\sigma_o - \sigma_1)$ 38 barns at low energies. Other elements present in the target do not contribute to the difference. The polarization is determined from the beam polarization, the geometry of the target, and the transmission counting rates before and after reversing polarization.

When the nucleus being polarized is radioactive, anisotropy of the decay can be used as a measure of polarization. It does not appear likely that such radioactive species can be gathered in sufficient concentration to be useful as a target. Since the nuclear polarization is usually dependent on nuclear moments, it does not appear that radioactive species can be used to monitor the polarization of stable isotopes in the same sample, either.

Even when use is made of nuclear methods of calibrating polarization, it is still necessary to have some monitor of fluctuations and changes in the target polarization. This may be provided by taking frequent calibrations, or by using one of the other methods as a secondary polarization measurement.

7. Spin-Echo Technique

The spin-echo method first devised by HAHN 1950 was used by CLARK and FEHER 1963 to measure polarizations in the semi-conductor InSb. This method overcomes some of the difficulties of the magnetic-resonance technique, and may prove to have great applicability.

In a sample containing polarized nuclei a short pulse rf power is applied at the Larmor frequency. The duration of the pulse is short enough so that the frequency is flat over the whole width of the resonance, making this method insensitive to line-shape changes. The amplitude of the pulse is carefully controlled to be always the same. The average spin direction of the polarized nuclei will precess by a certain angle (determined by the power and duration of the pulse) away from the direction of the magnetic field. The transverse component of the magnetization will then decay, giving rise to a free-induction decay signal in a pick up coil. The amplitude of this signal is proportional to the polarization of the nuclei. The thermal equilibrium signal provides the calibration.

The limitations on the use of this method have mainly to do with sensitivity. If one is willing to destroy the polarization, one may choose $\theta = 90^{\circ}$ for maximum sensitivity. However, in many cases this is not desired, and so a small angle must be used. The question then arises whether a thermal equilibrium signal sufficiently above noise level can be generated under these conditions. This can be answered only by trial in each case.

4. Further Discussion of Polarized Proton Targets

A. Other Materials Containing Hydrogen

1. Solid Hydrogen

The ground state of the hydrogen molecule is the $L = 0$, $S = 0$ parahydrogen state, in which the proton polarization is, of course, zero. The lowest excited state allowed by the Pauli exclusion principle is the $L = 1$, $S = 1$ orthohydrogen state. A solid that is initially 75% orthohydrogen can be prepared by quick-freezing natural molecular hydrogen.

Paramagnetic centers in the form of unpaired hydrogen atoms can be used as the starting points for the dynamic polarization process. These can be introduced into the sample by diffusing atomic hydrogen from an atomic beam apparatus into the material before freezing. Or one may create the centers within the solid by subjecting them to radiation damage. There is some evidence that permissible concentrations of atomic hydrogen may be limited to about 7×10^{16} atoms/cm^3 in the presence of molecular hydrogen (PIETTE el al 1959). At higher concentrations the atoms spontaneously recombine.

The number of protons that can be polarized by each paramagnetic center is given by the ratio of the spin-lattice relaxation time of the protons to the spin-relaxation time of the paramagnetic centers. In orthohydrogen the proton spin-relaxation time is short, as a consequence of the orbital motion, which leads to strong coupling with the lattice. Near 1° K, this time is of the order of 1 second (BLOOM 1957). The spin-relaxation time of the atomic hydrogen is about 0.1 second, according to ABRAGAM and BORGHINI 1964. When this ratio is compared with the small permissible concentration of paramagnetic centers, it appears that very

little polarization can be achieved by this method in solid hydrogen.

2. Solid Deuterium and Hydrogen Deuteride

The ground states of HD and D_2 have L = 0. As a consequence, the nuclear spin relaxation times are expected to be longer than in solid H_2, so there is some hope that sizable polarizations can be achieved in these molecules. Bose statistics permit S = 0 and S = 2 for the spin state of the deuterons in D_2. If the populations are divided according to the statistical weights, 5/6 of the deuterium will be in the S = 2 state and therefore polarizable. In the case of hydrogen deuteride, dissociation of the HD according to the reaction:

$$2HD \rightleftharpoons H_2 + D_2$$

If statistical weights govern the distribution, one expects 6/7 of the hydrogen (and deuterium) atoms to be found in HD molecules.

REBKA and WAYNE 1962 and SHARNOFF, SANDERSON, and POUND 1962 have reported some success in the dynamic polarization of solid deuterium. No attempt has yet been made to polarize solid HD.

Paramagnetic centers are unpaired atoms of deuterium. One method of creating them was to include a small amount of tritium in the sample. When the tritium undergoes radioactive decay, radiation damage results in broken molecular bonds near the site of the decay. SHARNOFF and POUND 1963 found that the number of centers created was an order of magnitude greater than by other methods. Nevertheless, this method did not prove as successful in producing nuclear polarization as one employed by REBKA 1963 and private communication. Samples were grown by passing gaseous

deuterium through an electrodeless, rf discharge in a "dry-filmed", glass tube and condensing it into a microwave cavity. The magnetic field used was 8500 oersteds. The maximum microwave power used was estimated at about 200 milliwatts delivered to the cavity (Q 3000 - 5000). Because of the hyperfine interaction in the atoms, the ESR line has a triplet structure. Polarization in the neighborhood of the central line leads to higher enhancements than in the neighborhood of the lateral lines. In fact, there is evidence that, under the conditions stated, the lateral lines are being saturated at 1.2°K. An enhancement of 160 is obtained at 4.2°K, equivalent to 0.7% polarization. At 1.2°K an enhancement of 80, equivalent to 1.2% polarization has been achieved. The density of free deuterium atoms was 5×10^{16} per cubic centimeter; there is hope to increase this by almost two orders of magnitude with improved sample-growing technique.

