Slow-neutron scattering by ^{186}W-enriched tungsten

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Joint Institute for Nuclear Research
(Submitted 16 February 1985)
Zh. Eksp. Teor. Fiz. **89**, 34–40 (July 1985)

Experiments with polarized and unpolarized neutrons have been carried out to identify the enhanced diffraction scattering of slow neutrons by tungsten which had been discovered previously. The results are reported. This scattering is probably caused by magnetic clusters which form near microscopic cobalt impurities in the tungsten. The slope of the curve of the electric form factor of the neutron is determined from the experimental data available: $(\partial G/\partial q^2)_{q^2=0} = (-0.0231 \pm 0.0009)$ fm^2.

1. INTRODUCTION

At thermal neutron energies the coherent-nuclear-scattering length of the tungsten isotope ^{186}W is small and negative.[1] This property raises the possibility of studying some subtle effects of neutron-matter interactions which are ordinarily masked by strong nuclear scattering. For example, the interaction between free neutrons and atomic electrons of tungsten (the so-called *ne* interaction) was studied in Refs. 2 and 3. These experiments were involved slow-neutron diffraction by single crystals synthesized from two isotopic mixtures, for which the scattering amplitudes were comparable in magnitude to the *ne* interaction and opposite in sign. Since tungsten is paramagnetic, it was expected that the total intensities of the diffraction reflections of monochromatic neutrons from an (hkl) family of planes would be described by

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

where K is a constant coefficient 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 of tungsten; a_{ne} is the scattering length due to the *ne* interaction; $A_{(hkl)}$ is an absorption factor; $\theta_{(hkl)}$ is the Bragg angle; $\exp(-2W_{(hkl)})$ is the Debye-Waller factor, where $W=B(\sin\theta/\lambda)^2$ and λ is the neutron wavelength; and the quantity $\gamma^2\operatorname{ctg}^2\theta$, where

$$\gamma=(\mu_n\hbar/2Mc)(Ze^2/\hbar c),$$

reflects Schwinger scattering.

It follows from (1) that the Zf dependence of the quantity

$$[I_{(hkl)}\sin 2\theta\exp(2W)(A_{(hkl)}K)^{-1}-\gamma^2\operatorname{ctg}^2\theta(1-f)^2]^{1/2} = a+Zfa_{ne} \quad (2)$$

[i.e., the dependence on various successive reflections (hkl)] should be linear, and the slope of the straight line should be governed by a_{ne}. However, the measurements of $I_{(khl)}$ showed that the slopes of this line for the two different isotopic mixtures differed by a factor of more than two. This result has been obtained in several experimental configurations, differing in various parameters (the geometry, the monochromator, etc.), but for approximately the same neutron wavelength ($\lambda\approx 1.14$–1.16 Å). Since no simple factor was found which could explain the deviation of the experimental results from the expected behavior, (1) (see Refs. 3 and 4), it was suggested[5] that additional scattering was occurring and was contributing to the diffraction peaks. Working from this hypothesis, we can rewrite expression (1) as

$$I_{(hkl)}=K\{(a+Zfa_{ne})^2+(1-f)^2\gamma^2\operatorname{ctg}^2\theta+p^2\} \times A_{(hkl)}\exp(-2W_{(hkl)})/\sin 2\theta, \quad (3)$$

where p^2 is some exponential function of $\sin(\theta/\lambda)$. Expression (3) successfully describes all the experimental data available, yielding the same slope for line (2) for the two isotopic mixtures of Ref. 3. It has been suggested[2,5] that this additional scattering is magnetic in nature.

In the present paper we report some experimental results which cast light on the nature of this hypothetical additional scattering.

2. MEASUREMENTS WITH UNPOLARIZED NEUTRONS

a) Measurements with neutrons of various wavelengths. The total intensities of the neutron diffraction reflections of a tungsten single crystal were measured on the SPN-100 spectrometer[6] of the institute of Nuclear Physics, Czechoslovak Academy of Sciences, Řež. The Co-Fe single crystal, which polarizes neutrons, was replaced by a zinc single crystal [the (0002) reflection]. This apparatus is designed to allow measurements of the wavelength λ of the monochromatic neutrons (by rotating the zinc single crystal and the spectrometer arm) between 0.7 and 1.6 Å. The integral intensities $I_{(hkl)}$ were measured for three reflections: (110), (220), and (330). As the targets we used the same tungsten samples as in Refs. 2 and 3. All the measurements of $I_{(hkl)}$ were carried out by the θ-2θ method. The detector aperture and the dimensions of the collimating system were chosen to satisfy conditions for measurement of the total integral intensity of all the reflections (see Ref. 7, for example).

