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Thorium Coherent Scattering Length Measured by Neutron Interferometry

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Abstract

A very accurate determination of the neutron coherent scattering length of ^{232}Th, b_{Th}, was performed at the neutron interferometer installed at the high-flux reactor of the Institut Laue-Langevin, Grenoble. The value $b_{Th} = 10{\cdot}52$ (3) fm was obtained for a neutron wavelength $\lambda = 1{\cdot}8389$ (6) Å.

The description of the neutron-nucleus scattering problem at low energy is usually based on the nuclear scattering length a. For applications of neutron scattering in condensed-matter investigations the knowledge of the bound scattering length b is needed. In the frame of the Fermi pseudopotential approximation one has $b = a(A+1)/A$, where A is the nucleus-neutron mass ratio. The Fermi model gives good predictions provided that the correct value for b is used.

Many theoretical approaches have been proposed to evaluate the scattering length: for light nuclei few-body models are used (Levinger, 1974; Karchenko & Levashev, 1976; Perne & Sandhas, 1978); for nuclei characterized by nearly closed shells an extension to the continuum of the Bloch and Gillet shell-model formalism has been proposed (Normand, 1977).

However, owing to the still incomplete knowledge of nuclear forces, the neutron-nucleus scattering length must be regarded as a parameter to be determined experimentally.

In this paper, we report the results of a high-precision determination of the coherent scattering length of thorium (natural thorium contains 100% of ^{232}Th). A complete synopsis of the b_{Th} values available in the literature can be found in Koester & Rauch (1981). In the previous work, conventional techniques were used, namely neutron diffraction on ThO_2 powder or single crystals and neutron transmission measurements. The lowest b_{Th} value observed was $b_{Th} = 9{\cdot}8$ (1) fm (Roof, Arnold & Gschneidner, 1962) by neutron diffraction on ThO_2 powder. A very similar value, $b_{Th} = 9{\cdot}84$ (3) fm (Rayburn & Wollan, 1965), was obtained by transmission measurements, whereas higher values were obtained by neutron diffraction on ThO_2 crystals [$b_{Th} = 10{\cdot}00$ (9) fm (Willis, 1963)] and by the recently developed Christiansen filter tech-

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nique [b_{Th} = 10·52 (6) fm (Koester & Rauch, 1981)]. Such a spread is too large to allow a precise interpretation of the sophisticated experiment performed on thorium compounds. For this reason we have performed a b_{Th} measurement by neutron interferometry, a technique that is free of almost all the systematic errors from which the conventional techniques suffer.

Interferometry allows the observation of the modification of the beam state $|\psi\rangle$ induced by some interaction $\hat{A}$. The intensity of the beam in the state resulting from the superposition of the original state $|\psi\rangle$ and the altered state $\hat{A}|\psi\rangle$ is

$$I = \langle \psi \pm \hat{A}\psi \,|\, \psi \pm \hat{A}\psi \rangle. \quad (1)$$

If $\hat{A}$ is a unitary operator, it is possible to write $\hat{A} = \exp(i\hat{\varphi})$, $\hat{\varphi}$ being a Hermitian operator. Then,

$$I = I_0 \pm \langle \cos \hat{\varphi} \rangle. \quad (2)$$

In the case of a neutron wave travelling through a medium of thickness t, the interaction has the form $\hat{A} = \exp[i(2\pi/\lambda)(1-n)t]$, where λ is the neutron wavelength and n is the real part of the refraction index of the medium; n is related to b by

$$n = 1 - \frac{N\lambda^2}{2\pi}\left[b^2 + \left(\frac{\tau_r}{2\lambda}\right)^2\right]^{1/2}, \quad (3)$$

where N is the number of atoms per unit volume and τ_r is the sum of capture, fission and incoherent scattering cross sections. As Th is a zero-spin nucleus the incoherent scattering cross section is zero, and since $(\tau_r/2\lambda)^2 \sim 10^{-8}\, b^2$ the removal term can be neglected in (3); thus, the phase operator $\hat{\varphi}$ can be written as

$$\hat{\varphi} = \lambda N b t. \quad (4)$$

It is then evident that by measuring the intensity of the interfering beams it is possible to determine the b value. A review of the theory of neutron interferometry is given by Bonse & Graeff (1977).