The method looks very promising, but much development remains to be done.

3. Polyethylene

Perhaps more effort has been devoted to trying to obtain large proton polarizations in polyethylene, $(CH_2)_n$, than in any other substance to which dynamic methods have been applied. The attraction to polyethylene and other plastics is that they are solids at room temperature (therefore, easy to handle, shape, and store), have relatively high hydrogen fractions, and have actually higher absolute concentrations of hydrogen per cm^3 than even pure solid H_2. Paramagnetic centers have been obtained either by dissolving free radicals into the plastic or by radiation bombardment. The results have often been disappointing, and difficult to reproduce.

BORGHINI and ABRAGAM 1960 reported 4.5% proton polarization in polystyrene containing 10% by weight of the free radical di-phenyl picryl hydrazil DPPH. Magnetic field was 12 kOe, ESR frequency 33.5 GH_z, temperature 1.5°K. HWANG and SANDERS 1960 reported 1.2% polarization in high-density polyethylene irradiated by fast neutrons. Magnetic field was 3.2 kOe, microwave frequency 9 GH_z, and temperature 1.2°K. NEGANOV et. al. 1963 report about 1.4% under similar conditions but with T = 0.5° K. JEFFERIES 1963 summarizes the published work to date on irradiated and doped plastics. Hwang and Sanders report (private communication) that at 35 GH_z and 12.5 kOe they repeatedly observed proton polarization of 6%. Hwang calculates -- assuming Lorentzian line shape with 150 Oe linewidth for the forbidden transition--that one should achieve 20% polarization under these conditions, and 51% if frequency and field are doubled.

Some of the difficulties involved in using polyethylene must be mentioned. Samples must be stored at low temperatures (dry ice or liquid nitrogen) to keep paramagnetic centers from recombining. Polyethylene is a very poor heat conductor, so any large target will have to be made out of several thin pieces to insure proper cooling. The process of preparation of samples is difficult to control. Many more pieces must be prepared than are to be used, and selected on the basis of performance.

It may well be that, despite these difficulties, a high polarization can be achieved in a polyethylene target. Such a target would have considerable advantage over existing ones in some experiments.

4. Other Hydrogeneous Materials.

To the author's knowledge, there has been little or no experimental work done with proton polarization in compounds of hydrogen with elements

of the first shell in the periodic table. These compounds have a high percentage of hydrogen by weight and should therefore be considered on that ground alone. Paramagnetic centers are provided, as above, either by doping with free radicals, or by radiation damage and the creation of F centers.

Lithium hydride (14% hydrogen) has been suggested by ABRAGAM and BORGHINI 1964 . It has the advantage of being solid at room temperature. Frozen methane (CH_4 - 25% hydrogen) is second only to HD (33%) in hydrogen fraction, and does not have the complication of having other polarized nuclei beside the protons. Frozen ammonia (18% hydrogen) and ice (11%) form very anisotropic crystals, and large single crystals may be difficult to grow. Hydrogen fluoride (5%) has slightly higher hydrogen fraction than the lanthanum magnesium mitrate crystals (3%). One may also note that lanthanum ethylsulfate crystals -- $La(C_2H_5SO_4)_3 \cdot 9H_2O$ -- with paramagnetic impurities, may be useful as a polarized target material. This compound is 5% hydrogen by weight. Large single crystals are more difficult to grow than in double nitrate.

B. Separation of Hydrogen Events from Background

It does not appear likely that solid hydrogen will be a polarizable target. The free protons in any other material constitute a minority of all the nucleons present in the target. Therefore, it will be necessary in any experiment employing these targets to identify those events which are due to scattering by free protons.

The polarization itself can be used as a means of distinguishing free proton scattering. If two successive counting rates are measured under identical circumstances except that the direction of the polarization vector has been reversed between counts, the difference between the two

rates can only be due to the effect of the polarized nuclei. Unless other nuclei in the target are also polarized, the difference is entirely due to scattering by free protons. Polarization in undesired nuclei may be suppressed, if necessary, by saturating the sample with rf power at the appropriate Larmor frequency. In many experiments the difference in counting rate upon polarization reversal is a small fraction of the total rate. Consequently, there may be a large fractional uncertainty in this difference, to counting statistics. In some cases this can be the limiting factor in the experimental accuracy.

It is important to know what fraction of the average total counting is attributable to free protons. Often the total rate must be used as a normalization to the polarization-dependent effect. The non-hydrogen total can be estimated by substituting a dummy for the polarized target, duplicating the geometry and chemical makeup of the latter, but being free of hydrogen insofar as possible. One may also estimate the non-hydrogen background by measuring counting rates that, by their kinematic properties, cannot be due to scattering from free protons, and extrapolating these rates to their values under the free proton peak.

