Measurement of the amplitdues for scattering of slow neutrons by praseodymium nuclei

G. G. Akopyan, V. P. Alfimenkov, and É I. Sharapov

Joint Institute for Nuclear Research
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The coherent amplitude for scattering of neutrons by praseodymium nuclei is determined by the time-of-flight neutron-diffraction technique in a polycrystalline sample of PrO_2 with known structure. The value obtained is $(0.490\pm0.015)\times10^{-12}$ cm. The total cross section for scattering of neutrons by metallic praseodymium is measured in the energy range 0.5–100 eV and is found to be $\sigma_{sc}=3.1\pm0.2$ b. An upper limit of 0.4 b is obtained for the spin-incoherent cross section.

INTRODUCTION

The amplitudes for elastic coherent scattering b_{coh} and elastic incoherent scattering b_{inc} of neutrons are physical constants characterizing the interaction of slow neutrons with nuclei. They play an important role in diffraction and refraction phenomena in neutron optics.

Particular interest is presented by the spin-incoherent cross section σ_{inc} of praseodymium as a result of the possibility[1] of utilizing neutron diffraction for direct detection of nuclear magnetism in the intermetallic compound $PrCu_2$. Measurements of the specific heat and magnetic susceptibility of this compound[2] have given an indirect indication of antiferromagnetic ordering in it of Pr nuclei at a temperature of about 0.05°K.

In an appropriate diffraction experiment the existence of such ordering of nuclei should lead to appearance of additional coherent peaks with an intensity proportional to σ_{inc}. An accurate knowledge of this quantity is necessary. However, the data existing in the literature for praseodymium, $\sigma_{coh} = 2.4 \pm 0.2$ b and an estimate $\sigma_{inc} \approx 1.6$ b which follows from Table II of Bacon's book,[3] date from 1953 and have not been remeasured since that time.

In the work being reported we have measured the amplitude $b_{coh}(Pr)$ (by diffraction of thermal neutrons in the dioxide PrO_2) and the total cross section for scattering of neutrons in metallic praseodymium in the energy range 0.5–100 eV. These measurements have made possible a new estimate of the incoherent cross section of praseodymium.

METHOD OF MEASUREMENT

1. The structure of the PrO_2 crystal is known from x-ray data,[4] and on this basis it has been assigned to the space group $Fm3m\text{-}O_h^5$ (a structure of the fluorite type). The face-centered-cubic unit cell of PrO_2 has a constant a = 5.39 Å and contains four praseodymium atoms and eight oxygen atoms located as shown in Fig. 1.

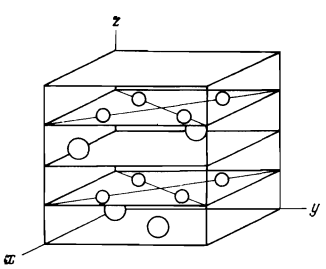

FIG. 1. Unit cell of PrO_2 crystal. The largest circles represent praseodymium ions, and the smaller circles oxygen ions.

In accordance with this structure, the lines of neutron diffraction patterns are separated by intensity into three groups. In the first group (the reflections (111), (311), (331), and so forth) the integrated intensity of the lines is determined by the contribution of only praseodymium atoms, which corresponds to a structure factor of $4b_{Pr}$. The structure factor of the reflections of the second group (the lines (222), (200), . . .) is equal to the difference $4b_{Pr}-8b_O$, and the structure factor of the lines of the third group ((220), 400), . . .) is determined by the sum of the contributions of praseodymium and oxygen, $4b_{Pr} + 8b_O$. This structure is favorable for measurement of $b_{coh}(Pr)$ relative to the well known oxygen scattering amplitude ($b_O = 0.580 \times 10^{-12}$ cm). The greatest accuracy is obtained by comparison of the intensities of the lines of the first and second groups, for which the ratio of the squares of the structure factors is $(1-2b_O/b_{Pr})^2$. In this case a 15% change in the ratio of the line intensities leads only to a 5% error in the value of b_{Pr}. For the neutron-diffraction time-of-flight method in polycrystalline materials, it is well known (see for example ref. 5) that the following relation is valid between the line intensities ΣN_{hkl} and the structure factor F_{hkl}:

$$\sum N_{hkl}\sim\lambda^4 I(\lambda)\varepsilon(\lambda)A(\lambda)\exp\left\{-2B\left(\frac{1}{2d}\right)^2_{hkl}\right\}(jF^2)_{hkl}. \qquad (1)$$

Here we have used the following notation: $I(\lambda)\epsilon(\lambda)$ is the dependence of the neutron flux and detector efficiency on the neutron wavelength, $A(\lambda)$ is the correction for neutron absorption in the sample, $\exp\{-2B(1/2d)^2_{hkl}\}$ is the Debye-Waller thermal factor, and j is a factor due to the recurrence interval of the planes taking part in a given reflection from the polycrystalline sample.