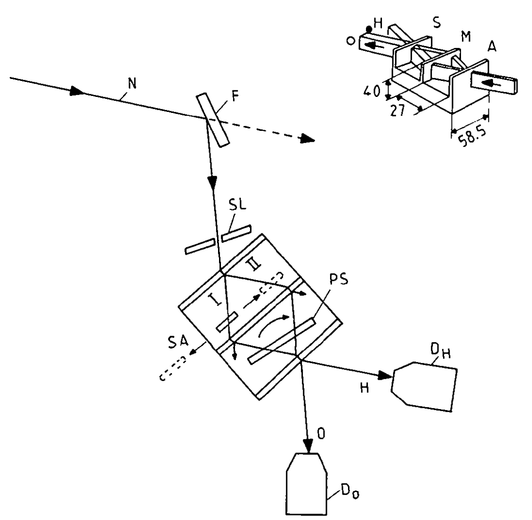

Fig. 1. Drawing of the experimental arrangement. The incident neutron beam N is monochromatized by the first crystal F. The beam cross section is reduced by the slit SL. PS is the Al phase shifter and SA is the thorium sample. The intensities of the outgoing beams O and H are measured by the neutron detectors D_O and D_H.

The experiment presented here was performed at the Laue-type neutron interferometer installed at the Institut Laue-Langevin (Bauspiess, Bonse & Rauch, 1978; Bauspiess, 1978) following an experimental procedure similar to the one we used to determine the coherent scattering length of natural uranium (Boeuf *et al.*, 1982). A diagram of the experimental arrangement is shown in Fig. 1. A monochromatic neutron beam is coherently split in the first of three parallel oriented Si crystals. The two beams so obtained are split again in the second Si slab and interfere on the third. Two ^{3}He detectors measure the intensities of the outgoing beams.

An Al phase shifter consisting of a 5·06 mm thick parallel-sided plate is inserted in the interferometer so as to cross both beams. The phase shifter is then rotated step by step around an axis perpendicular to the scattering plane. In this way, the relative phase of the interfering beams varies, producing a periodical modulation of the emerging neutron beam intensity. At each position of the phase shifter, the neutron intensity is measured with and without a thorium slab in one of the two beam paths. In this way, as shown in Fig. 2, two interferograms with a phase shift $\Delta\varphi = \lambda N b t$ are obtained, where t is the effective thickness of Th crossed by neutrons. However, from the data reported in Fig. 2, it is possible to measure $\Delta\varphi$ excluding any multiple of 2π degrees. In fact, if we put

$$\Delta\varphi = 2\pi m + \Delta P \quad (5)$$

only the ΔP term can be inferred from the data reported in Fig. 2. To determine m unambiguously one has to know b with sufficient accuracy. The spread of the b values reported in the literature being too large a second measurement was performed with a different experimental set up (Bauspiess *et al.*, 1978; Boeuf *et al.*, 1982). The Al phase shifter was replaced

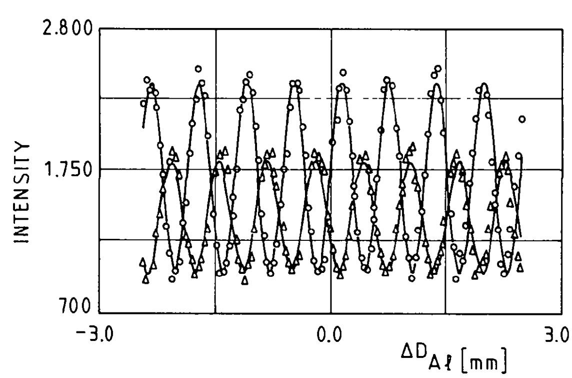

Fig. 2. Neutron intensity of the forward beam as a function of the difference between the path lengths of the two beams in the aluminium phase shifter. The line with circles represents the interferogram for sample out, and the line with triangles the interferogram for sample in.

by the thorium sample, which was rotated in the two interfering beams. An example of the interferograms obtained is reported in Fig. 3, which shows the intensity of the forward beam as a function of the difference between the path lengths of the two beams in thorium.

