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ACHILLES A. SUAREZ, OLGA Y. MAFRA and SILVIO B. HERDADE

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INSTITUTO DE ENERGIA ATÔMICA
Caixa Postal 11049 (Pinheiros)
CIDADE UNIVERSITÁRIA "ARMANDO DE SALLES OLIVEIRA"
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Achilles A. Suarez, Olga Y. Mafra and Silvio B. Herdade

Instituto de Energia Atômica
Nuclear Physics Division
São Paulo - Brasil

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RESUMO

Experiências foram realizadas com o reator de pesquisas tipo piscina do IEA para verificar uma anomalia, relatada por N. Mitrofanov, no espalhamento magnético de neutrons por amostras ferromagnéticas hidrogenadas.

Entretanto, nenhuma anomalia notável foi observada com neutrons epi-térmicos em ferro contendo hidrogênio absorvido e com neutrons térmicos em uma liga de niquel-paládio carregada com hidrogênio.

ABSTRACT

Experiments were carried out at the IEA swimming pool reactor to verify an anomaly, reported by N. Mitrofanov, in the magnetic scattering of neutrons by hydrogenated ferromagnetic samples.

No remarkable anomaly was observed, however, with epi-thermal neutrons in iron containing absorbed hydrogen and

with thermal neutrons in a nickel-palladium alloy charged with hydrogen.

RESUMÉ

On a réalisé des expériences à la pile de recherche type piscine de l'IEA pour vérifier une anomalie, rapportée par N. Mitrofanov, dans la diffusion magnétique de neutrons par des échantillons ferromagnétiques hydrogenés.

Cépendant, on n'a pas constaté d'anomalie frappante avec des neutrons épithermiques dans le fer qui contient hydrogène absorbé at avec des neutrons thermiques dans un alliage de nickel-palladium chargé en hydrogène.

1. INTRODUCTION

Thermal neutrons passing through ferromagnetic samples are polarized in a way to increase neutron population with spin antiparallel to the magnetization direction. One way of measuring neutron polarization is by observing the relative variation of the transmission due to the sample magnetization, what is known as the "single transmission effect" (that will be reffered to as STE in the present paper).

N. Mitrofanov (1) reported a considerable increase in the STE with hydrogen concentration in a Ni-Pd alloy using thermal neutrons. He also observed that epi-cadmium neutrons present a STE in hydrogenated iron similar to that observed with thermal neutrons in pure iron.

Results are presented in this paper for the STE due to the scattering of epithermal neutrons in hydrogenated Armco-type iron and to thermal neutrons in hydrogenated Ni-Pd (18% Ni) alloy.

Some results of control experiments, consisting in the measurement of the STE in pure iron and Ni-Pd (18%), are also presented.

2. MAGNETIC SCATTERING OF NEUTRONS. THE "SINGLE TRANSMISSION EFFECT" (3)

Neutrons have an intrinsic magnetic moment and therefore can interact with magnetic fields and be scattered by them.

The transmission of a beam of neutrons through a magnetized (to saturation) sample is described in terms of two cross sections,

$$\sigma^{\pm} = \sigma_0 \pm p \qquad (1)$$

where the two signs refer, respectively, to neutron spins parallel and antiparallel to the direction of magnetization. σ_0 is the cross section for a completely unmagnetized sample, and includes the effects of magnetic as well as nuclear scattering; p, arises from the interference between the nuclear and magnetic amplitudes.

The most direct and simple method of determining σ_0 and p is by measuring the neutron transmission of unmagnetized and magnetized samples. Consider a slab of a ferromagnetic substance of thickness d and atomic density N. If the sample is in an unmagnetized state, the transmission is:

$$\frac{I}{I_0} = e^{-N \sigma_0 d} \qquad (2)$$

When the sample is magnetized to saturation, the two

possible neutron spin orientation will have different cross sections and, hence, different transmissions. For an initially unpolarized beam ($I_+ = I_- = 1/2\ I_o$):

