

ОБЪЕДИНЕННЫЙ  
ИНСТИТУТ  
ЯДЕРНЫХ  
ИССЛЕДОВАНИЙ  
ДУБНА

E3-85-87

Yu. A. Alexandrov, J. Vavra,\* M. Vrana, I. Kulda,\*  
T. A. Machekhina, P. Mikula,\* R. Michalec,  
V. M. Nazarov, A. I. Okorokov,\*\* V. F. Peresedov,  
V. V. Runov,\*\* L. N. Sedlakova, B. Chalupa \*

SCATTERING OF SLOW NEUTRONS  
ON TUNGSTEN ENRICHED  
WITH  $^{186}\text{W}$  ISOTOPE

Submitted to ЖЭТФ

---

\* Institute of Nuclear Physics (Řež, ČSSR)

\*\* Leningrad Institute of Nuclear Physics (Catchina  
near Leningrad, USSR)

1985

## INTRODUCTION

As is known the isotope  $^{180}\text{W}$  has a small negative nuclear coherent scattering length  $^{1/}$  in the thermal neutron energy range. Due to this one can study some hyperfine effects of neutron-substance interaction which are usually cancelled by strong nuclear scattering. So the interaction between free neutrons and electrons of tungsten atoms (the so-called ne-interaction) was studied in refs.  $^{2, 3/}$ . The reported there experiments were carried out by the method of diffraction of slow neutrons on single crystals prepared from two isotopic mixtures having the scattering amplitudes considerable with those of ne-interaction, but opposite in sign. Since the tungsten is a paramagnetic it was expected that integral diffraction intensities of reflections of monochromatic neutrons on a set of planes  $(hkl)$  should be

$$I_{(hkl)} = K \{ (a + Zf_{(hkl)} a_{ne})^2 + (1 - f_{(hkl)})^2 \gamma^2 \text{ctg}^2 \theta_{(hkl)} \} A_{(hkl)} \frac{\exp(-W_{(hkl)})}{\sin^2 \theta_{(hkl)}}, \quad (1)$$

where  $K$  is the coefficient constant for all the measured reflections,  $a$  is the nuclear scattering length,  $Z$  is the number of electrons in an atom,  $f_{(hkl)}$  is the atomic form-factor for tungsten,  $a_{ne}$  is the scattering length due to ne-interaction,  $A_{(hkl)}$  is the absorption factor,  $\theta_{(hkl)}$  is the Bragg angle,  $\exp(-2W_{(hkl)})$  is the Debye-Waller factor,  $\lambda$  is the neutron wave-

length,  $\gamma^2 \text{ctg}^2 \theta$ , where the term  $\gamma = \frac{1}{2} \mu_n \frac{Ze^2}{Mc^2}$ , takes into account,

the Schwinger scattering.

From eq. (1), it follows that

$$\left[ \frac{I_{(hkl)} \sin^2 \theta \exp(2W)}{kA_{(hkl)}} - \gamma^2 \text{ctg}^2 \theta (1 - f)^2 \right]^{1/2} = a + Zfa_{ne} \quad (2)$$

should linearly depend on  $Zf$  (various consequent reflections  $(hkl)$ ) and that the slope of the curve is due to  $a_{ne}$ . However, the measurements of  $I_{(hkl)}$  have shown that the slopes of the curves for two different isotopic mixtures differ by more than

ОБЩЕИЗВЕСТНОЕ НАУЧНОЕ  
 РАССУЖДЕНИЕ НЕОПРЕДЕЛЕННО  
 2-11-1954

a factor of 2. This result was confirmed in the experiments carried out using various instruments with different parameters (geometry, monochromators, etc.) but operating in about the same neutron wavelength range ( $\lambda = 1.14-1.16 \text{ \AA}$ ). No simple reason for the discrepancy of experimental results from the expected dependence (1) was found (see refs. <sup>3,4/</sup>) and therefore, a hypothesis of additional scattering making a contribution into diffraction peaks was drawn in ref. <sup>5/</sup>. Taking it into account eq. (1) can be transformed into

$$I_{(hkl)} = K \{ (a + Z f_{(hkl)} a_{ne})^2 + (1 - f_{(hkl)})^2 \gamma^2 \text{ctg}^2 \theta_{(hkl)} + p^2 \} \times \exp(-2W_{(hkl)}) / \sin^2 \theta_{(hkl)} \quad (3)$$

where  $p^2$  is some exponential function of  $\frac{\sin \theta}{\lambda}$ . Eq. (3) allowed

one to describe all the experimental data and obtain the same slope of the curves for two isotopic mixtures <sup>3/</sup>. In refs. <sup>2,5/</sup> there was also made an assumption of the magnetic nature of the hypothetical additional scattering.

