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# Experiments on Dynamic Polarization of Protons in Polyethylene by the 'Solid Effect' — A Proposed Polarized Proton Target[1])

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## Introduction

We wish to report here on the «state of the art» with regard to the polarization of nuclei, in particular of protons, in environments suitable for use as polarized targets for scattering experiments. The program of this talk will be to discuss:

1. design criteria for a polarized proton target;
2. possible methods of polarization;
3. recent experimental work in various laboratories which encourages one to believe that a useful polarized target can be realized;
4. the studies made at the University of Minnesota on this subject.

We wish to emphasize that our experimental work is still in a preliminary state. Although the prospect of a useful polarized proton target is encouraging, we have not a this moment such a target in final form.

## Design Criteria for a Polarized Proton Target

Many nuclear physics experiments utilizing oriented nuclei, a typical example being that by C. S. WU *et al.* [1][3]), have already been performed. Elegant angular correlation and other experiments involving radioactive decay of oriented nuclei have yielded many results. However, it is clear after some reflection that most of the orientation methods which have been applied in these radioactive decay experiments are not

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[2]) ALFRED P. SLOAN Foundation Research Fellow.

[3]) Numbers in brackets refer to References, page 133.

suitable for producing a polarized target for scattering experiments. We will state now the criteria, or perhaps the prejudices, we have applied in our thinking about possible polarized targets. These criteria are as follows:

A. The number of target nuclei must be large enough and the polarization must be sufficiently great to produce detectable assymmetries in scattered beams. For this reason we have restricted our consideration to condensed matter, and we have felt that only polarizations above ten percent would be useful. This separation at ten percent polarization is of course rather arbitrary and the position of the demarkation line actually depends on the particular experiment that is contemplated. In considering $p$–$p$ scattering at 40 and 68 Mev – the available energies of the Minnesota proton linear accelerator – we have assumed that the polarization of the incident beam might be of the order of thirty percent. Since assymmetries below one percent are for all practical purposes unobservable, we felt that a minimum target polarization of ten percent would be necessary.

B. The polarization must be maintained in the presence of the incident beam. This is the consideration which makes the decay experiments so different from scattering experiments. A useful figure to keep in mind is that if each particle loses 1 Mev in the target, a beam current of one microampere will deliver one watt to the target. This means that with this energy loss per particle a beam of $10^{10}$ particles per second dissipates approximately one milliwatt in the target. This need to handle power inputs in the range of a few milliwatts and to maintain a constant polarization over the long time intervals required to obtain counting statistics eliminates from consideration all orientation methods requiring such very low temperatures as can only be achieved by the method of adiabatic demagnetization.

C. The material containing the target nucleus to be studied (e.g. protons) should be simple enough so that the scattering process to be studied (e.g. $p$–$p$ scattering) can be separated from background scattering by other nuclei. In the experiments involving radioactive decay the radioactive nuclei may be embedded in a medium containing several different nuclear species without any complications ensuing. In the case of scattering experiments, however, each scattering process by nuclei other than those under study provides an undesirable background. If in addition, other nuclei with spins (which may or may not be oriented with the target nuclei) are present, spin correlation experiments become even more difficult. Of course energy discrimination will be helpful to distinguish beam particles scattered by protons from those scattered by other nuclei in the case of $p$–$p$ or $n$–$p$ experiments. Nevertheless even energy discrimination may not be adequate if foreign nuclei

possessing many low level excited states are present. Therefore we decided at the outset of our work that we would look into the question whether useful polarization can be produced in very simple materials. We have not explored the question whether there may be other less simple materials in which larger polarizations are realizable. (Work at other laboratories shows that larger polarization can indeed by achieved in various materials.)

D. It should be possible to fabricate the material readily into a suitable form for use as a target. This is not really a very serious criterion, involving as it does only questions of engineering design. However, on this basis we have excluded from consideration some otherwise fairly promising techniques. We have not considered for example, a target composed of hydrogen atoms dissociated in a discharge tube and deposited in a solid rare gas matrix at liquid helium temperatures.