It is of course desirable to have the ratio of hydrogen events to background as high as possible. This makes it easier to determine the total free-proton event rate. Moreover, when this ratio is much less than unity, the difference in rate due to polarization reversal is made that much less than unity, difficult to measure. In principle one can measure a small effect as accurately as desired by taking sufficiently long counting time. In practice it is not possible to measure effects very much smaller than one percent, because of the difficulty of maintaining constant conditions for the long counting time required.

A choice must sometimes be made between two target materials, one of which has a greater hydrogen fraction, and the other is capable of yielding higher polarizations. Let H be the counting rate due to free protons, and B the background due to heavier nucleus. One may presume that the ratio H/B is, other factors remaining the same, proportional to the ratio of free protons to bound nucleons in the target material. The fractional change in rate upon reversal of polarization, P, will be proportional to PH/(H + B). This factor may be taken as a figure of merit in deciding upon target material. For example, suppose H/B is 1/5 in some experiment when lanthanum magnesium nitrate is chosen as the target material, in which P = 0.60. The figure of merit is then 0.10. For the same experiment with a polyethylene target, one may expect H/B $\simeq$ 1, and one achieves the same figure of merit if P $\simeq$ 0.20. However, if H/B is much above unity for both materials, one would choose the target material with the higher polarization. In special cases the target material may be selected for such features as high density, or absence of elements with high atomic number. The choice of target material is thus seen to depend strongly on the nature of the experiment to be performed.

The background can be suppressed considerably by making use of two-body kinematics. When an incident particle of known energy strikes a free proton at rest, and there are only two particles in the final state, the final energies and angles are uniquely correlated. The energies and angles of both final-state particles are completely determined by the measurement of any one of them. No such exact correlation exists when the beam particle collides with a bound nucleon. Two-body final states include all cases of elastic scattering, and also many inelastic reactions, such as

$$t + p \rightarrow d + d$$

$$+ \quad + \quad +$$
$$+P \rightarrow \Sigma^{+} + K^{+} .$$

In the following sub-s ections we discuss the merits of various ways to use two-body kinematics to suppress background.

1) The Direction Of Both Final Particles

In experiments performed with the Berkeley polarized target SCHULTZ, et al. 1963, STEINER et al. 1964), arrays of small scintillation counters were placed on either side of the beam, in the median plane of the magnet. Coincidence counting rates were recorded between each pair of counters on opposite sides of the beam. Most such coincidence rates gave a low level corresponding to background events. When a pair of counters was so situated that their two angles were those of the products of an elestic scattering from a free proton, the coincidence rate was higher. This indicated that both free proton and background evernts were being counted. It was found that in π^{-}-p scattering at 250 MeV ,H/B was about 1.5; in proton-proton scattering at 2 to 6 GeV, H/B was between 5 and 10. An one-inch-cube sample of lanthanum magnesium nitrate crystal was used in these experiments. These hydrogen-to-background ratios were very gratifying, if unexpected. One may conclude that in these experiments at least, little is to be gained by using materials with greater hydrogen fraction.

There is reason to expect that the background suppression will continue to be as good as this when the experiments are extended to higher energies. At very high energies the kinematics are such that the elastic events are compressed into a narrow cone in the forward direction. Adequate geometrical angular resolution can in principle always be

obtained by placing small counters far enough downstream from the target. The number of such counters needed to obtain the same solid angle will increase; spark chambers might be employed to relieve some of this problem. The ultimate limit on the angular resolution is determined by (a) angular divergence of the incident beam, or (b) multiple scattering in the crystal.

The mean angle of multiple Coulomb scattering is proportional ~~ity~~ to the square-root of the thickness of the crystal. At high energies it is inversely proportional to the momentum (actually it goes as $1/p\beta$). This means that the product of beam momentum and the mean scattering angle, i.e., the effective transverse momentum imparted by Coulomb scattering, is independent of energy. For a 1 inch-cube sample of lanthanum magnesium nitrate this mean transverse momentum is about 10 MeV/c. On the other hand, scattering from bound nucleons is characterized by transverse momentum of about 200 MeV/c, the Fermi momentum of particles bound in a finite nucleus. The suppression of the background is given by the square of the ratio of these characteristic transverse momenta, a factor of 400. This is so because the background events may be thought of as smeared out, in each of two transverse dimensions, 20 times as broadly, from the ideal two-body kinematic angles, as the free-proton events. The results are independent of energy so long as the angle subtended by each counter is smaller than the mean multiple-scattering angle.

This method is limited at low energies and small scattering angles by the failure of low-energy recoil protons to reach the counters. With the 1 inch lanthanum magnesium nitrate crystal, protons of less than 60 MeV could not be counted. Low-energy protons either stopped within the

target, or emerged with such low momentum that they were trapped in the magnetic field. The more energetic protons below this limit escaped, but had a varying loss of energy that depended on path traveled within the crystal. The consequent varying amount of curvature in the field served to destroy all information about the initial direction of these slower protons. When it is important to observe low-energy recoils, the crystal must be made thinner, or tilted with respect to beam direction, or both.