#### EXPERIMENTS WITH UNPOLARIZED NEUTRONS

##### a) Measurements Using the Neutrons with Different Wavelength

Integral intensities of diffraction reflections of neutrons on the tungsten single crystal were measured with the SPN-100 <sup>6/8/</sup> spectrometer (Institute of Nuclear Physics, Rež, CSSR). It has a Zn single crystal (reflection (0002)) instead of the neutron polarizing Co-Fe single crystal. The construction of the spectrometer allows one to change the wavelength of monochromatic neutrons in the range from 0.7 to 1.6 Å by rotating the crystal and the spectrometer shoulder. The integral intensities  $I_{(hkl)}$  of the three reflections (110), (220), (330) were measured. The targets were the same samples from tungsten as in refs. <sup>2,3/</sup>. All the measurements were performed by the  $\theta-2\theta$  method. The detector aperture and the size of the collimation system were chosen to meet the conditions of measuring the total integral intensity of all the reflections (see, for example, ref. <sup>7/</sup>).

Every reflection intensity  $I_{(110)}$ ,  $I_{(220)}$ , and  $I_{(330)}$  was measured for the following neutron wavelengths 0.7; 1.1; 1.3; 1.6 Å. With account for absorption and angular dependence on  $\theta_{(hkl)}$ , the results were in coincidence with those of refs. <sup>2,3/</sup> within experimental errors not exceeding 1%.

##### b) Measurements at Liquid Nitrogen Temperature

In order to clear up the effect of temperature on the intensity of diffraction reflections of neutrons on the samples under investigation the integral intensities of diffraction peaks (110), (220), (330), (440), (550), and (660) were measured at room temperature (293 K) and at temperature of liquid nitrogen (about 80 K). The measurements were carried out at the IBR-30 reactor by the time-of-flight method without changing the geometry of the arrangement allowing us to determine simultaneously the integral intensities of six orders of reflections. It was obtained that within experimental errors below 3% the temporal dependence of diffraction reflections can be explained by the temperature dependence of the Debye-Waller factor in the frame of the Debye approximation.

#### SMALL ANGLE SCATTERING OF POLARIZED NEUTRONS

Small angle neutron scattering occurs due to diffraction or reflection of neutron waves on inhomogeneities in the sample (e.g., domains in ferromagnetics). The small angle scattering on the inhomogeneities in grains of tungsten powder was mentioned in ref. <sup>5/</sup>. The authors of the present paper have undertaken a search for small angle scattering on the tungsten single crystal using the multichannel apparatus for small angle scattering of polarized neutrons constructed in the Leningrad Institute of Nuclear Physics (Gatchina near Leningrad, USSR). The principal parameters of the apparatus were: mean neutron wavelength  $\langle \lambda \rangle = 8.8 \text{ \AA}$  with a half width  $\Delta \lambda / \langle \lambda \rangle \approx 30\%$ ; the number of counters with the analysis of the scattered neutron polarization was 20. The installation was equipped with a system providing the polarization analysis of scattered neutrons in the three interperpendicular directions: Z - in the direction of the beam, X - perpendicular to the scattering plane, Y - in the scattering plane and perpendicular to Z. The shape of neutron beams with opposite spins directed in the ( $I_{0+}$ ) direction and in the opposite ( $I_{0-}$ ) direction along the X-axis is shown in Fig. 1. In the directions of Y and X axes the picture is the same.

The distribution of the degree of neutron polarization defined by the relation  $P_0 = \frac{I_{0+} - I_{0-}}{I_{0+} + I_{0-}}$  is shown in the same figure.