## Possible Methods of Polarization

We divide the possible polarization methods into two categories, static and dynamic [2]. In the static method, polarization is produced in a material in thermal equilibrium subject only to time-independent external magnetic fields. In the dynamic methods, time-dependent external magnetic fields are used.

### A. *The static methods applicable to nucleus with spin* $I = 1/2$

1. Polarization in an external field (the «brute force» method). This method requires $H_0 = 10^5$ gauss and $T = 0.02^\circ$ $K$ in order to produce polarization of approximately 50%. Thus this method is out of the question since adiabatic demagnetization is required to reach such low temperatures.

2. Polarization in an internal field («Rose-Görter method»). This method is not completely out of the question if one could find a material with $H_{int} \cong 10^6$ gauss. However for an election in the 1 $s$ state about a proton, the internal field is $H_{int} \cong 1.5 \times 10^5$ gauss. Thus $T = 0.03^\circ$ $K$ is necessary to achieve a polarization of approximately fifty percent. The internal fields in ferromagnetic media are also typically of the same order of magnitude. Hence it seems unlikely that large proton polarization can be produced by static methods without the use of adiabatic demagnetization. As we have seen, this means that it is not likely that a useful polarized proton target can be achieved by this method. The situation will be more favorable for polarization of nuclei of high $Z$ value where the internal field at the nucleus can be considerably higher.

## B. *Dynamic Methods of Polarization*

Dynamic polarization of nuclei has been produced by many methods, some of which have been applied to radioactive decay experiments. Among the names of authors associated with the different methods are OVERHAUSER [3], ABRAGAM [4], JEFFRIES [5], FEHER [6] and others. Some of the methods have steady-state characteristics, producing polarization which can be maintained at a constant level for long periods. Others, notably two methods associated with the names of ABRAGAM and FEHER, are transient methods producing a polarization which decays with time. The time constant of decay is equal to the nuclear spin-lattice relaxation time which may be many minutes at low temperatures. We will not consider the transient methods here.

A common characteristic of the steady-state methods is, that the material containing the nuclei to be polarized must also be one which contains unpaired electron spins. The system is perturbed by time-dependent (radio-frequency or microwave) magnetic fields which induce transitions. The transitions of the Overhauser Effect involve the flip of the electron spin only. On the other hand, the transitions of the Jeffries method involve the simultaneous flip of both the electron and the nuclear spins. These transitions are possible due to the magnetic interaction between the nuclei and the electrons. In either case the system is allowed to come to a perturbed equilibrium state in the presence of the *rf* field and the thermal relaxation processes. Under favorable conditions the population distribution over the nuclear energy levels is no longer a Boltzmann distribution with characteristic energy $\hbar\omega_p$ but now involves an energy of the order $\hbar\omega_e$ where $\nu_e = \omega_e/2\pi$ and $\nu_p = \omega_p/2\pi$ are the Larmor frequencies of the electron and proton respectively in a field $H_o$.

Thus the nuclear polarization in the perturbed equilibrium state can be of the order of the electron polarization in thermal equilibrium, and a sizeable polarization can exist at a temperature obtainable without adiabatic demagnetization.

The original proposal by OVERHAUSER was for nuclear polarization in metals. Shortly after his work however, BLOCH [7], KITTEL [8], ABRAGAM [9] and others showed that the method has wider applicability. Since then, the Overhauser method has been applied to align nuclei in metals [10], free radicals [11], solvent nuclei in solutions of free radicals [12] and other materials. The proton-bearing materials to which this method might be applied appear generally to fail our criteria A (large polarization) and/or C (simple enough material).

The Jeffries method was originally applied by him to polarization of nuclei in paramagnetic ions, and as so applied also fails our criteria A

and C, for the paramagnetic ions must themselves be present in rather small concentration in a matrix of diamagnetic material.