Each value of $I(110)$, $I(220)$, and $I(330)$ is determined

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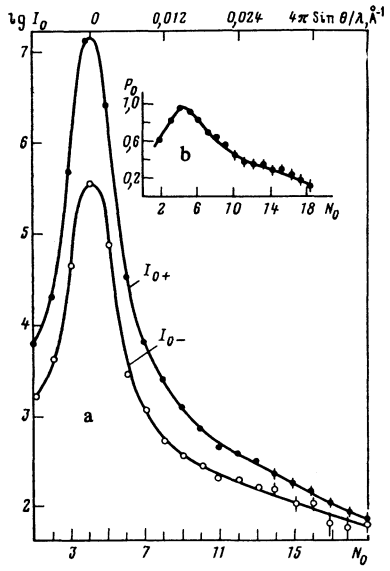

FIG. 1. a—Shape of the neutron beam emerging from the apparatus for two opposite spins of the neutrons, parallel to (I_{0+}) and opposite (I_{0-}) the X axis. The quantities plotted along the abscissa axes are the counter number and the momentum transfer $4\pi\sin\theta/\lambda$, in $Å^{-1}$. The distance between counters is $4.1\cdot10^{-3}$ rad; b—distribution of the degree of polarization P_0 of the neutron beam.

for neutron wave-lengths of 0.7, 1.1, 1.3, and 1.6 Å. Within the experimental error—no greater than 1%—the results of the measurements agree with the corresponding results of Refs. 2 and 3, after correction for absorption and the angular dependence (on $\theta_{(hkl)}$). We found no dependence of any sort of the wavelength.

b) Measurements at liquid-nitrogen temperature. To determine a possible effect of the temperature on the intensity of the neutron diffraction reflections of the test samples, we measured the total intensities of the (110), (220), (330), (440), (550), and (660) diffraction peaks at room temperature (293 K) and liquid-nitrogen temperature ($\approx$80 K). These measurements were carried out at the IBR-30 pulsed reactor of the JINR by a time-of-flight method with a fixed experimental geometry. A temporal analysis of the pulses from the neutron detector made it possible to simultaneously determine the total intensities of all six reflection orders. The results showed that, within the experimental errors, no greater than 3%, the changes in the intensities of the various diffraction reflections as functions of the temperature can be explained on the basis of the temperature dependence of the Debye-Waller factor corresponding to the Debye approximation.

3. SMALL-ANGLE SCATTERING OF POLARIZED NEUTRONS

Small-angle scattering of neutrons arises during the diffraction or refraction of neutron waves by individual inhomogeneities in the test sample (e.g., domains in ferromagnets). That a small-angle scattering of neutrons occurs at internal inhomogeneities in grains of a tungsten powder was pointed out in Ref. 5. In the present study we searched for small-angle neutron scattering in tungsten single crystals using the multichannel apparatus of the Leningrad Institute of Nuclear Physics of studying the small-angle scattering of polarized neutrons.[8] The average neutron wavelength here was $\langle\lambda\rangle = 8.8$ Å; the half-width was $\Delta\lambda/\langle\lambda\rangle \approx 30\%$; and there were 20 data channels for analyzing the polarization of the scattered neutrons. This apparatus was equipped with a system for analyzing the neutron polarization after scattering for three mutually perpendicular initial directions of the neutron spin: Z, along the beam direction; X, perpendicular to the scattering plane; and Y, lying in the scattering plane and perpendicular to Z. Figure 1 shows the shape of the neutron beam for two opposite spin directions, along (I_{0+}) and opposite (I_{0-}) the X axis. The patterns for the Y and Z axes are analogous.

Also shown in Fig. 1 is the distribution of the degree of polarization of the neutrons, found from the relation $P_0 = (I_{0+} - I_{0-})(I_{0+} + I_{0-})$. The samples for these measurements were prepared from both a natural isotopic mixture of tungsten and tungsten enriched to 90.7% in the isotope ^{186}W. Figures 2 and 3 show the results found on the characteristics I_+, I_-, and P of beams transmitted through the tungsten samples. Comparison of these figures shows that in the tungsten enriched in ^{186}W the neutrons are scattered through angles up to $\sim 3°$, and the scattering is accompanied by a depolarization of the neutron beam. Similar results were found for the Y and Z directions. Within the measurement errors, we find no differences among the X, Y, and Z directions. The scattering and depolarization of the beam are also observed when neutrons are passed through a sample with a natural isotopic mixture of tungsten (sample No. 1). On the other hand, the scattering and depolarization do not occur when neutrons are passed through sample of a natural isotopic mixture of tungsten (No. 2).