2) The Energy of Both Final Particles

ABRAGAM, et al. 1962 , in a proton-proton scattering at 20 MeV, detected both final recoil particles with two large angle CsI crystals located at 45° on either side of the beam. Two coincident particles were counted only if the energies of each of them was above 1.5 MeV and if their sum was above 10 MeV. No quantitive estimate of the background rate is given, but the intimation is that it was small compared with the free-proton rate. This method of background suppression is particularly effective when the total energy available is comparable to the binding energy of a proton in a bound nucleus.

It would seem that solid-state detectors, which provide both energy and spatial resolution might find considerable application with this regard. If both particles are detected the reaction can be two times overdetermined with consequent almost total suppression of background.

3) The Energy and Angle of One Final Particle

When it is impossible to detect one of the final particles, recourse may be had to this less effective method of background suppression.

The occasion may arise, for example, when one of the final particles is an unstable hyperon that decays within the crystal, sending decay products mostly into the pole faces.

In the -p experiment at 250 MeV with the Berkeley target, a range telescope was set up to select those forward-scattered pions which stopped at the appropriate moderator thickness. The "appropriate" thickness is the range of free-proton scattered pions emerging at that particular angle. The free-proton events could be clearly distinguished, but the background rate was four or five times as great. The method suffers in its background suppressing power from the fact that only one parameter (the energy) is being used as kinematic criterion. When two particles are detected in coincidence, the requirement of coplanarity with the incident beam is applied automatically.

Range telescopes are not convenient at energies very much above 250 MeV. Here one may use magnetic analysis of the outgoing particle to determine its momentum. Since the target is located with a strong honogeneous field, it seems practical to use this same field in the momentum analysis.

C. Size of the Target

Experimental considerations often dictate the limits to the size of the polarized target. Its thickness must be such that the lowest energy particle one hopes to detect will lose only a small part of its energy in traversing it. The dimensions of the target transverse to the beam direction need be no larger than the size of the beam spot to be used. When angular resolution is important, the thickness is limited by the need to keep multiple Coulomb scattering down.

When none of these factors is limiting, the general desire is to make the target as large as possible. This will result in higher counting rates, and so make most efficient use of accelerator time. The present limits on the physical size of the target are set by the effort and expense one is willing to devote to construction of necessary apparatus. Sufficient microwave power is needed to saturate the forbidden transition in the large crystal. Carcinotrons that produce 15 watts at 70 GH_z seem to meet all conceivable needs on this point. Cooling capacity is needed to handle this amount of power; this seems to be a matter of straightforward cryogenic engineering. A magnetic field is needed that is homogeneous to one oersted out of 20,000, over the whole target volume; this problem is surmountable with careful design and enough material. There is no practical reason why larger polarized targets than existing ones will not be built. The limitations are set only by considerations of how worthy is the effort to build them.

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APPENDIX A

Time Reversal and Polarized Target Experiments

In this section we consider the relation between single elastic scattering involving a polarized target and the more conventional experiment in which the polarization of the recoil particle is measured after a scattering from an unpolarized target. For simplicity we will assume that the projectile particle is either spinless, or that it is unpolarized before scattering, and not analyzed for polarization afterward. Let the projectile have momentum p_i before scattering, and p_f after scattering. Choose the z-axis in the direction $(p_i \times p_f)$, normal to the plane of the reaction. We shoose the wave-functions to be plane spinor waves, before and after the reactions, and eigenfunctions of I_z, where I is the spin angular momentum, with eigenvalues m_i (before) and m_f (after). Let $p(m_i)$ be the relative population of the various m_i states before the reaction; for a completely unpolarized target $p(m_i)$ $1/(2I + 1)$. Finally, let the differential cross-section for scattering from the state m_i into the state m_f be $R(m_i m_f)$. The polarization, P, induced in the recoil particle in scattering from an unpolarized target is given by

$$P\frac{m_f}{I} = \frac{\sum_{m_i, m_f} \frac{m_f}{I} \; R(m_i \rightarrow m_f)}{\sum_{m_i, m_f} R(m_i \rightarrow m_f)} \qquad \text{(A.1)}$$

Now consider the left-right asymmetry, P', in scattering from a polarized target; or equivalently, the fractional dependence of the

left-scattering on complete reversal of the target polarization. The transition from left to right-scattering is effected by a 180° rotation about the incoming beam direction, thus reversing the z-direction and taking $p(m_i)$ into $p(-m_i)$. It is not difficult to see that

$$P' = \frac{1}{P_T}\,\frac{L - R}{(L + R)} = \frac{\sum_{m_i m_f} \left[p(m_i) - p(-m_i) \right] R(m_i \to m_f)}{P_T \sum_{m_i m_f} \left[p(m_i) + p(-m_i) \right] R(m_i \to m_f)} \qquad (A.2)$$

where

$$P_T = \sum_{m_i} \frac{m_i}{I}\, p(m_i) \qquad (A.3)$$

is the initial target polarization.