C. *Polarization by the Solid Effect*

The method which appears to give hope that a polarized proton target can be realized was discovered in 1958 when ABRAGAM and PROCTOR [13] and ERB, MOTCHANE and UEBERSFELD [14] independently announced the observation of dynamic polarization of nuclei in various solids containing unpaired electron spins [15]. The observations indicated that polarization occurred not just in the vicinity of a paramagnetic center, but could spread throughout a sample of bulk material. The explanation of this effect, given by ABRAGAM and PROCTOR, indicated that such spreading of polarization could be the case and that this method of polarization should be a very general method of polarizing nuclei. The method, named «l'Effet Solide» or the «Solid Effect» by ABRAGAM, is like that of JEFFRIES in that it employs a microwave magnetic field which induces transitions in which an electron spin and a nuclear spin are simultaneously flipped. A nuclear polarization results when a perturbed equilibrium state is achieved in the presence of the relaxation process. Unlike the Jeffries method, however, the nuclear spins flipped by the microwave field can be rather distant from the paramagnetic center, and communicate with it only via the magnetic dipolar interaction between electron and nuclear spins. The internal magnetic field at such a nucleus is very nearly the external magnetic field. Thus these nuclei, after being polarized, can transmit the polarization to other nuclei far away from the paramagnetic center by inducing flips of the more distant nuclei due to the dipolar interaction among the nuclei themselves. The polarization thus spreads away from the paramagnetic centers by a «spin-diffusion» process. We may see in detail the process by which a nucleus reasonably near the paramagnetic center becomes polarized, as is shown in figure 1. We have drawn the figure for the relevant case where the nuclear Zeeman energy $\hbar\omega_p$ is much greater than the interaction energy $E_i$ between the electron and the nuclear moments. The allowed transition occurs at circular frequency $\omega_e$. However, because of the electron – nucleus dipolar interaction $m_I$ is not a rigorously good quantum number and the forbidden transitions A and B, at frequencies $\omega_{\substack{B \\ A}} = \omega_e \pm \omega_p$ can be induced, with a matrix element lower than that for the «allowed transitions» by a factor of order $E_i/\hbar\omega_p$. We suppose that the applied microwave field is sufficiently strong so that the «forbidden transition» is saturated, i.e. the population of the initial and final states of the transition are equalized. This will always be the case for a sufficiently strong microwave field because the trans-

ition probability of induced emission always equals that for absorption (Microwave power of the order of one milliwatt is sufficient for our case at 1.2 °K). The presence of this microwave field now «short-circuits» some of the relaxation processes. The fastest of these processes is generally that which keeps the electron spins in thermal equilibrium with the lattice at temperature $T$. The transition probabilities associated with these thermal relaxation process must have such values as to maintain the ratio of the populations in the upper and the lower states of the «allowed transitions» at the electron Boltzmann factor $\epsilon_e = \exp(-\hbar\omega_e/kT)$. Thus the result of saturating a «forbidden transition» will be to enhance greatly the nuclear polarization, since the distribution over the nuclear energy levels is now governed by the electron Boltzmann factor. The polarization now spreads to nuclei more distant from the magnetic centers by the «spin-diffusion» process mentioned earlier. It is clear from figure 1 that saturation of the «forbidden transition A» yields polarizations of equal magnitude but opposite sign to that due to the saturation of the «forbidden transition B». Hence polarization of nuclei by the «Solid Effect» has the added advantage that the sign of the polarization can be reversed easily by changing the microwave frequency from $\nu_A$ to $\nu_B$.