$$\frac{I_+}{I_o} = 1/2\ e^{-N(\sigma_o + p)\,d}$$

$$\frac{I_-}{I_o} = 1/2\ e^{-N(\sigma_o - p)\,d} \qquad (3)$$

The "single transmission effect", (STE), E_1 is defined as the relative increase in transmission due to the magnetization of the sample:

$$E_1 = \frac{\Delta I}{I} = \frac{(I_+ + I_- - I)}{I} \qquad (4)$$

From equation (3) we can write:

$$E_{1s} = -1 + \cosh Npd \simeq 1/2\ (Npd)^2 \qquad (5)$$

Equation (5) applies only to samples which are magnetized to complete saturation. For unsaturated samples, the single transmission effect has shown, by Halper and Holstein (4), to result in

$$E_1 = E_{1s}\ f\left(\frac{\epsilon\, d}{\delta_\epsilon}\right) , \qquad (6)$$

where $\epsilon = (M_S - M) / M_S$ is the relative deviation of the magnetization from saturation, and δ_ϵ is the characteristic lenght, properties of the sample.

3. EXPERIMENTAL ARRANGEMENT FOR THE MEASUREMENT OF THE SINGLE TRANSMISSION EFFECT (STE). RESULTS OBTAINED FOR ARMCO - -TYPE IRON AND Ni-Pd (18% Ni) ALLOY.

Preliminary experiments were carried out using a neutron beam from the IEAR-1 reactor for the measurement of the STE in Armco-type iron and nickel palladium alloy samples. These preliminary experiments had the purpose of obtaining data related to samples without hydrogen. These data have been afterwards compared with the STE results obtained with the same samples charged with hydrogen in various concentrations. Results of these preliminary experiments were also compared with those published in the literature (5) (6). Nevertheless, the dependence of the unsaturated STE on the crystalline properties of the sample (Eq. 6), makes this comparison not absolute for samples from different origins.

Figure 1 gives the experimental set-up for the measured of the STE. A collimated beam of neutrons (of the order of 10^6/n /cm^2/sec) from beam hole Nº7 of the IEA swimming pool reactor is transmitted through samples placed between the pole pieces of an electromagnet. Cadmium slits are used to define the incident and transmitted beam. The intensity of the transmitted beam is measured by a BF_3 detector. The room background is minimized by a paraffin + boron shield around the detector. The beam transmitted through both sample and detector is stopped by a beam-catcher.

The STE was measured by counting the transmitted neutrons with the magnetic field on and off. Many cycles, of a few minutes counting time in each situation, were repeated during several hours. This procedure was adopted to minimize the influence of the reactor power fluctuations on the measurements. Epithermal neutrons were obtained with a cadmium filter in the beam. Care was taken in order to assure that hysteresis effects did not influence the field-off counts. The applied magnetic field was 10.7 Kilogauss.

For thermal neutrons with a most probable velocity of 2180 m/sec, Hughes, Wallace and Holtzman (6) have obtained, for a 1 cm thick sample of Armco iron, a value of $(2.0 \pm 0.1)\%$ for the STE, using an applied magnetic field of 7000 oersteds. The same experimenters have observed an increase of the STE with the decreasing of neutron velocities until the Bragg cut-off is reached, at about 1000 m/sec.

In our experimental set-up, lead filters with several thicknesses were used in beam hole Nº 7 in order to provide thermal neutron beams with different mean velocities. Table I gives the thermal neutron spectrum mean wave lenght as a function of lead filter thickness.

STE was measured in samples of Armco-type iron, 1 cm thick, consisting of 10 plates 3 x 1.5 x 0.1 cm, at room temperature. Table II gives results obtained with different mean neutron wave lenght $\bar{\lambda} = 3.34$ Å, near the Bragg cut-off of iron.

STE in a Ni-Pd (18% Ni) alloy was measured with neutrons having this mean energy since the Bragg cut-off wave-lenght for nickel is very close to the one for iron. For this Ni-Pd alloy a STE equal to $(0.127 \pm 0.039)\%$ was obtained, at room temperature.

