

Single Transmission Effect For Slow Neutrons Passing Through Magnetized Pd-Ni Alloy, Containing Absorbed Hydrogen

N. MITROFANOV

Instituto de Fisica y Matematicas, Universidad de Chile, Santiago, Chile

(Received January 26, 1961; revised manuscript received September 12, 1961)

Results are reported on experiments in which a single transmission effect was observed when passing thermal neutrons through magnetized Pd-Ni (46 at.% Ni) alloy containing absorbed hydrogen. A striking correlation was found between the single-transmission effect and the amount of hydrogen in the alloy, when applying an external field of 5000 gauss, the maximum was $(5.2 \pm 0.9)\%$ with a total of 10 sheets. No single-transmission effect was observed with epithermal cadmium neutrons nor with Pd-Ni samples free of hydrogen. The results of polarization measurements with iron and nickel samples agree with the results reported in the literature.

The observed decrease in the magnetic moment of Pd-Ni alloy with added hydrogen excludes common Bloch-type neutron polarization as a possible explanation of the effect mentioned. The phenomenon could be qualitatively understood on the assumption that protons are aligned in the magnetic field of the crystalline structure of the alloy.

INTRODUCTION

IT is well known, that when passing thermal neutrons through magnetized ferromagnetic materials (iron, nickel), the neutrons are polarized and their transmission changed in comparison with the transmission through unmagnetized samples.

The "single-transmission effect" is defined as a relative increase in transmission, due to the magnetization of the sample.¹ The difference in thermal-neutrons transmission, in this case, should be explained as a result of the interference between nuclear and magnetic scattering of the neutron which change the value of the total cross section. The magnetic scattering is a result of the neutron-electron magnetic interaction.

This paper reports on a new effect—the single-transmission effect of thermal neutrons during their passage through the hydrogenated sheets of Pd-Ni alloy. This effect is caused by the change of the total neutron cross section due to neutron scattering.

The Pd-Ni alloy samples, before the transmission experiments, were saturated with hydrogen until they reached 7–8 at.% of hydrogen saturation. The saturation difficulties were overcome by introducing a new electrolytic saturation method.

The ferromagnetic behaviors of the Pd-Ni samples change by means of their hydrogen saturation, so that in the hydrogenated samples the ferromagnetic properties appear more weakly. This fact excludes the explanation of the effect by means of magnetic neutron-electron scattering.

EXPERIMENTAL PROCEDURE AND RESULTS

The experiments were performed with the 800-keV Philips cascade accelerator of the University of Chile. A beam of slow neutrons from beryllium bombarded with 650-keV deuterons, and moderated in a block of paraffin $90 \times 70 \times 70$ cm surrounding the target, passes

through a 30-cm long collimator. This collimator has an opening of $4 \text{ cm} \times 4 \text{ cm}$, it is lined with cadmium, and its two parts are attached to the entrance and exit faces to the ferromagnetic sample. The samples of $4 \times 4 \text{ cm}$ fit snugly into the gap of the electromagnet. The direction of magnetization is at right angles to the neutron beam.

After passing through the sample, the neutrons entered a boron-containing scintillation detector² through a cadmium channel 5 cm in length and an opening of $4 \times 4 \text{ cm}$. The crystal of about 1 mm thickness was mounted with a 50-cm long "light pipe" onto a Du Mont type 6291 photomultiplier carefully shielded from the influence of stray magnetic fields.

The polarization measurements were monitored by a BF_3 proportional counter embedded in the paraffin block 60 cm from the target on the side opposite to the collimator. A series of control experiments were carried out with a soft iron sample, 2 cm thick, in a magnetic field of about 5000 gauss. Counts were recorded with the field alternately on and off for a certain number of monitor pulses. The cadmium background was 35% and was subtracted from the results. The single-transmission effect η is given by

$$\eta = (N' - N) / (N - N_{\text{Cd}}),$$

where N' , N , and N_{Cd} are the total number of counts for an equal number of monitor counts with the iron magnetized, unmagnetized, and with a cadmium shield, respectively. Our value of $\eta = 3.5\% \pm 0.7\%$ for 2-cm iron is somewhat lower than the result of previous measurements of Staub *et al.*,³ which is probably due to a nonideal Maxwellian spectrum of slow neutrons.

In order to determine whether the magnetic field caused any change in the counting rate aside from the polarization effect, a zero check was made by using a

² The scintillator consisted of a compressed mixture of $\text{ZnS}(\text{Ag}) + \text{H}_3\text{BO}_3$, prepared in our Laboratory by N. Mitrofanov.

³ J. Fleeman, D. B. Nicodemus, and H. H. Staub, *Phys. Rev.* **76**, 1774 (1949).

¹ B. T. Feld, *Experimental Nuclear Physics*, edited by E. Segré (John Wiley & Sons, Inc., New York, 1953), Vol. II, p. 557.

TABLE I. The neutron polarization effect in palladium-nickel with different concentrations of hydrogen. Sample No. 1.

Thickness Pd-Ni (cm)	Weight ^a H (mg)	η_{obs}^b (%)
1.0	...	<0.5
0.5	26	≤0.5
0.5	40	1.3±0.5
0.5	47	1.8±0.6
1.0	70	3.4±0.7

^a Total weight of hydrogen in the palladium-nickel.

^b Errors are statistical. The limit of observation of the single-transmission effect was estimated to be 0.5%.

brass block instead of an iron block, with all other conditions unchanged. With such an arrangement it was found that there was no change in the transmission with application of the magnetic field.