PROTON POLARIZATION BY SATURATION OF FORBIDDEN TRANSITIONS

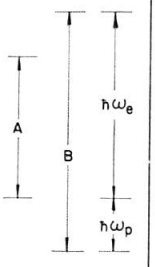
| STRONG FIELD QUANTUM NOS. $m_s$ | $m_i$ | ENERGY LEVELS AND "FORBIDDEN" TRANSITIONS | POPULATIONS $\left(\epsilon_e = e^{-\frac{\hbar\omega_e}{KT}},\ \epsilon_p = e^{-\frac{\hbar\omega_p}{KT}}\right)$ THERMAL EQUIL. | A SATURATED | B SATURATED |
|---|---|---|---|---|---|
| + | − |  | $N_1\epsilon_e\epsilon_p$ | $N_2\epsilon_e$ | $N_3$ |
| + | + | | $N_1\epsilon_e$ | $N_2$ | $N_3\epsilon_e$ |
| − | − | | $N_1\epsilon_p$ | $N_2$ | $N_3/\epsilon_e$ |
| − | + | | $N_1$ | $N_2/\epsilon_e$ | $N_3$ |
| POLARIZATION $\equiv \langle \frac{I_z}{I} \rangle$ | | | $= \frac{1-\epsilon_p}{1+\epsilon_p}$ | $= \frac{1-\epsilon_e}{1+\epsilon_e}$ | $= \frac{\epsilon_e - 1}{\epsilon_e + 1}$ |
| | | | $= \tanh\frac{\hbar\omega_p}{2KT}$ | $= \tanh\frac{\hbar\omega_e}{2KT}$ | $= -\tanh\frac{\hbar\omega_e}{2KT}$ |
| | | | $\cong \frac{\hbar\omega_p}{2KT}$ | $\cong \frac{\hbar\omega_e}{2KT}$ | $\cong -\frac{\hbar\omega_e}{2KT}$ |
| ENHANCEMENT FACTOR | | | $= 1$ | $\cong \frac{\omega_e}{\omega_p} = 660$ | $\cong -\frac{\omega_e}{\omega_p} = -660$ |



Figure 1
Schematic Diagram of the «Solid Effect»

Now the nuclear physicist may ask, since neither radioactive decay nor scattering experiments have been performed, how is the nuclear polarization produced by this method measured? The answer is by measuring the strength of a nuclear magnetic resonance signal produced by these nuclei. 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 is proportional to the nuclear polarization which is defined as

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

and which has the value

$$P = tanh \frac{\hbar\omega_p}{2kT} \cong \frac{\hbar\omega_p}{2kT}$$

at thermal equilibrium.

Thus we know the absolute polarization if we measure the ratio of the power absorption of the nuclear magnetic resonance in the presence of the microwave field to that occuring in the absence of the microwave field.

Observation of nuclei polarized by the «Solid Effect» has now been reported by several workers and on a variety of substances. Protons in water of hydration in paramagnetic crystals have been polarized at Oxford [16], Berkeley [17] and Saclay [18]. Nuclei in damaged crystals [16], and polymers [17] [18] have also been polarized as have nuclei in a polymer doped with free radicals [18].

## A Proposed Polarized Proton Target

We have made some preliminary studies hoping to achieve a polarized proton target by the use of the «Solid Effect». These studies require an apparatus in which a nuclear magnetic resonance signal may be observed while a microwave magnetic field is applied to the sample. Workers in this field have not to date published the details of how this may be accomplished, but the method we have used (after trying other techniques with varying degrees of failure) is shown in figure 2. We wind a coil for use in the nuclear resonance measurement directly on the sample and place both inside a rectangular microwave cavity (slotted to break eddy current paths) resonating in the $TE_{012}$ mode. Our nuclear resonance circuitry is shown in figure 3 and is notable only for its crudity, which is permissible because of the large nuclear resonance signal encountered here. Earliest measurements were made with a marginal oscillator circuit which was considerably more sensitive

than this arrangement is. However, we grew to mistrust the linearity of the marginal oscillator over the wide range of nuclear resonance absorption and emission strength encountered here. In figure 4 we show a copy of a typical recorder tracing which shows a base-line, the thermal equilibrium nuclear magnetic resonance signal, and the enhanced nuclear magnetic resonance signal. The ratio of these signals is called the enhancement factor.