The samples were prepared both from natural tungsten and from that enriched with 90.7% <sup>188</sup>W isotope. Data on the characteristics  $I_+$ ,  $I_-$ , P of the beam transmitted through tungsten samples are presented in Figs. 2, 3. From the comparison of the curves it

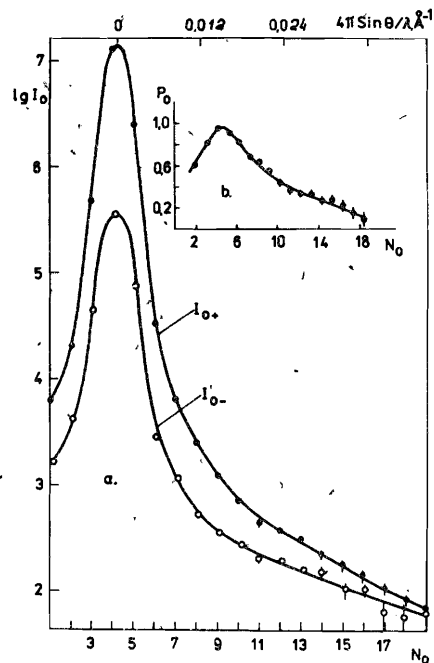


Fig. 1. a) The shape of the neutron beam leaving the installation for the two opposite spin directions: in the direction of the X-axis ( $I_{0+}$ ) and backwards ( $I_{0-}$ ). There is indicated along the X-axis the number of the counter and the momentum transfer. The distance between the counters is  $4.1 \times 10^{-3}$  rad.

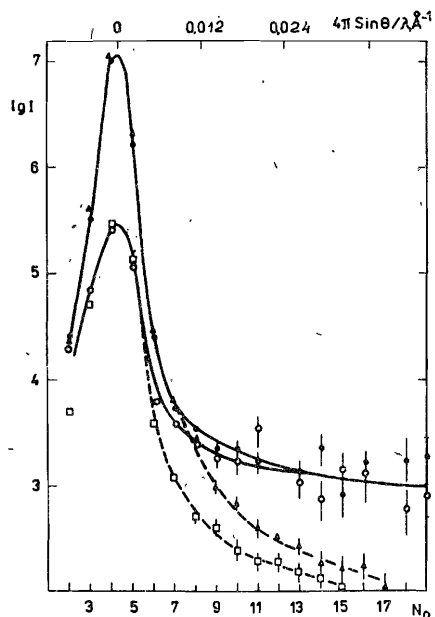


Fig. 2. Characteristics  $I_+$  and  $I_-$  of the beam transmitted through the enriched tungsten sample ( $\bullet - I_+$ ,  $\circ - I_-$ ) and through the sample No. 2 from natural tungsten ( $\Delta - I_+$ ,  $\square - I_-$ ). The radius of the inhomogeneities evaluated from the GUINIER plot of the scattering curve is about  $40 \text{ \AA}$ .

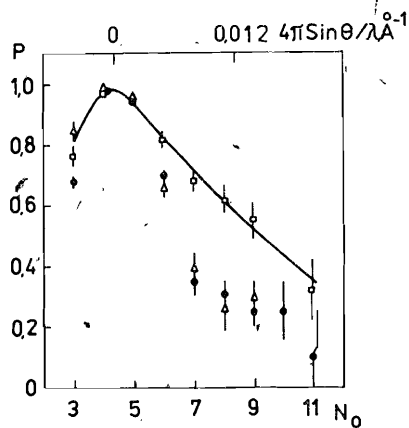


Fig. 3. Distribution of the degree of polarization of the beam transmitted through the sample from enriched tungsten -  $\bullet$ , through the samples No. 1 -  $\Delta$  and No. 2 -  $\square$  from natural tungsten. Solid curve is the distribution  $P_0$ .

Table

Element	Na	Ca	Mg	Cu	Al	Fe	Ni	Cr	Si	Mo
Content in the sample in %	<0.01	<0.01	0.01	0.01	0.07	0.02	0.01	<0.01	0.06	<0.03

is seen that neutron scattering on the enriched with  $^{186}\text{W}$  sample occurs up to  $\sim 2-3^\circ$  and is followed by the depolarization of the beam. Analogous results were also obtained in the Y and Z directions. No difference in X, Y, and Z directions is observed within experimental errors. The scattering and depolarization of the neutron beam is also observed for neutron transmission through one of the samples from natural tungsten (No. 1). But no scattering and depolarization occurred during the neutron beam transmission through another sample from natural tungsten (No. 2).