4. SINGLE TRANSMISSION EFFECT IN HYDROGENATED SAMPLES

4.1 Mitrofanov's "uncommon" single transmission effect in hydrogenated samples

N. Mitrofanov (1), utilizing neutrons produced by a Cockroft-Walton accelerator, at University of Chile, has observed an "uncommon" STE in a Ni-Pd (46% Ni) alloy containing absorbed

hydrogen at room temperature. A striking correlation was found between the STE and the amount of hydrogen in the alloy. When an external magnetic field of 5 Kilogauss was applied, the STE varied from a value close to zero, for non hydrogenated samples, 1 cm thick, to a maximum value of $+(5.2 \pm 0.9)\%$ for samples of the same thickness containing 145 mg of absorbed hydrogen. The observed decrease in the magnetic moment of the Ni-Pd alloy with the addition of hydrogen excludes common Bloch type neutron polarization as a possible explanation of this "uncommon effect". Mitrofanov suggests that the phenomenon could be qualitatively understood on the assumption that protons are aligned in the magnetic field of the crystalline structure of the alloy. He also mentions the possibility of proton polarization in the internal magnetic field (about 0.1% and perhaps more) (see appendix I). In a private communication, the explanation of proton alignment by the presence of strong indirect spin - -exchange forces was proposed by Mitrofanov. Hydrogen was introduced into the alloy by electrolysis of an aqueous solution of sulfuric acid to which a small quantity of As_2O_3 was added. Palladium was chosen to form an alloy with a ferromagnetic substance (nickel) because of its capability of absorbing great amounts of hydrogen.

Mitrofanov also suggested that the same "uncommon" effect should be observed with epithermal neutrons in hydrogenated iron. The normal STE in iron without hydrogen is close to zero at epithermal energies.

It was recommended that these experiments should be carried out again with higher fluxes of neutrons, and a program was started in São Paulo, using the IEAR-1 reactor . At the beginning, this program was developped in collaboration

with N. Mitrofanov but, after a few months, it became an independent project that was carried out by the IEA Nuclear Physics Division.

4.2 Measurement of the STE in hydrogenated samples at the IEAR-1

Samples of Armco-type iron 3 x 1.5 x 0.1 cm were electrolytically hydrogenated. Prior to the hydrogenation process the sample surfaces were mechanically and electrolytically polished to avoid the formation of molecular hydrogen by the recombination of ions trapped in irregularities presented by these surfaces. Mechanical polishing has been carried out using a fine sand-paper and diamond powder. Electrolytical polishing (7) has been carried out using the following bath, refrigerated by water at 0°C : 500 cc acetic acid, 25 cc perchloric acid. Stainless steel has been utilized as cathode. The current density was 5 Amp/dm^2 and the polishing time of order of 5 minutes.

Following this polishing procedure the surfaces were examined at a special metallographic microscope in order to verify if some scratches were still remaining.

In the electrolytic hydrogen charging process (7) (8) the iron samples were used as cathode and a lead plate as anode. These electrodes were immersed in a 10 N H_2SO_4 aqueous solution. The current density employed was 10 Amp/dm^2 and the hydrogenation time from one to several hours. To avoid liberation of hydrogen due to heating, the electrolytic cell was cooled by an ice $CaCl_2$ bath. 50 mg of As_2O_3 per liter were added to the electrolyte to facilitate the penetration of hydrogen in iron. Arsenic was used to cause an increase in the ratio of hydrogen absorption.

The above described hydrogenation method turned out to be more effective than the one suggested by Mitrofanov (2) in which a 1N H_2SO_4 solution saturated with arsenic was used. Smialovski (9) has observed that arsenic as a poison in the electrolyte caused an increase of the hydrogen absorption by the cathode only within certain limits of the electrode potential in which the volatile compound AsH_3 is probably formed.

The hydrogen contend of the samples was determined by heating them in vacuum at 300°C and measuring the amount gas evolved by reading the pressure at a constant known volume. It was assumed that at this temperature about 80% of the hydrogen content in iron is released.