The effect of various symmetry principles can now be stated. Time reversal invariance requires, for an elastic scattering, that

$$R(m \to m') \quad R(m' \to m). \qquad (A.4)$$

Space-reflection invariance ("parity" or "R-invariance," see BOHR 1959), says that

$$R(m \to m') \quad 0 \text{ if } m'-m \text{ is odd.} \qquad (A.5)$$

If target and projectile are identical particles, the positive z-direction sense is undefined, and we must have

$$R(m \to m') \quad R(-m \to -m') \qquad (A.6)$$

If I 1/2, anyone of the conditions (A.4), (A.5), or (A.6) is sufficient to guarantee that P P'. This is not a trivial statement. In an inelastic process, such as associated production of strange particles, it is possible that none of these conditions are valid. BILENKY 1958 suggested just such an experiment to test the

relative parities of strange particles.

When $I = 1/2$, it is obvious that a given P_T can be obtained with many different distributions of $p(m_i)$, leading to different values of P'. A direct relation between P and P' can be obtained if we specify that p is a linear function of m_i:

$$p(m_i) = \frac{1}{2I + 1}\left(1 + a\frac{m_i}{I}\right) \qquad (A.7)$$

with $|a| \leq 1$. With this form of distribution (obtainable, for example, with brute force methods when $\mu H/IkT \ll 1$) one obtains, using (A.4) to interchange m_i with m_f in (A.1).

$$P = \frac{P_T}{a} P' \qquad (A.8)$$

From (A.3) and A.7)

$$\frac{P_T}{a} = \frac{\sum_{m_i=-I}^{I} m_i^2}{I^2(2I + 1)} \qquad (A.9)$$

For $I = 1/2, 1, 3/2, 2, 5/2, 3,$ and $7/2$,

$\frac{P_T}{a} = 1,\ 2/3,\ 5/9,\ 1/2,\ 7/15,\ 4/9,$ and $3/14$, respectively.

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FIGURE CAPTIONS

Figure 1. Energy level diagram for system of a paramagnetic center weakly coupled to a neighboring nucleus, in an external magnetic field. The frequencies quoted are those appropriate for producing dynamic proton polarization in a lanthanum magnesium nitrate crystal, with neodymium doping, in a field of 9.1 kOe.

Figure 2. Lanthanum magnesium nitrate crystals used in Berkeley polarized target. Scale is in inches.

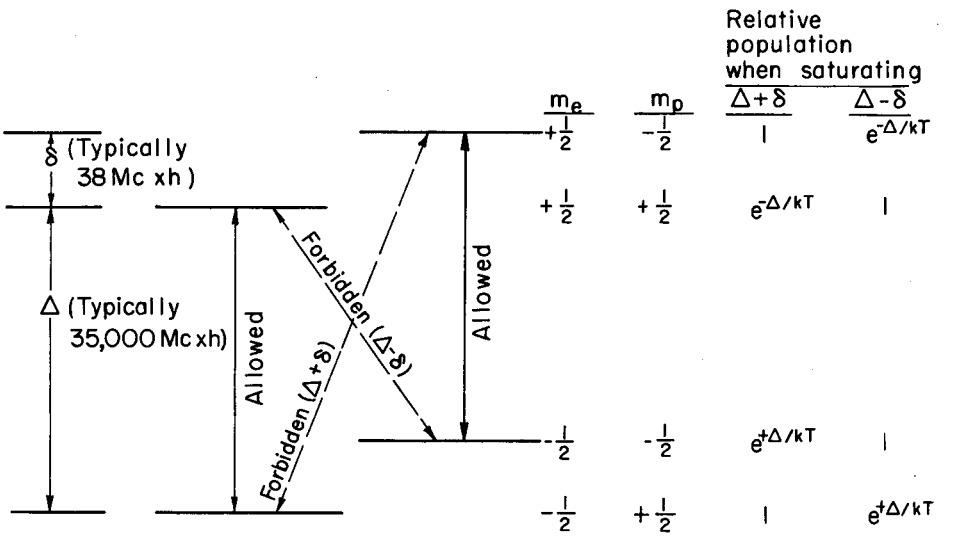
Figure 3. Cryostat used to maintain Berkeley polarized target at 1.2° K.

Figure 4. Microwave system used with Berkeley polarized target.

Figure 5. Mounting of Berkeley polarized target with respect to magnet. The particle beam is usually incident from the left.

Figure 6. Nuclear magnetic resonance system used to measure target polarization.

Figure 7. Microwave cavity used to contain Berkeley polarized target. Shown in cutaway view is the winding of the nuclear induction pickup coils, and the copper septum used to provide uniform detection sensitivity. The crystals (not shown) are mounted parallel to the septum, two on either side, filling the space between the pickup coil windings.