A possible reason for the observed phenomenon may be the interaction of neutrons with magnetic clusters arising due to magnetic impurities in the sample. Therefore, there was made an analysis of other elements impurities content in tungsten samples. The earlier available data from such an analysis carried out by the "GOSFOND" of isotopes for the enriched tungsten are summarized in the Table. Since in the Table there are no data on the content of a ferromagnetic - Co - a search for its microimpurities was undertaken with a REGATA<sup>9/</sup> apparatus installed at the IBR-2 reactor, JINR.

The cobalt content in tungsten samples was determined using the conventional neutron activation technique. The investigated tungsten samples together with a Co standard sample were irradiated for an hour in the active core of IBR-2. The Co content in tungsten samples was measured by measuring the radioactivity of  $^{60}\text{Co}$  isotope - a product from the  $^{59}\text{Co}(n, \gamma)^{60}\text{Co}$  reaction. The activation of  $^{60}\text{Co}$  in the tungsten sample and in the standard Co sample was determined with the GeLi detector. The areas under photopeaks of  $\gamma$ -ray energy of the radionuclide  $^{60}\text{Co}$  were calculated ( $E = 1173 \text{ keV}$  and  $E = 1332 \text{ keV}$ ). As a result we obtained:

- The content of Co in the sample from tungsten enriched with  $^{186}\text{W}$  isotope was  $(0.6 \pm 0.2)\%$ .
- The content of Co in the sample No. 1 from natural tungsten was  $(0.3 \pm 0.1)\%$ .
- No Co was found in the tungsten sample No. 2.

So there exists a correlation between the small angle scattering of neutrons on tungsten followed by the neutron

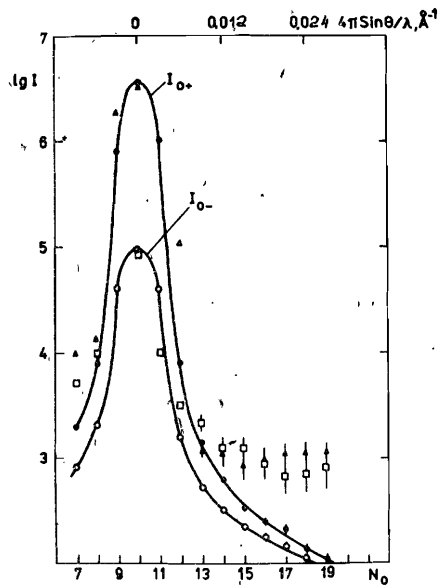


Fig. 4. Small angle neutron scattering on Cd (a foil  $\gamma$  0.05 mm thick).  $\bullet$  -  $I_{0+}$ ,  $\circ$  -  $I_{0-}$ ,  $\Delta$  -  $I_{0+}$ ,  $\square$  -  $I_{0-}$ .

beam depolarization and the content of Co in tungsten samples. As is already mentioned above this may occur due to the formation of magnetic clusters around to Co atoms. Knowing the geometry of the experiment and the number of incident and detected neutrons one may estimate the equivalent scattering amplitude (per an atom of tungsten):  $a_M \sim 3 \cdot 10^{-14}$  cm which coincides with the earlier estimates made in the analysis of neutron diffraction on tungsten single crystals <sup>3/</sup>.

In conclusion let's note that small angle neutron scattering was also observed on other nonmagnetic metals containing magnetic impurities. Fig. 4 illustrates our data on small angle scattering of neutrons on a thin cadmium foil ( $\sim 0.5$  mm) containing about 0.5% of Fe atoms.