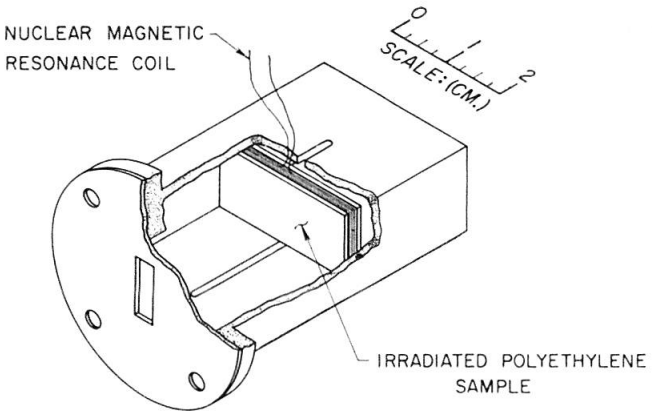



Figure 2

Microwave cavity with sample and nuclear magnetic resonance coil

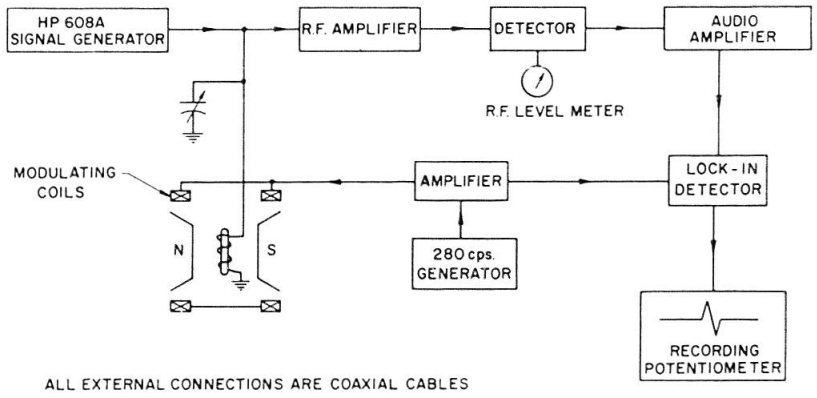



Figure 3

Block diagram of nuclear magnetic resonance spectrometer used for measurement of proton polarization

We have concentrated our attention on the study of proton polarization in polyethylene. We chose polyethylene as the target material because it is simple, containing no other nuclear species than $C^{12}$ which has spin zero and a few low level excited states. Moreover polyethylene also has a very high proton density (0.14 g of proton per $cm^3$ as compared to 0.07 g of proton per $cm^3$ for liquid hydrogen). Paramagnetic

centers are introduced successfully via radiation bombardement of the sample by fast neutrons or 40 Mev protons. Table 1 gives a summary of our results to this date. All data tabulated were obtained at $H_0 \cong 3200$ gauss, $\nu_e \simeq 9$ kmc/s and $T = 1.2°$ K. Note that even the largest enhancement factor observed at liquid helium temperature, which is forty (corresponding to P = 1.2%), falls short of the theoretical expectation which is approximately six hundred sixty. One may well ask what are the factors which limit the enhancement. In the case of polyethylene, one of the limitations may be readily seen by examining the shape of the electron spin resonance line. The absorption line is primarily due to the allowed transitions which we have previously considered to occur at one frequency $\nu_e$, for a given external magnetic field $H_0$. The observed electron spin resonance absorption line actually has a width $\Delta H_0$ of approximately 60 gauss for $\nu_e$ fixed at 9 kmc/s, or correspondingly a frequency width $\Delta\nu_e = \Delta\omega_e/2\pi = H_0 . d\nu_e/dH_0 = 150$ mc/s. The line shape of the electron spin resonance here represents the distribution of the $\nu_e$ values for the various paramagnetic centers. This distribution results from the fact that near each paramagnetic center are many