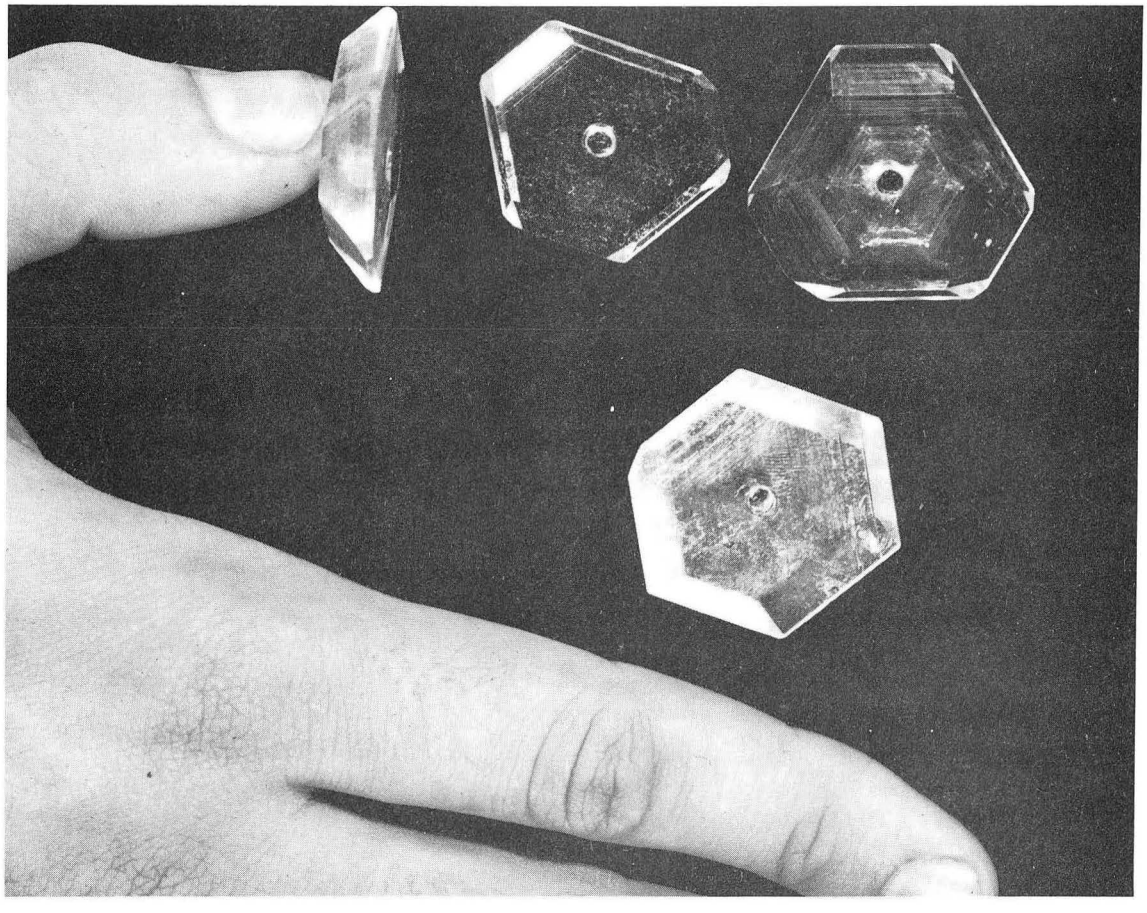
Proton polarization (saturating $\Delta+\delta$) $\longrightarrow \frac{\Sigma (m_p)_i P_i}{S \Sigma P_i} = \frac{(-\frac{1}{2})(1) + (+\frac{1}{2})(e^{-\Delta/kT}) + (-\frac{1}{2})(e^{+\Delta/kT}) + (+\frac{1}{2})(1)}{\frac{1}{2}(1 + e^{-\Delta/kT} + e^{+\Delta/kT} + 1)}$

$= -\tanh \frac{\Delta}{2kT}$

Proton polarization (saturating $\Delta-\delta$) $\longrightarrow +\tanh \frac{\Delta}{2kT}$