One possible cause of these effects might be the interaction of a neutron with magnetic clusters which form in a sample containing magnetic impurities. We accordingly

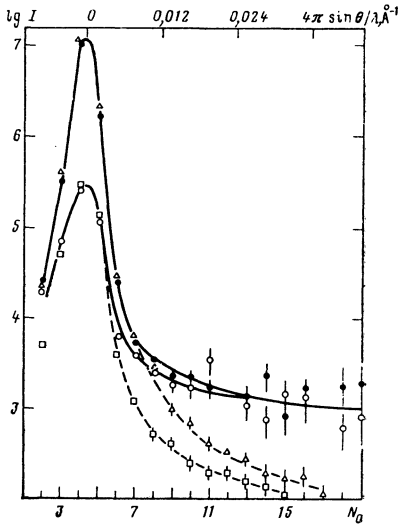

FIG. 2. Characteristics I_+ and I_- of the beam transmitted through an enriched tungsten sample (●—I_+; ○—I_-) and through sample No. 2, of a natural isotopic mixture of tungsten (△—I_+; □—I_-). The scale size of the inhomogeneities found from a Guinier plot is $R \sim 40$ Å.

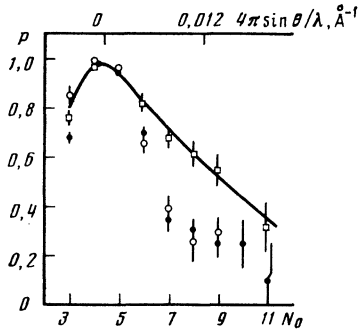

FIG. 3. Distributions of the degree of polarization of the transmitted neutron beams. ●—Enriched tungsten; ○—sample No. 1, of a natural isotopic mixture of tungsten; □—sample No. 2, of a natural isotopic mixture of tungsten. The solid curve is the distribution of P_0.

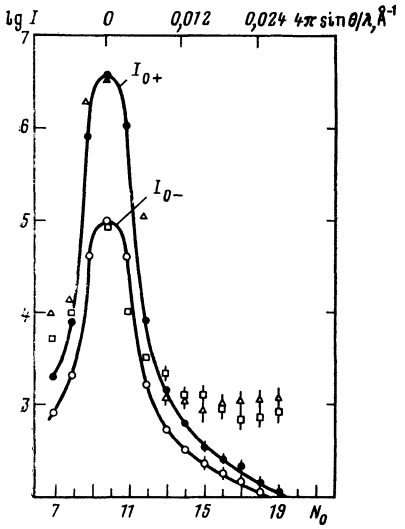

FIG. 4. Small-angle neutron scattering by cadmium (a sheet ~0.05 mm thick). ●—I_{0+}; ○—I_{0-}; △—I_{+}; □—I_{-}.

analyzed the tungsten samples for impurities of other elements. The results of this analysis, which had been carried out previously for the enriched tungsten by the State Isotope Foundation, are listed in Table I. Since this table has no data on the concentration of ferromagnetic cobalt, a search was made for microscopic impurities of this element at the REGATA device[9] of the IBR-2 pulse reactor at the JINR.

The cobalt concentration in the tungsten samples was determined by a standard neutron-activation method. The tungsten samples were bombarded for 1 h in the active zone of the IBR-2 reactor along with cobalt standards. The Co concentration in the tungsten samples was found from the radioactive isotope ^{60}Co, which is the product of the reaction ^{59}Co$(n,\gamma)^{60}$Co. The ^{60}Co activation in the tungsten and in the Co standards was measured with a GeLi detector; the areas under the photopeaks were calculated for the energies of the γ rays from the radioactive nuclide ^{60}Co ($E = 1173$ and 1332 keV). As a result it was found that (a) the cobalt concentration in the ^{186}W-rich tungsten sample is $(0.6 \pm 0.2)\%$, (b) the cobalt concentration in sample No. 1 of natural tungsten is $(0.3 \pm 0.1)\%$, and (c) there was no cobalt in sample No. 2 of natural tungsten.

There is accordingly a correlation between the small-angle scattering of neutrons by the tungsten, accompanied by a depolarization of the beam, and the concentration of the cobalt impurity in the tungsten. As we have already mentioned, the reason for this scattering might be the formation of magnetic clusters around impurity cobalt atoms in the tungsten. Knowing the experimental geometry and the numbers of incident and detected neutrons, we can estimate the equivalent scattering amplitude (per tungsten atom): $a_M \approx 3 \cdot 10^{-14}$ cm. This value agrees with estimates found previously in an analysis of diffraction experiments with neutrons and tungsten single crystals.[3]

To conclude this section, we note that small-angle neutron scattering is also observed for other nonmagnetic metals containing magnetic impurities. Figure 4 shows our data on the scattering of neutrons by a thin ($\approx$0.05-mm) sheet of cadmium containing something on the order of 0.5% iron atoms.