Table 1

Proton Polarization in Irradiated Polyethylene

| Radiation | Dosage | Sample | Temp. | Max. Enhancement |
|---|---|---|---|---|
| | | | °K | |
| 40 MeV Proton . . . . . | $\sim 2\times10^7$ rad | High Density | 77 | 45 |
| 1.3 MeV $\gamma$ of $Co^{60}$ . . . . | $1.4\times10^7$ roentgen | High Density | 77 | 10·5 |
| Fast Neutron (Brookhaven) | 6 h | High Density | 77 | 8 |
| Fast Neuron (Brookhaven) | 12 h | High Density | 77 | 15 |
| Fast Neutron (Brookhaven) | 60 h | High Density | 77 | 36 |
| Fast Neutron (Argonne) . | Light Brown Color | Low Density | 77 | 4 |
| Electrons . . . . . | $10^7$ rad | Low Density | 77 | 1 |
| Fast Neutron (Argonne) . | Lemon Color | Low Density | 2.1 | 2 |
| Fast Neutron (Argonne) . | Light Brown Color | High Density | 2.1 | 40 |
| 40 MeV Proton . . . . . | $\sim 2\times10^7$ rad | High Density | 1.2 | 22 |
| 1.3 MeV $\gamma$ of $Co^{60}$ . . . . | $1.4\times10^7$ roentgen | High Density | 1.2 | 5 |
| Fast Neutron (Argonne) . . | Light Brown Color 24 h | High Density | 1.2 | 40 |
| Fast Neutron (Brookhaven) | 6 h | High Density | 1.2 | 3 |
| Fast Neutron (Brookhaven) | 12 h | High Density | 1.2 | 8 |
| Fast Neutron (Brookhaven) | 60 h | High Density | 1.2 | 20 |

Radiation dosage of fast neutrons are given either in terms of the color of the sample or the length of bombardment time at a given location in the reactor.

protons, each of which may have its spin up or down, and each of which produces a contribution to the total internal magnetic field at the paramagnetic center. In any event we have an ensemble of paramagnetic centers with a distribution of $\nu_e$ values, whose width is 150 mc/s. We also have corresponding distributions for $\nu_A = \nu_e - \nu_p$ and $\nu_B = \nu_e + \nu_p$ each with a width of 150 mc/s. Thus for a given value of $H_o$ and $\nu$, we will satisfy the condition $\nu = \nu_e$ for some centers, $\nu = \nu_A$ for others and $\nu = \nu_B$ for still others. Consequently we can never find a condition in which we saturate only one type of transition. It has been shown by ABRAGAM [13] that saturation of the «allowed transition» ($\nu = \nu_e$) does not affect nuclear polarization. However, as we have shown previously, saturation of the two «forbidden transitions» will produce competition between centers. Some centers tend to produce enhanced polarization of one sign, whereas others tend to produce enhanced polarization of opposite sign. The most favorable condition in such a case is that which maximizes the difference between the numbers of centers of types A and B. This difference is proportional to the slope of the absorption line shape times $\nu_p$ if $\nu_p \ll \Delta\nu_e$. Both the electron spin resonance absorption derivative and the enhancement are plotted against $H_o$ in figure 5 for a fixed frequency $\nu$. Materials in which $\nu_p > \Delta\nu_e$ also exist and may be expected to yield larger enhancements. Such materials have been and are being studied at Saclay [18] as is reported elsewhere in these proceedings.