MU-28789

Fig. 1



ZN-4154

Fig. 2

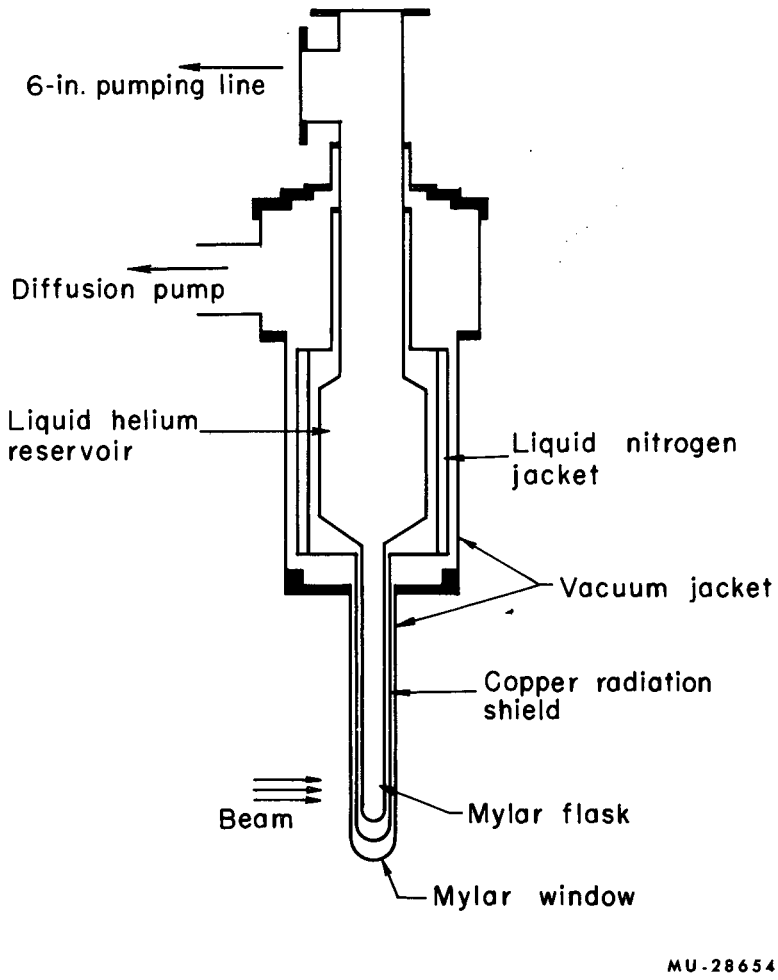

6-in. pumping line
Diffusion pump
Liquid helium reservoir
Liquid nitrogen jacket
Vacuum jacket
Copper radiation shield
Beam
Mylar flask
Mylar window
MU-28654

Fig. 3

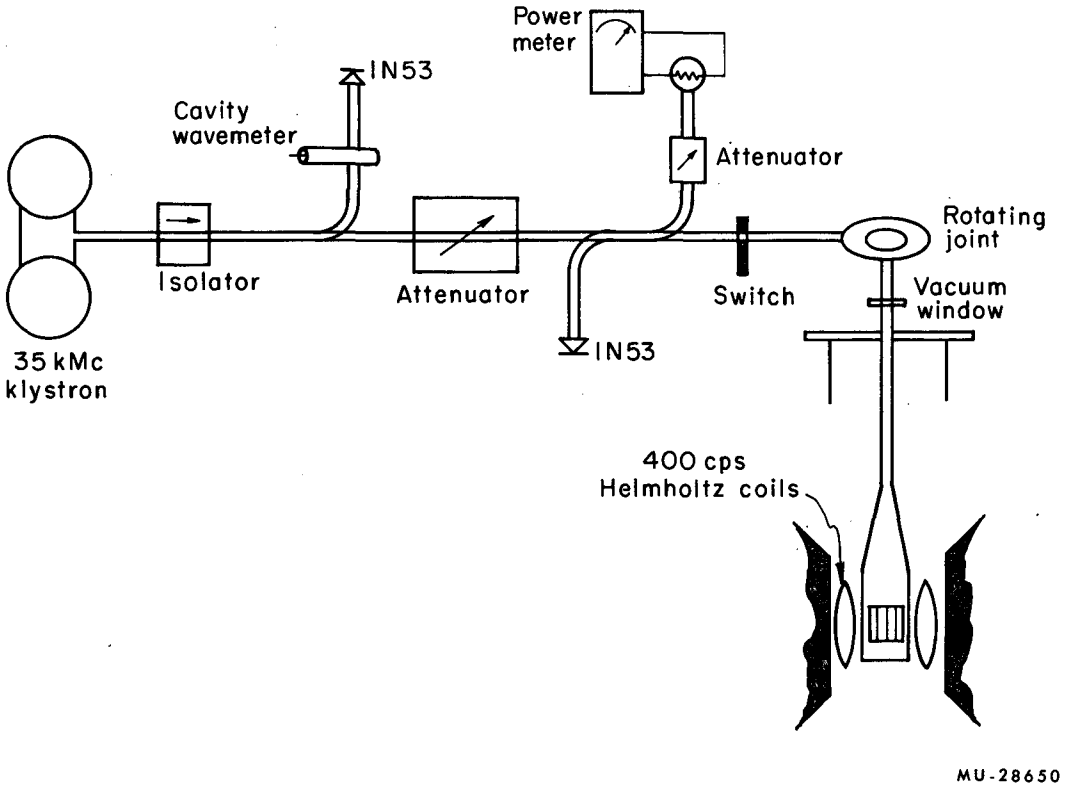
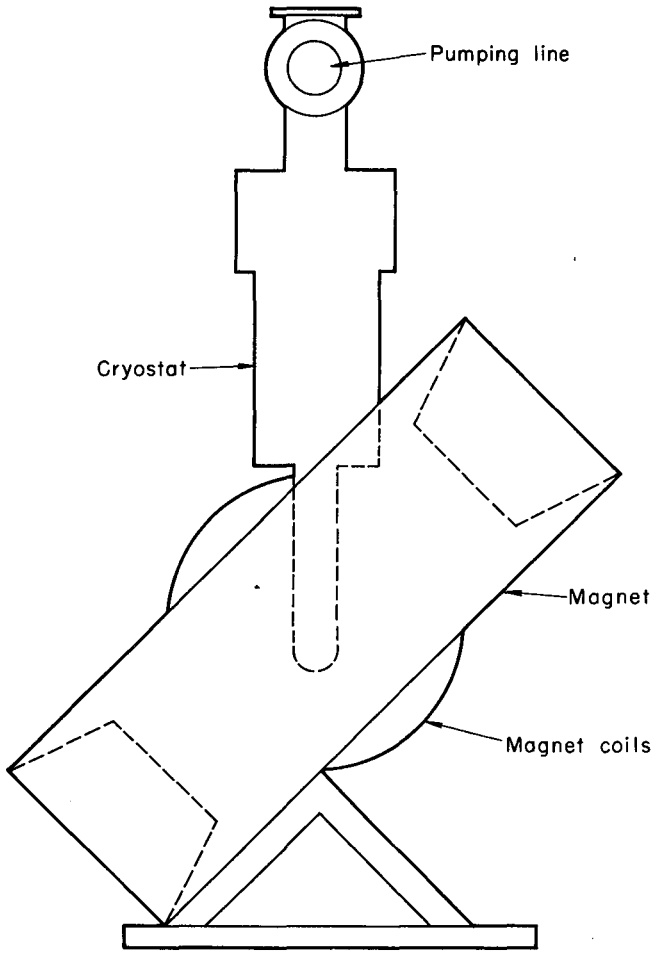

