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Neutron Interferometric Determination of the Coherent Scattering Length of Natural Uranium

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A very accurate determination of the neutron coherent scattering length of natural uranium b_U was carried out with the neutron interferometer installed at the high-flux reactor of the Institut Laue-Langevin, Grenoble. A recently developed technique was used including the rotation of a parallel-sided Al phase shifter with and without the presence of the uranium sample in one path of the neutron beam. From the observed phase shift between the two interferograms the value $b_U = 8.417 \pm 0.005$ fm was obtained.

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The neutron coherent scattering length b describes the behavior of a neutron close to the boundary of a bound nucleus of a given isotope. The precise determination of the neutron coherent scattering length of a nucleus is of importance in fundamental nuclear physics as well as in condensed-matter investigations. A recent review of the b values of several nuclei is given by Koester.[1]

Recent works have been dedicated to the theoretical evaluation of the scattering length in the frame of the nuclear shell model[2] and to the relationship between the b values for bound and free nuclei.[3] However, the neutron scattering length must be still regarded as a phenomenological parameter to be determined experimentally. In recent years a new experimental technique appeared and led to a more precise determination of b, namely neutron interferometry.[4-6]

This paper reports a very accurate determination of the neutron coherent scattering length of natural uranium performed on the neutron interferometer installed at the Institut Laue-Langevin, Grenoble. All the values of b for natural uranium which appear in the literature were obtained, with the exception of one, by collecting Bragg peak intensities on powder or single crystals. These values range from 8.4 ± 0.2 to 8.61 ± 0.4 fm.[7] This last value was obtained through the Christiansen filter method.[1]

Recently Cooper and Sakata[8,9] have reanalyzed the different single-crystal and powder neutron-diffraction data which were all obtained with UO_2 samples. They derived a value of the ratio between the neutron coherent scattering length of

U and O as $b_U/b_O = 1.451 \pm 0.002$. Using this result and the value $b_O = 5.805 \pm 0.004$ fm given by a recent compilation[10] one obtains $b_U = 8.42 \pm 0.013$ fm. These determinations of b_U based on Bragg diffraction are affected by several uncertainties.

The measurements were performed on UO_2 samples and not on pure metallic uranium; consequently they take as reference the b of oxygen which has to be known very precisely, and furthermore a very precise determination of the stoichiometry of the sample has to be done which is generally quite difficult. Moreover, in this case the b values were obtained by a multiparameter best fit of the Bragg peak intensities which implied a coupling of the uncertainties on b and the various other parameters: Debye-Waller factor, absorption coefficient, extinction coefficient, and geometrical constant related to the sample detector arrangement. None of these sources of errors are associated with the much more direct neutron interferometric method.

The neutron interferometric technique used has been described by Bonse and Kischko.[11] A monochromatic neutron beam impinges in Laue position on the first of three parallel-oriented perfect Si crystals. The two obtained coherent beams are then diffracted by the second crystal and interfere on the third. The intensity of the emerging neutron beam is measured by two ^{3}He detectors. Both beams cross a phase shifter consisting of a 5-mm-thick parallel-sided Al plate which is rotated step by step around an axis perpendicular to the scattering plane. At each step the neutron intensity was measured with and without an uranium sample crossing one of the two neutron paths. A scheme of the experimental arrangement is shown in Fig. 1 and the interferograms obtained are reported in Fig. 2. The physical quantity which must be derived from Fig. 2 is the phase shift $\Delta\varphi$ due to the presence of the uranium sample. This phase shift is given by

$$\Delta\varphi = (2\pi/\lambda)(1-n)t, \tag{1}$$

where λ is the wavelength of the incident neutron, n the real part of the refractive index, and t the effective thickness of the sample (thickness crossed by the neutron). b is related to n by

$$n = 1 - (N\lambda^2/2\pi)[\,b^2 + (\sigma_r/2\lambda)^2\,]^{1/2}, \tag{2}$$

where N is the number of atoms per unit volume and σ_r is the sum of capture, fission, and incoherent scattering cross sections ($\sigma_r = \sigma_{n,\gamma} + \sigma_{fis} + \sigma_{inc}$). For natural uranium $(\sigma_r/2\lambda)^2 \sim 10^{-8} b^2$; then Eq. (1) becomes

$$\Delta\varphi = N\lambda b t. \tag{3}$$

If t is large enough (typically a few millimeters) the phase shift is greater than 2π and can be