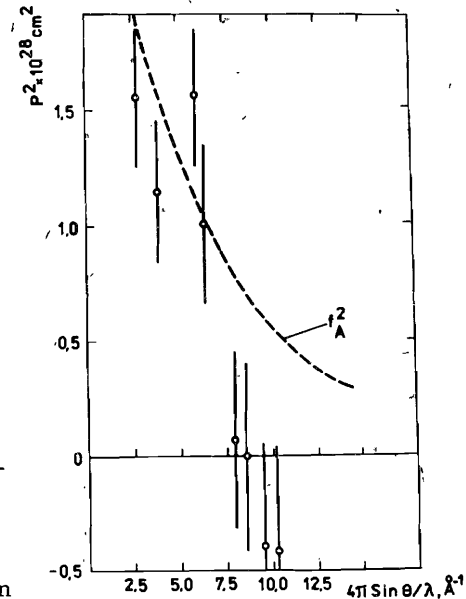
#### ON THE LENGTH OF (ne) - SCATTERING

If the additional diffraction scattering of neutrons on tungsten has a magnetic nature, then in order to find  $a_{ne}$  in the diffraction experiments with tungsten single crystals one should measure the dependence on momentum transfer of the magnetic scattering formfactor connected with  $p^2$  as follows  $p^2 = \frac{2}{3} f_M^2 a_M^2$ . One may try to determine this dependence from the data available in ref. <sup>3/</sup>. Let us assume that at  $\frac{\sin \theta}{\lambda} = 0.663$  (reflection (400))

$p^2 = 0$ , since the magnetic form-factor is a sharply decreasing function of  $\sin \theta / \lambda$ . Data procession under this assumption allows one to obtain both  $p^2 \left( \frac{\sin \theta}{\lambda} \right)$  (see Fig. 5) and  $a_{ne} = (-1.60 \pm 0.05) \times 10^{-16}$  cm (at  $\chi^2 = 1.25$ ). The minimal value of  $|a_{ne}| = 1.55 \times 10^{-16}$  cm is found under condition that  $p^2 \geq 0$  (within er-

\* An assumption of  $p^2 = 0$  at other  $\frac{\sin \theta}{\lambda}$  leads to a rise of  $\chi^2$ .

Fig. 5. Dependence of  $p^2$  on momentum transfer.  $f_A^2$  is the curve for the squared atomic formfactor of tungsten.



rors) in the interval considered, and the maximal value is determined under the condition that not less than two of sixteen points are coming out for two errors off the function (3) (at  $|a_{ne}| = 1.65 \cdot 10^{-16}$  cm).

Thus the most probable value of  $a_{ne}$  obtained in the diffraction on a single crystal of tungsten appeared to be in contradiction with the analogous value from neutron scattering on noble gases <sup>10/</sup>  $(-1.33 \pm 0.03) \cdot 10^{-16}$  cm and about the same as the value obtained in neutron scattering on liquid lead and bismuth <sup>11/</sup>  $(-1.56 \pm 0.05) \cdot 10^{-16}$  cm). The value of  $a_{ne}$  is connected with the mean squared radius of charge distribution in the neutron  $\langle r^2 \rangle = 3 \frac{\hbar^2}{Me^2} a_{ne}$  and with the derivative over squared transferred momentum of electrical formfactor:  $\frac{\partial G(0)}{\partial q^2} = \frac{\hbar^2}{2Me^2} a_{ne}$ . At  $a_{ne} = -1.60 \cdot 10^{-16}$  we have  $\langle r^2 \rangle = -0.14 \cdot 10^{-26}$  cm<sup>2</sup> and  $\frac{\partial G(0)}{\partial q^2} = -0.0231$  fm<sup>2</sup> which coincide with the results of the analysis of data on elastic ed-scattering in the high energy range reported in ref. <sup>12/</sup>.

#### CONCLUSIONS

It is known that there appears a sequence of states (Kondo effect, spin glasses, micromagnetic state) in dependence on the concentration of magnetic impurities in nonmagnetic materials. (e.g., see refs. <sup>13, 14/</sup>). Most studied are at present the disordered magnetics \*  $Pd_{1-x}Fe_x$  and  $Pd_{1-x}Co_x$ , where  $x$  is the concentration Fe and Co impurity atoms. In the fall of 60's it was established with the help of diffuse scattering of neutrons that in Pd there exists a cloud of polarized d-electrons of the matrix around an atom of magnetic impurity <sup>15/</sup>. In recent

\* Systems where magnetic atoms do not form a regular lattice.



years<sup>10</sup> the data are obtained revealing that the magnetic clusters of iron atoms exist in paramagnetic phase. Their size is about 15 Å for the PdFe alloy. Besides these data establish the existence of the quasidomain structure near the critical point (at  $\frac{T_c - T}{T_c} = 10^{-2}$ ). The discovered<sup>2,3,5</sup> in the end of 60's additional scattering of neutrons on tungsten which contributed to Bragg peaks was confirmed by the observed nowadays small angle scattering. It seems to give evidence for the fact that at least some nonmagnetic metals with magnetic impurities have in the paramagnetic phase a tendency of arising a large range ordering leading to the formation of comparatively large magnetic clusters (of the order of dozens of angstrom and more).