4. *ne* SCATTERING LENGTH

If the additional diffractive scattering of neutrons by tungsten is of magnetic origin, the problem of determining a_{ne} from diffraction experiments with tungsten single crystals reduces to determining the momentum-transfer dependence of the magnetic-scattering form factor F_M, which is related to p^2 by $p^2 = (2/3) f_M^2 a_M^2$. We can attempt to determine this dependence from the diffraction data reported in Ref. 3. Since the magentic form factor is a rapidly decreasing function of $\sin\theta/\lambda$, we assume that at $\sin\theta/\lambda = 0.663$ Å^{-1} [the (400) reflection] we have $p^2 = 0$. Analysis of the experimental data under this condition reveals both $p^2(\sin\theta/\lambda)$ (Fig. 5) and the value $a_{ne} = (-1.60 \pm 0.05) \cdot 10^{-16}$ cm (for[3] $\chi^2 = 1.25$). The minimum value, $|a_{ne}| = 1.55 \cdot 10^{-16}$ cm, is determined by the condition $p^2 > 0$ (with allowance for the errors) in this interval of $\sin\theta/\lambda$, while the maximum value is determined by the condition that at least two of the 16 experimental points deviate from the functional dependence (3) by two standard deviations (with the value $|a_{ne}| \geqslant 1.65 \cdot 10^{-16}$ cm).

The most probable value for a_{ne} found from the diffraction experiments with tungsten single crystals thus contradicts the corresponding value found from experiments on neutron scattering by inert gases[10] [$(-1.33 \pm 0.03) \cdot 10^{-16}$ cm], while it agrees approximately with a value found from experiments on the scattering of neutrons by liquid lead and

TABLE I.

Element	Na	Ca	Mg	Cu	Al	Fe	Ni	Cr	Si	Mo
Conc., %, in sample	<0,01	<0,01	0,01	0,01	0,07	0,02	0,01	<0,01	0,06	<0,03

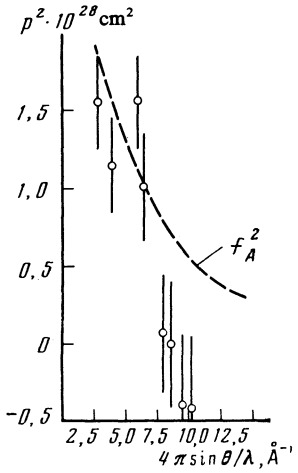

FIG. 5. Dependence of p^2 on the momentum transfer. Here f_A^2 is the square of the atomic form factor of tungsten.

liquid bismugh[11] [$(-1.56 \pm 0.05)\cdot 10^{-16}$ cm]. The quantity a_{ne} is related to the mean square radius of the charge distribution in the neutron,

$$\langle r^2\rangle = 3\hbar^2 a_{ne}/Me^2,$$

and to the derivative of the electric form factor (G) of the neutron with respect to the square of the momentum transfer, q^2,

$$(\partial G/\partial q^2)_{q^2=0} = \hbar^2 a_{ne}/2Me^2.$$

With $a_{ne} = -1.60\cdot 10^{-16}$ cm we find $\langle r^2\rangle = -0.14\cdot 10^{-26}$ cm^2 and $(\partial G/\partial q^2)_{q^2=0} = -0.0231$ fm^2, in agreement with results found in Ref. 12. in an analysis of data on high-energy elastic *ed* scattering.

5. CONCLUSION

A sequence of different states (the Kondo effect, spin glasses, and micromagnetic states; see Refs. 13 and 14, for example) can arise in nonmagnetic metals, depending on the concentration of magnetic impurities. The systems which have been studied most extensively at this point are the disordered magnetic materials in which magnetic atoms do not form a regular crystal lattice) $Pd_{1-x}Fe_x$ and $Pd_{1-x}Co_x$, where x is the concentration of the impurity of Fe or Co atoms. In the late 1960s it was shown by diffuse neutron scattering that magnetic impurity atom in a palladium alloy is surrounded by a cloud of polarized *d* electrons of the matrix.[15] Recent data[16] point to the existence of magnetic inhomogeneities of impurity from atoms (the size of the clusters is on the order of 15 Å) in the paramagnetic phase of the alloy PdFe, while a quasidomain structure is found near the critical point [at $(T_c - T)/T_c \approx 10^{-2}$]. An additional scattering of neutrons by tungsten found[2,3,5] in the late 1960s, which contributes to Bragg peaks and which is confirmed by the existence of small-angle scattering, apparently shows that at least certain nonmagnetic metals containing magnetic impurities have a tendency, in their paramagnetic phase, to form a state with a long-range order which leads to the formation of comparatively large magnetic clusters (with sizes on the order of tens of angstroms and more).

We wish to thank V. L. Aksenov, A. M. Balagurov, S. V. Maleev, R. P. Ozerov, N. M. Plakida, and B. P. Toperverg for useful discussions. We also thank Chosa Mandrika for assistance in analyzing the experimental data.

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Translated by Dave Parsons