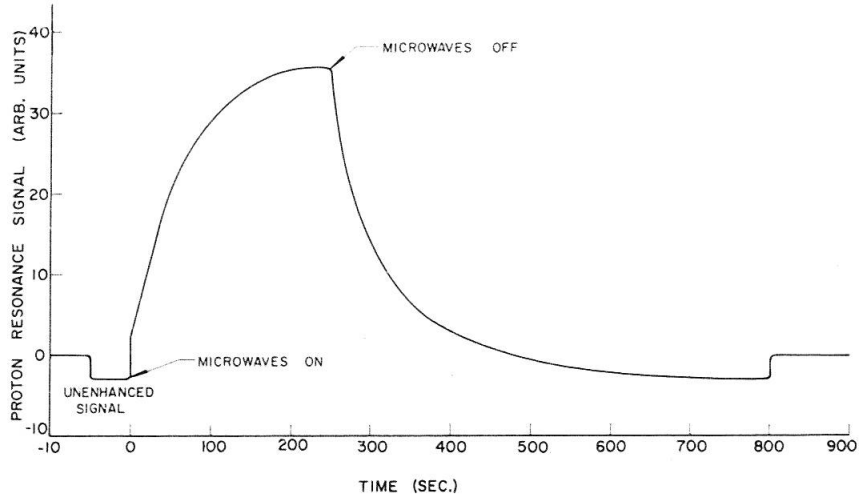



Figure 4

Copy of a typical enhancement observation

For a material like polyethylene the electron spin resonance line shape and $\Delta\nu_e$ are independent of $H_o$, whereas $\nu_p$ is proportional to $H_o$. Thus we would expect to observe increasing enhancement as we increase $H_o$ [19]

(and $\nu_e$) so long as we continue to satisfy $\nu_p < \Delta\nu_e$. Our next step is therefore to perform similar measurements at higher values of $H_o$ and $\nu_e$. We hope to see proton polarization in the range of 5–15% at 12,000 gauss and 1.2° K. The 5% figure assumes no increase of enhancement factor upon raising $H_o$. As it is rather simple today to extract power inputs of the order of a few milliwatts at temperatures as low as 0.4°–0.5° K by using $He^3$ as the refrigerant, we are designing our present apparatus accordingly and hope to gain an additional factor of two to three in proton polarization by lowering the temperature.

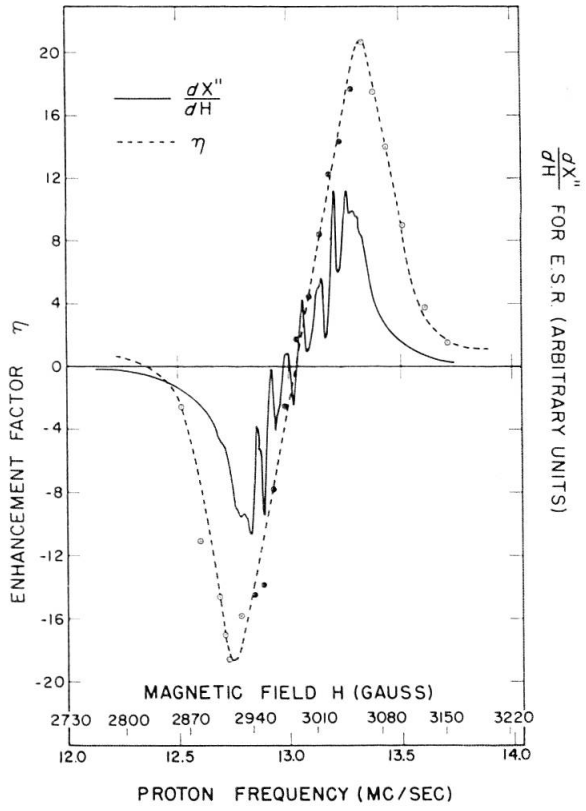



Figure 5

Plot of electron spin resonance line shape (absorption derivative) and enhancement factor for protons in radiation damaged polyethylene at 1.2° K.

In summary the study of the dynamic polarization of protons by the «Solid Effect» in radiation damaged polyethylene at Minnesota has yielded a proton polarization of 1.2% using $H_o \cong 3200$ gauss, $\nu_e \cong 9$ kmc/s. and $T = 1.2°$ K. It appears to be quite feasible to produce proton polarization of 10% or more using $H_o \cong 12{,}000$ gauss, $\nu_e \cong 36$kmc/s and $T = 0.5°$ K. Such a polarized proton target can then be used for scattering experiments. Unfortunately we de not have an operating polarized proton target at this moment, but the design and construction work are now under way.

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[19] Note added in proof: Measurements performed at 3,000 gauss and 4,500 gauss and 1.2° K verified the fact that the enhancement factor is approximately proportional to $H_0$.