The authors wish to express their gratitude to V.L.Aksenov, A.M.Balagurov, S.V.Maleev, R.P.Ozerov, N.M.Plakida, and B.P.Toperverg for fruitful discussions and to Jose Manrike for his help in data procession.

#### REFERENCES

1. Alexandrov Yu.A. JINR, 3-3442, Dubna, 1967, p. 112; Alexandrov Yu.A. et al. Sov.J.Jad.Fiz., 1969, 10, p. 328.
2. Alexandrov Yu.A. JINR, P3-4783, Dubna, 1969.
3. Alexandrov Yu.A. et al. Sov.J.Yad.Fiz., 1974, 20, p. 1190.
4. Alexandrov Yu.A. JINR, E3-5713, Dubna, 1971.
5. Alexandrov Yu.A., Ignatovich V.K. JINR, E3-6294, Dubna, 1972.
6. Michalec R. et al. Yad.Energia, 1966, c. 11, p. 12. 12.
7. Arndt V.W., Willis B.M: Single Crystal Diffractometry, Cambridge, 1966.
8. Mikhailov B.E. et al. Preprint of Leningrad Nucl.Phys.Inst. No. 696, Leningrad, 1981.
9. Nazarov V.M., Frontasieva M.V. Second All-Union Meeting "Nuclear Physics Methods in Environmental Control", Riga, USSR, 1982, p.62.
10. Krohn V., Ringo G. Phys.Rev., 1973, D8, p. 1305.
11. Melkonian B. et al. Phys.Rev., 1959, 114, p. 1571.
12. Bilen'kaja C., Kazarinov Yu.M. Sov.J.Yad.Fiz., 1980, 32, p. p. 742.
13. Korenblit I.Ya., Shender E.F. Uspekhi Fiz.Nauk, 1978, 126, p. 233.
14. Aksenov V.L. et al. IV International School on Neutron Physics, JINR, P3,4-82-704, Dubna, 1982, p. 389.
15. Glow G., Holden T.M. Proc.Phys.Soc., 1966, 89, p. 119.
16. Akselrod L.A. et al. Sov.J.Exp. and Theor.Phys., 1980, 78, Sov.J.Fiz.Tverd.Tela, 1980, 22, p. 3513.

Received by Publishing Department  
on February 7, 1985.

Александров Ю.А. и др.

E3-85-87

Рассеяние медленных нейтронов на вольфраме, обогащенном изотопом <sup>186</sup>W

Приведены результаты экспериментов с неполяризованными и поляризованными нейтронами, выполненных для выяснения природы дополнительного дифракционного рассеяния медленных нейтронов на вольфраме, обнаруженного ранее. Делается вывод, что вероятной причиной такого рассеяния являются магнитные кластеры, образующиеся в вольфраме вокруг микропримесей кобальта. Из имеющихся экспериментальных данных определена величина наклона кривой электрического формфактора нейтрона  $\left(\frac{\partial G}{\partial q^2}\right)_{q^2=0} = -0.0231 \pm 0.0009 \text{ Фм}^2$ .

Работа выполнена в Лаборатории нейтронной физики ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1985

Alexandrov Yu.A. et al.

E3-85-87

Scattering of Slow Neutrons on Tungsten Enriched with <sup>186</sup>W Isotope

The results are reported of the experiments with polarized and unpolarized neutrons carried out in order to understand the nature of the earlier discovered additional diffraction scattering of slow neutrons on tungsten. The conclusion is arrived at that the most probable reason for it is the formation of magnetic clusters around Co microimpurities present in tungsten. The value of the slope  $\left(\frac{\partial G(0)}{\partial q^2}\right)$  of the electric formfactor of the neutron is found on the basis of the experimental data to be  $\frac{\partial G(0)}{\partial q^2} = -0.0231 \pm 0.0009 \text{ fm}^2$ .

The investigation has been performed at the Laboratory of Neutron Physics, JINR.

Preprint of the Joint Institute for Nuclear Research. Dubna 1985