Thus, for measurement of b_{coh} from the ratio of line intensities, it is necessary to know the dependence of the quantity $I\epsilon$ on the wavelength and to take into account accurately the thermal motion of the atoms and the absorption of neutrons in the sample.

2. For neutron energies of 1–100 eV praseodymium has no strong resonances. The effects of the crystal structure and the binding of the atoms in the lattice already have no effect on the scattering cross section in this energy region. If the scattering angle is sufficiently large, paramagnetic scattering also drops out, and measurements in this range of energies give the cross section for scattering by the free nucleus σ_{free} and consequently also the cross section for elastic scattering in a bound atom σ_{bound}, which is related to b_{coh} and σ_{inc} by the expression

$$\sigma_{bound}=\left(\frac{A+1}{A}\right)^2\sigma_{free}=4\pi b_{coh}^2+\sigma_{inc}. \qquad (2)$$

This relation permits the elastic incoherent cross section to be obtained from the known total and coherent cross sections for neutron scattering.

EXPERIMENT AND RESULTS

A powdered sample of PrO_2 of weight 95 g was prepared from the oxide Pr_6O_{11} by the method of slow leaching under the action of dilute acetic acid.[6] Measurements of neutron diffraction by the time-of-flight method were carried out in the IBR pulsed reactor working at a power of 14 kW. The experimental geometry is shown in Fig. 2. The flight path consisted of a distance of 34.5 m from the reactor to the sample and 1.5 m from the sample to the detector. The detector was a battery of 14 boron counters and 5 small helium counters. The sample had transverse dimensions of 90 × 45 mm and was placed in a reflecting geometry. The scattering angle 2θ was 144°. The neutron diffraction pattern obtained in 37 hours is shown in Fig. 3.

The quantity $I\epsilon$ was measured with reduced reactor power with the battery of counters mentioned above in the direct neutron beam. In addition we measured neutron diffraction patterns of tungsten and lead and obtained from them also values of $I\epsilon$, which turned out to be in good agreement with the direct measurements.

In order to take into account accurately the absorption, we measured the transmission (total cross section) of all samples used. In these measurements we used a liquid scintillation neutron detector[7] placed beyond the samples at a distance of 58.5 m from the reactor. To evaluate the Debye-Waller thermal factor we made measurements with the sample at helium temperature.

The measured values of b_{coh} are given in the table. The values of the thermal factor were calculated for B = 0.9. The effective flux $I\epsilon\lambda^4$ is given in relative units. The correction for absorption was calculated from the known formula for symmetric reflection (see for example ref. 5) with use of the measured total cross section. The large value of absorption is due to the presence of water and praseodymium acetate adsorbed on the surfaces of the crystals in preparation of the sample. Experimental values of the areas of the peaks ΣN_{hkl} are given with their statistical errors. Values of the squares of the structure factors (with an accuracy to an unknown normalization constant C) were obtained from Eq. (1). The agreement of the F^2 values for the (220) and (400) reflections indicates the correct allowance for the wavelength-dependent factors in Eq. (1). In the last column we have given the coherent amplitudes and their statistical errors, calculated from the ratios of the structure factors of the corresponding reflections to the value of F_{311}. The averaged value of the amplitude is

hkl	λ, Å	j	$\exp\{-2B\left(\frac{1}{2d}\right)^2\}$	$I\epsilon\lambda^4$	$A(\lambda)$	ΣN_{hkl}	cF^2	b_{coh}, 10^{-12} cm
200	5.0	6	0.940	0.40	0.335	3180±200	4210	0.485±0.012
220	3.58	12	0.885	0.69	0.370	61 000±400	22600	0.512±0.010
311	3.05	24	0.840	0.85	0.405	15 000±450	2160	—
222	2.9	8	0.819	0.90	0.420	11 200±450	4500	0.476±0.014
400	2.53	6	0.782	0.97	0.450	46 600±600	22800	0.515±0.011

$$b_{coh}=(0.490\pm0.015)\cdot10^{-12}\ \text{cm}$$

The error in this amplitude value is determined mainly by the accuracy in measurement of the ratio of the quantity $I\epsilon A$ for different wavelengths, which is estimated as 10%(systematic error). The measured amplitude value corresponds to a coherent cross section value

$$\sigma_{coh}=3.02\pm0.19\ \text{b}$$

2. The total cross section for scattering by praseodymium in the energy range 0.5–100 eV was measured in the same geometry with the IBR reactor working with an electron linear accelerator. The sample used was metallic praseodymium in the form of a hollow cylinder with outer diameter 100 mm, inner diameter 80 mm, and height 200 mm. The sample was placed vertically in a neutron beam of width 40 mm. The effective thickness of the sample in this geometry was 5.8×10^{22} nuclei/cm^2.