Power
meter
IN53
Cavity
wavemeter
Attenuator
Rotating
joint
Isolator
Attenuator
Switch
Vacuum
window
IN53
35 kMc
klystron
400 cps
Helmholtz coils
MU-28650

Fig. 4



MU-28652

Fig. 5

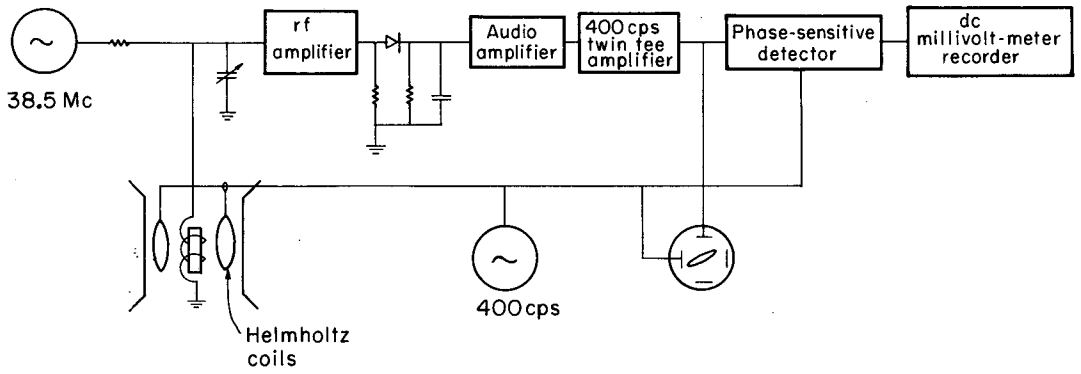

38.5 Mc
rf amplifier
Audio amplifier
400 cps twin tee amplifier
Phase-sensitive detector
dc millivolt-meter recorder
400 cps
Helmholtz coils
MU-28651

Fig. 6

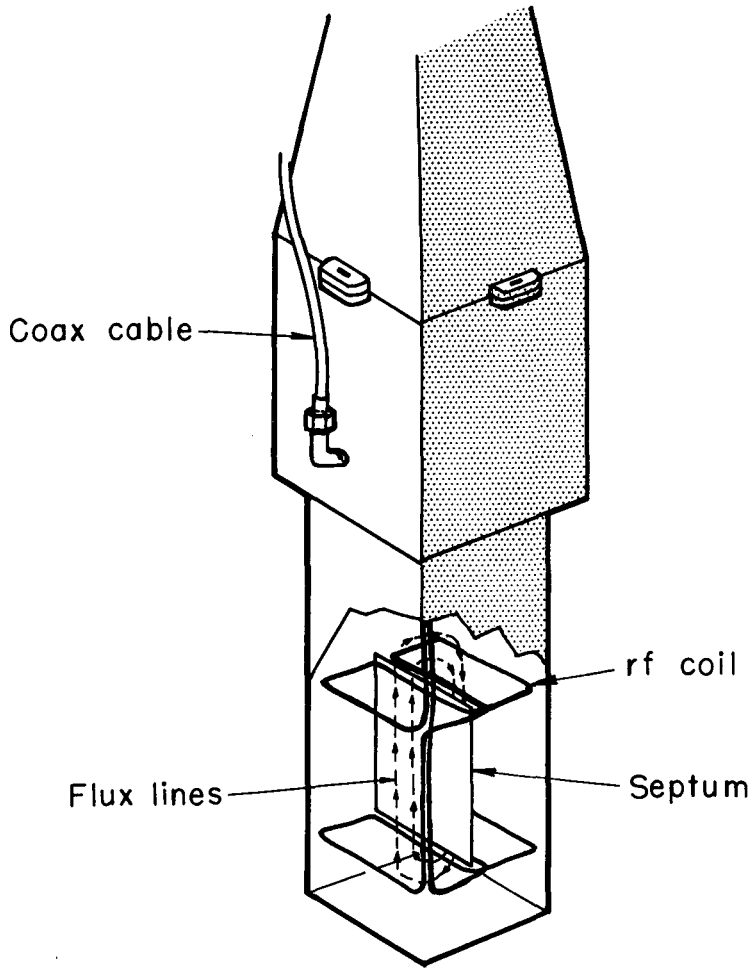


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Fig. 7