The oscillation frequency K of the interferogram is related to the b value by (Bauspiess, 1978)

$$K = \lambda N b. \tag{6}$$

Using this technique, the precision with which the b value can be measured depends on the number of cycles used to determine K. However, this number is limited for geometrical reasons by the dimensions of the interferometer and of the sample itself. In our case, from a best fit of the collected data the value $K = 0{\cdot}0579\,(9)\ \mu\text{m}^{-1}$ was obtained. This gives $\Delta\varphi = 66\,(1)$ [$t = 1140\,(2)\ \mu$m] corresponding to a value of $m = 10$. By inserting in (5) $m = 10$ and the ΔP values measured with the first experimental set up, a much more precise value of $\Delta\varphi$ can be found. In fact, a given absolute error in ΔP leads to a very small relative error in $\Delta\varphi$, because of the large m value. For instance, using the data reported in Fig. 2, after correction for the small phase shift due to the air volume corresponding to the sample [$\Delta P_{\text{air}} = 0{\cdot}02\,(1)$] one finds $\Delta P = 3{\cdot}52\,(1)$ and then $\Delta\varphi = 66{\cdot}35\,(1)$, which is in agreement, within the experimental uncertainties, with the previous less-accurate value. Finally, to calculate the b value, λ, N and the effective thickness t have to be measured accurately.

The neutron wavelength λ has been measured by using the interferometer crystal as a spectrometer. The value obtained was $\lambda = 1{\cdot}8389\,(6)$ Å. The atomic density N is related to the mass density ρ and the atomic mass A by $N = \rho N_{\text{av}}/A$, where $N_{\text{av}} = 6{\cdot}0220941 \times 10^{23}\ \text{mol}^{-1}$ is Avogadro's number. The mass density ρ was measured by an Archimedes balance. The measurement was performed on two parallel-sided plates of metallic thorium, 99·5% purity, optically polished. The ρ values obtained were $\rho = 11{\cdot}60\,(1)\ \text{g cm}^{-3}$ for the first sample (sample A) and $\rho = 11{\cdot}58\,(1)\ \text{g cm}^{-3}$ for the sample B, in agreement with the value $\rho = 11{\cdot}72\ \text{g cm}^{-3}$ calculated from the lattice-parameter measured by X-ray diffraction.

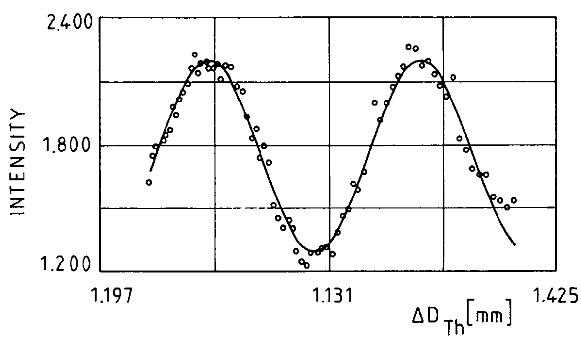

Fig. 3. Neutron intensity of the forward beam as a function of the difference between the path lengths of the two beams in thorium.

In fact, it is possible that some microcavities exist in the sample, so that the measured ρ value is lower than the calculated one. The sample thicknesses were measured with a comparator device and the parallelism checked; the value $t_0 = 1002\,(1)\ \mu$m was obtained for both sample A and sample B. However, the effective thickness t depends on the angular position of the sample with respect to the neutron path. In our case, the thorium slab was inserted in the interferometer parallel to the silicon crystal surface. The orientation in the vertical plane was checked by an optical method while the angular position in the horizontal plane was directly measured by the interferometer. In fact, if the sample is slightly misoriented by an angle δ, the phase shift $\Delta\varphi_{\text{I}}$ produced by the sample in beam path I is different from the phase shift $\Delta\varphi_{\text{II}}$ produced by the sample crossing beam path II, because the effective thickness for the two configurations is different. It is easy to see that

$$\delta = \tan^{-1}\left[\cot\theta_B \frac{\Delta\varphi_{\text{I}} + \Delta\varphi_{\text{II}}}{\Delta\varphi_{\text{II}} - \Delta\varphi_{\text{I}}}\right], \tag{7}$$

where θ_B is the angle corresponding to the Si 220 Bragg reflection at the wavelength used. Therefore, during the rotation of the Al phaseshift, three sets of measurements were performed, namely with the sample out of the neutron beam, with the sample in beam path I and finally with the sample in beam path II.

From the measurements with sample A, the value $\delta = 0{\cdot}0171\,(1)$ rad was found, whereas the value $\delta = 0{\cdot}0165\,(2)$ rad was obtained for sample B. Following the procedure described above we found the value $b = 10{\cdot}54\,(3)$ fm for sample A and the value $b = 10{\cdot}51\,(3)$ fm for sample B. Thus the neutron coherent scattering length of thorium is

$$b_{\text{Th}} = 10{\cdot}52\,(3)\ \text{fm}.$$

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