Besnard (8) charged Armco iron with hydrogen up to concentrations of 50 cc H/100g Fe without causing any damage to the samples. Beyond this concentration surface bubbles and other surface and internal deffects may appear in the metalic samples. Therefore, if the samples were supersaturated with hydrogen, some internal deffects should be expected and magnetization properties would be altered in a non-controlable caotic way. In the present work surface bubbles were observed when hydrogen concentrations reached 105.5 cc H/100g Fe indicating that supersaturation was probably attained. Duflot (7) supersaturated iron with 140 cc H/100 g Fe.

The hydrogen content of the samples utilized in the verification of the "uncommon single transmission effect" has been determined before and after the neutron transmission measurements, through some monitoring samples.

The hydrogenation of the Ni-Pd samples was also done by electrolysis in a 0.1 N H_2SO_4 aqueous solution with the

addition of 50 mg of As_2O_3 per liter (10). An electric current density of 0.02 Amp/cm^2 was used in this case, and several different charging times. The amount of absorbed hydrogen was measured just like in the case of the iron samples. It was assumed that at 300°C Ni-Pd liberates close to 100% of the total hydrogen content.

The same experimental arrangement and procedures described for the measurement of STE in iron and Ni-Pd, was used in the study of hydrogenated samples, at beam hole nº 7. Ten plates 3 x 1.5 x 0.1 cm were used together to make a 1 cm thick sample. Having in mind the magnetization effects, it would be better to use just one piece of magnetic material but for better penetration of hydrogen the best samples are the very thin ones. So, a compromise has to be assumed and it was decided to use a stack of 10 plates 1 mm thick each.

The results obtained with epithermal neutrons (reactor beam with cadmium filter) in hydrogenated iron are shown in Table III. No remarkable anomaly in the single transmission effect was observed with hydrogen concentrations up to 53.8 cc H/100g Fe.

Table IV shows the results obtained with neutrons having a mean wave lenght $\bar{\lambda}$ = 3.34 angstroms in hydrogenated Ni-Pd (18% Ni) alloy.

5. CONCLUSIONS

The hydrogen concentrations attained by us in Ni-Pd were not large but we have found no reason to improve hydrogenation conditions in view of the negative results obtained with iron samples in our laboratory and results of experiments on Ni-Pd performed simultaneously in other research centers. Van

Loef and Van Dingenen (Mol, Belgium) (5) have observed a small decrease instead of a remarkable increase in the STE with increasing hydrogen concentration in a Ni-Pd alloy. In an experiment carried out with polarized neutrons by Schermer and Sailor (Brookhaven, USA) (11), it was observed a proton polarization of $+(2.5 \pm 5)\%$ in a sample of Ni-Pd containing hydrogen. These authors conclude that the effect observed by Mitrofanov is at least two orders of magnitude too large to be explained in this manner.

So, the results obtained in the present work for iron containing hydrogen as well as the results obtained for hydrogenated Ni-Pd alloys in this laboratory, at the Mol reactor, Belgium (5) and at Brookhaven (11) did not confirm the new effect observed by Mitrofanov (1).