In the neutron-proton scattering experiments, an alloy was used which combined ferromagnetic properties with a high absorption power for hydrogen. Because of Schindler's studies on the magnetic and electric properties of palladium-nickel alloys,⁴ we decided to use in our experiments laminas of Pd-Ni, 1 mm thick and 16.4 g in weight, in an atomic ratio of 46% Ni and 54% Pd, which were obtained from Engelhard Industries, Inc., Newark, New Jersey.

Hydrogen was introduced into the alloy by electrolysis of an aqueous solution of sulfuric acid to which a small quantity of As₂O₃ was added. The presence of the latter was essential in order that the laminas absorbed hydrogen in measurable quantity. The amount of hydrogen was estimated by weighing each lamina before and directly after the process of electrolysis. In order to obtain different concentrations of hydrogen, palladium-nickel laminas were electrolyzed during periods varying from a few hours up to within 24 hr. In the latter case, on the average, 8½ at. % of hydrogen was absorbed per lamina.

Single-transmission experiments were done at a constant magnetic field with different thicknesses of pal-

ladium-nickel samples and different degrees of saturation with hydrogen. The experimental conditions were the same as in the case of iron. The laminas were weighed just before each measurement in order to know the amount of hydrogen present. Since each measurement took only a few hours, no correction was applied for the loss hydrogen during the experiment.⁵ The measured single-transmission effects, each with their standard deviation, are given in Table I for the first sample and Table II for the second sample (the second sample has the same composition as sample No. 1). Data were taken so that the statistical counting error in the neutron scintillation detector was less than 1%, and the single-transmission effect was obtained from a series of 20–50 runs with the magnetic field alternately on and off. The most striking result is that η increases with the amount of hydrogen in the palladium-nickel.

The maximum single-transmission effect for the first sample was obtained with 1 cm of palladium-nickel and 70 mg hydrogen. No single transmission effect was observed when a cadmium sheet of 1 mm thickness was present between the sample and the neutron scintillation detector. This means that epicalcium neutrons do not give polarization.

No single transmission effect was observed with 1 cm Pd-Ni without hydrogen. As the magnetic properties of palladium-nickel with 46 at. % nickel are very similar to those of pure nickel,⁶ the polarization due to coherent magnetic scattering should also be comparable. On the basis of the data of Hughes *et al.*⁷ for nickel, η was estimated to be less than 0.5% for a 1-cm sample, which is consistent with our findings. From these results, it is concluded that the single-transmission effect obtained with different thicknesses of palladium-nickel must depend on neutron-proton scattering.

Palladium-nickel laminas that were charged with hydrogen up to supersaturation after repeating the electrolysis several times did not give any single-transmission effect. These laminas were strongly deformed probably as a consequence of some change in the lattice and lost hydrogen very rapidly.

DISCUSSION

Neutron scattering by magnetic atoms is a well known physical process by means of which thermal neutrons can be polarized, and as a consequence of this, the neutron-single-transmission effect appears. However, in the experiments with hydrogen-loaded palladium-nickel sheets, the single-transmission effect was obtained through neutron scattering by protons in the presence of an external magnetic field. The effect is due probably

⁵ The loss of hydrogen was about 5% during the first few days after the electrolysis; afterwards, it slowed down to about 10% in a month.

⁶ R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1956).

⁷ D. J. Hughes, J. R. Wallace, and R. H. Holtman, *Phys. Rev.* **73**, 1277 (1948).

TABLE II. The neutron polarization effect in palladium-nickel with different concentrations of hydrogen. Sample No. 2.

Thickness of Pd-Ni (cm)	Weight of hydrogen (mg)	$\eta = (N' - N)/(N - N_{\text{Ca}})$ (%)	$(N' - N)/N$ (%)
1	0	...	+0.5 ± 0.5
1	90	+3.5 ± 1.4	
1	87	+3.3 ± 1	
1	85	+3.1 ± 1	
1	80	+2.8 ± 0.4	
1	145	+5.2 ± 0.9	
With Cd sheet between sample and detector			$[(N' - N)/N]_{\text{Cd}}$
1	90	...	+0.3 ± 1
1	85	...	-0.3 ± 0.9
1	80	...	+0.25 ± 1.5

⁴ A. I. Schindler, R. J. Smith, and E. I. Salkovitz, *Phys. Rev.* **108**, 921 (1957).

to the difference in the neutron-proton cross section in the presence and in the absence of the magnetic field. It might be suspected that a large part of the protons is aligned along the interior magnetic field and the orientation of the iron microcrystals in the exterior magnetic field might produce a correlation between proton and neutron, causing a change in the cross section. In our next paper we will show this effect.

It is probable that a small part of the interstitial protons are polarized in the interior magnetic field (about 0.1% and perhaps more).

The various methods applied to obtain nuclear polarization which are summarized by Blin-Stoyle and Grace,⁸ are effective at temperature below 1°K only.

⁸ R. J. Blin-Stoyle and M. A. Grace, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42.

In discussing nuclear polarization of impurity atoms in ferromagnetic materials at very low temperature, Marshall⁹ came to the conclusion that the effective magnetic field at these nuclei amounts to 10^5 - 10^6 gauss, with the same intensity and direction throughout a domain.

Our experiments were performed at room temperature and under these conditions a high percentage proton-polarization is improbable, since this requires an exceptional high value of the interior magnetic field.

All these effects can produce proton coupling and alignment in a ferromagnetic crystalline structure at room temperature, which we have experimentally observed.

⁹ W. Marshall, *Phys. Rev.* **110**, 1280 (1958).