The praseodymium scattering cross section was measured relative to the known scattering cross section of lead (σ_{Pb} = 10.90 b). A hollow cylindrical sample of lead 100 mm in diameter was used, with a scattering equivalent to that from the praseodymium sample. In this experimental arrangement the correction due to radiative capture of neutrons by praseodymium nuclei (σ_γ = 2b at 1 eV) is taken into account. The amount of the correction was 11% at 1 eV and 4% at 10 eV. The

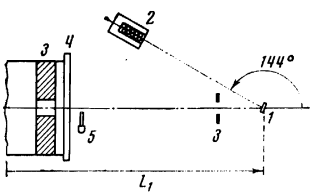

FIG. 2. Experimental geometry: 1–sample, 2–detector in shield, 3–collimators, 4–evacuated neutron pipe, 5–monitor.

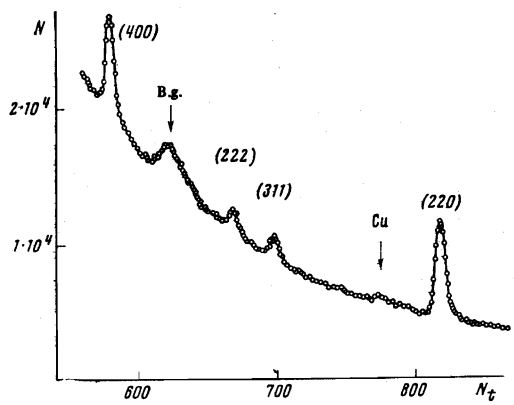

FIG. 3. Part of the experimental neutron diffraction pattern. The horizontal axis is the number of the time-analyzer channel (width 40 μsec), and the vertical axis is the detector counting rate. The peak labeled B.g. is background due to the particular mode of operation of the pulsed reactor (a satellite).

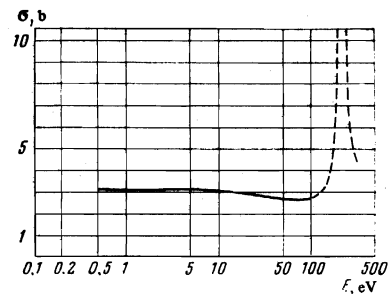

FIG. 4. Neutron scattering cross section in praseodymium. The dashed portion shows the behavior of the cross section in the vicinity of the 216-eV and 234-eV resonances without correction for radiative capture of neutrons.

measured scattering cross section of praseodymium is shown in Fig. 4. In the energy range up to 10 eV no resonance effects are felt, the scattering cross section is

$$\sigma_{sc}=3.1\pm0.2 \text{ b}.$$

On the basis of Eq. (2) with use of the result for the coherent cross section this gives

$$\sigma_{inc}=0.08^{+0.28}_{-0.08} \text{ b},$$

which is equivalent to an upper limit estimate of the incoherent cross section equal to 0.4 b.

DISCUSSION OF RESULTS

The value obtained for the coherent amplitude b_{coh} is 10% higher than the 1953 results.[3] The difference is outside of the experimental error. An argument in favor of the new value is the high reliability of the method used here for normalization to the coherent amplitude of oxygen, which enters into the composition of the sample studied.

A feature of the energy behavior of the measured scattering cross section of praseodymium is a decrease in the cross section in the region 30–100 eV. This is the result of the combined action of interference between resonance and potential scattering and interference between the 216-eV and 234-eV resonances, which have identical spins.

Reduction of the estimate of σ_{inc} from 1.6 b to a value less than 0.4 b has an important bearing on the proposed experiment[1] to observe nuclear magnetism in $PrCu_2$. As a result of the smaller j factor, the intensity of the coherent nuclear maxima with the greatest contribution from Pr atoms in polycrystalline $PrCu_2$ will be 5–10 times weaker than the intensity of peaks of the (311) type in the compound PrO_2 (Fig. 3). For a cross section σ_{inc} 10 times smaller than σ_{coh}, the additional peaks on ordering of the Pr nuclei should be respectively 10 times weaker still. The existence of an appreciable background in diffraction in polycrystalline samples does not permit work with such weak reflections.

The possibility of observing the effect (for the condition $\sigma_{inc} \sim 0.1$ b) remains if a $PrCu_2$ single crystal is used. It is therefore necessary to measure $\sigma_{inc}(Pr)$ more accurately in a direct experiment.

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