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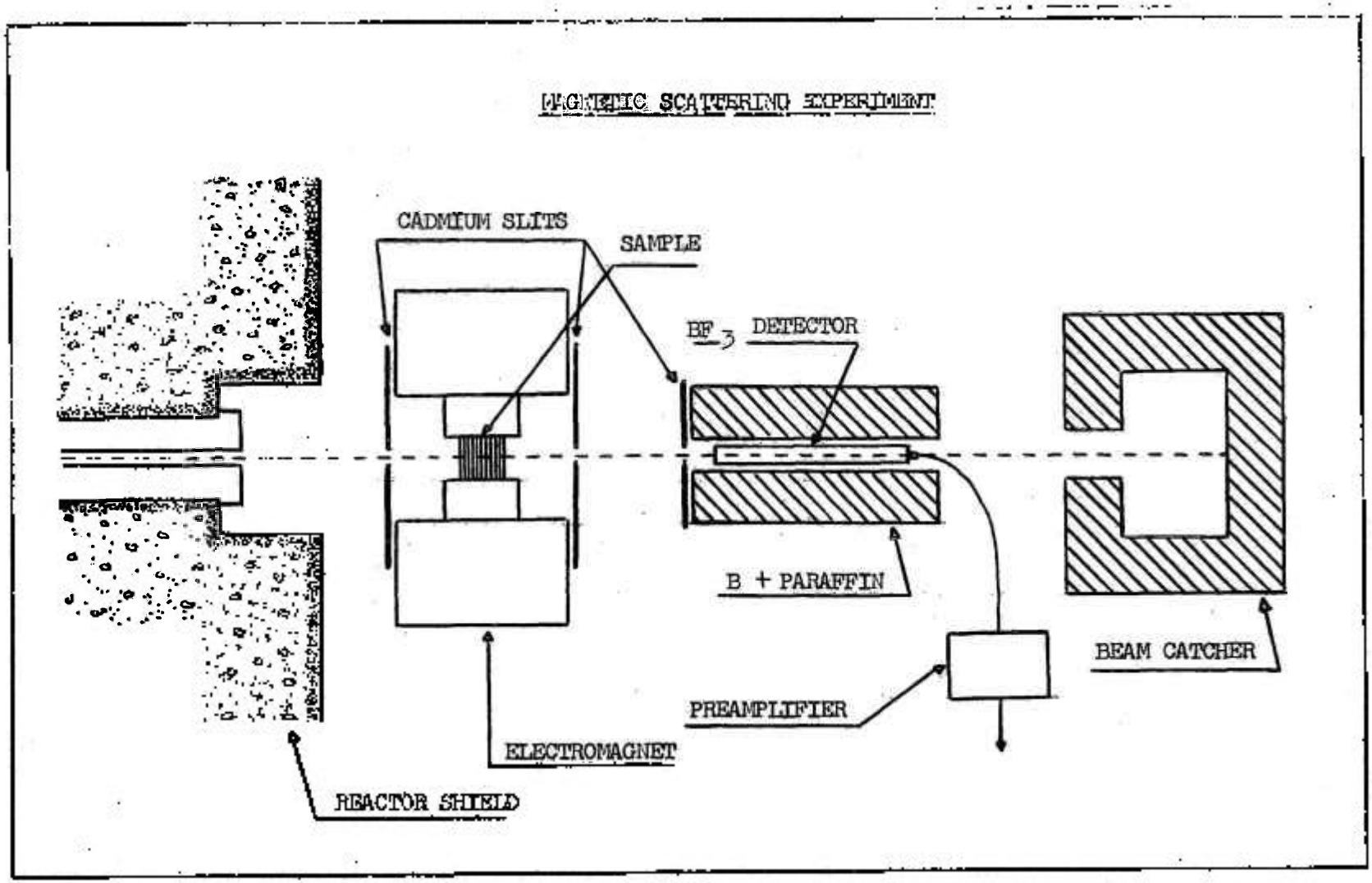
MAGNETIC SCATTERING EXPERIMENT
CADMIUM SLITS
SAMPLE
BF3 DETECTOR
B + PARAFFIN
BEAM CATCHER
PREAMPLIFIER
ELECTROMAGNET
REACTOR SHIELD

FIGURE -I-

TABLE I

Mean neutron velocity, energy and wave lenght as a function of lead filter thickness in the reactor beam

Thickness of lead filter	$\bar{v}$	$\bar{E}$	$\bar{\lambda}$
cm	m/sec	ev	Angstrom
0.0	2719	0.039	1.45
2.5	2633	0.035	1.53
5.0	2385	0.029	1.66
10.0	1885	0.018	2.14
12.5	1625	0.014	2.43
15.0	1446	0.011	2.73
17.5	1313	0.0091	3.01
20.0	1182	0.0074	3.34
25.0	968	0.0049	4.10

The thermal neutron spectrum of the reactor without filter was assumed to be maxwellian. The mean energy of this spectrum was determined experimentally by measuring the transmission of gold with a thin BF_3 detector. The mean energy of the filtered beam was calculed using BNL-325 data for lead.

TABLE III

STE for epi-cadmium neutrons in hydrogenated iron

cm^3 H/ 100g Fe	STE (epi-cadmium neutrons) %
0.0	+(0.006 ± 0.018)
47.7	-(0.050 ± 0.022)
53.8	-(0.196 ± 0.002)

Larger amounts would cause the formation of molecular hydrogen (cracking).

TABLE II

Variation of the STE in pure iron with the mean wave length of the thermal neutron spectrum

Mean wave-lenght of neutron spectrum $\bar{\lambda}$, Angstroms	STE for 1 cm Armco-type iron %
1.53	+(2.18 ± 0.05)
1.66	+(2.51 ± 0.04)
2.14	+(3.26 ± 0.03)
2.43	+(3.69 ± 0.18)
2.73	+(3.94 ± 0.03)
3.01	+(4.03 ± 0.03)
3.34	+(4.00 ± 0.04)
4.10	+(3.97 ± 0.08)

TABLE IV

STE for thermal neutrons ($\bar{\lambda}$ = 3.34 angstroms) in hydrogenated Ni-Pd (18% Ni) alloy

cm^3 H/ 100g Ni-Pd	STE (thermal neutrons) %
0.0	+(0.1270 ± 0.0390)
135.6	+(0.1340 ± 0.0620)
138.4	+(0.0578 ± 0.0500)
170.4	+(0.0190 ± 0.0553)

APENDIX I

One of the hypothesis presented by Mitrofanov as a possible justification for his results, that is, the interstitial proton polarization in the internal magnetic field of the sample, does not explain the change in the STE.

To estimate the proton polarization in the internal magnetic field we can use the well known Brillouim formula for nuclear polarization in magnetic fields (12):

$$f_n = I^{-1} \frac{\sum m_i \exp(-W(m_i)/kT)}{\sum \exp(-W(m_i)/kT)} \qquad (1)$$

where I is the proton spin, m_i the component of this spin in the direction of the field and the sums are over all magnetic sub-states of energy $W(m_i)$; $W(m_i) = m_i \mu H/I$, where μ is the proton magnetic moment. For hydrogen in Ni-Pd alloy at room temperature and internal magnetic fields of about 10^6 gauss the calculated polarization is 3.5×10^{-4}. This assumed value for 0,1°K (3.42×10^5 gauss) (13).

Let's assume that the change in the STE observed by Mitrofanov is due to interactions between polarized neutrons. The cross section for polarized hydrogen is given by (12) : $\sigma = \sigma_0 (1 + \varrho)$ where σ_0 is the cross section without hydrogen and/or neutron polarization.

The relative change in the cross section is:

$$\varrho = 2 I f_n f_p (b/a) \frac{1 - b/2a}{1 + I(I+1)(b/a)^2} \qquad (2)$$

where: $a = (4\pi/k)\ \dfrac{I\eta' + (I+1)\eta''}{2I+1}$

$b = (4\pi/k)\ \dfrac{\eta'' - \eta'}{2I+1}$

f_n = neutron polarization

f_p = proton polarization

k = neutron wave number

η' and η'' are the fase shifts corresponding to the total angular momentum for the singlet and triplet interactions.

If we introduce this expression in the STE definition (1) we get:

$$E_s = 1/2\ (Npd)^2 - n\,\sigma_o\,\rho\,d$$

The value of b/a is negative for hydrogen; f_n and f_p have oposite signs so that $\rho > 0$ and the STE decrease with hydrogen concentration.

For our samples of Ni-Pd, the theoretical saturated STE is

$$E_s = 1/2\ (Npd)^2 = 1.34 \times 10^{-4}$$

where we assumed p = 1 barn for Ni (thermal neutrons) (6)

For the same samples saturated with hydrogen (0 .7 H/Pd) we have:

$$E_s = 1/2\ (Npd)^2 - n\sigma_o\rho\,d = 1.34 \times 10^{-4} - 1.54 \times 10^{-7}$$

From the above theoretical result one should expect significant effect of proton polarization only with magnetic field

of the order of 10^{10} gauss. Any way, this effect would be in a sense to decrease and not to increase the STE.

Note that the calculations above were made supposing that there is no depolarization and the neutrons are completely polarized.

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