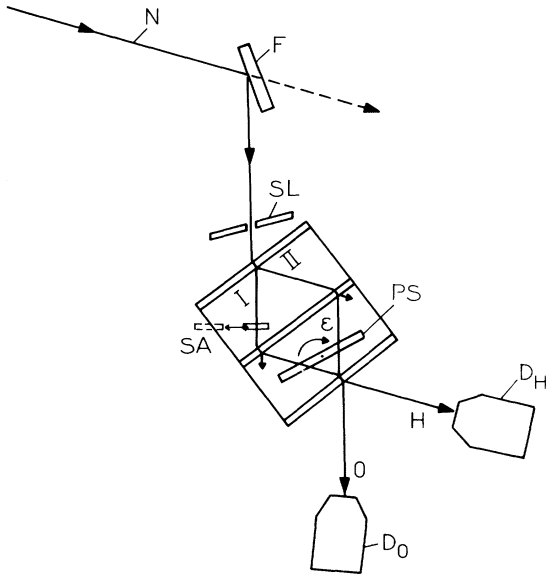

FIG. 1. Drawing of the experimental arrangement. The incident neutron beam N is monochromatized by the fore crystal F. The beam cross section is reduced by the slit SL. PS is the Al phase shifter and SA is the uranium sample. The intensities of the outgoing beams O and H are measured by the neutron detectors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

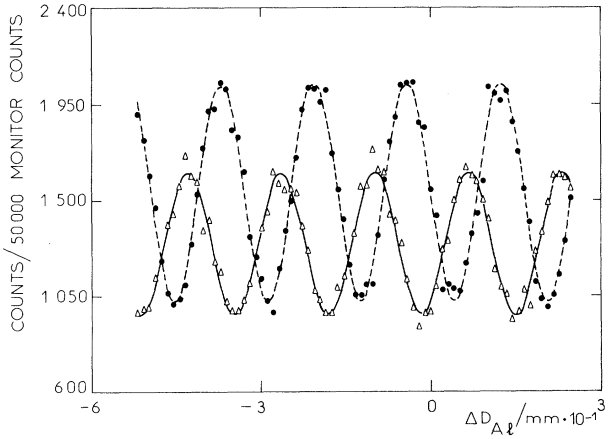

FIG. 2. Neutron intensity of the forward beam as a function of the difference between the path lengths of the two beams in the aluminium phase shifter. The dotted line represents the interferogram for sample out, and the full line the interferogram for sample in.

written as

$$\Delta\varphi_{\text{expt}} = 2\pi m + \Delta P, \quad (4)$$

m being an integer and $\Delta P < 2\pi$. However, from Fig. 2 only ΔP is obtained. This experimental ΔP_{expt}, once corrected for the small phase shift due to the air slab equivalent to the sample ($\Delta P_{\text{air}} = 0.05 \pm 0.01$), becomes $\Delta P = 4.08 \pm 0.01$.

In order to determine the integer number m, a separate less refined experiment was performed.[4] The phase shifter was removed and replaced by the uranium sample which was crossed by the two beams during its rotation. Figure 3 reports the interferogram obtained. The neutron intensity was fitted by the function

$$I = I_0[1 + A\cos(K\Delta D + d)], \quad (5)$$

where ΔD is the difference between the two neutron path lengths inside the uranium sample, A the interference contrast, d the phase shift for $\Delta D = 0$, I_0 the average intensity, and K the oscillation frequency which is related to b by[5]

$$K = Nb\lambda. \quad (6)$$

From a best fit of the data reported in Fig. 3 the value $K = 0.0748 \pm 0.0003$ μm^{-1} was obtained. This gives $\Delta\varphi = 269 \pm 2$ leading to a value of $m = 42$ to be used in the previous more accurate measurement of b. It has to be noted that this method could also be more precise if the number of cycles used to determine K were increased. But this implies a larger rotation of the sample inside the interferometer which is difficult because of a lack of space.

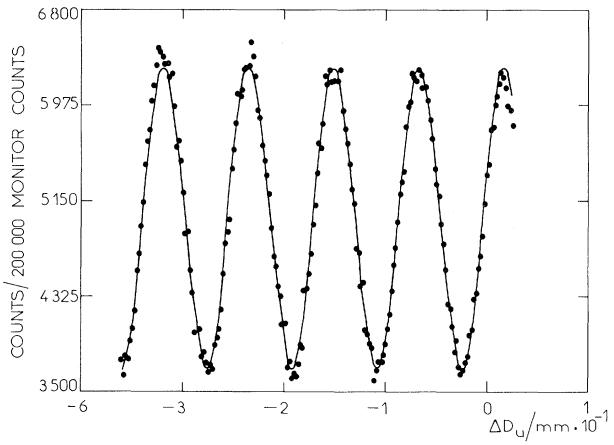

FIG. 3. Neutron intensity of the forward beam as a function of the difference between the path lengths of the two beams in uranium.

From the value of ΔP and m, a precise value of $\Delta\varphi$ is obtained: $\Delta\varphi = 267.97 \pm 0.014$. The advantage of the method used is now evident. The fact that the measurement is based on a determination of ΔP instead of $\Delta\varphi$ implies that a given absolute uncertainty in ΔP leads to a very small relative error in $\Delta\varphi$ because of the large m value.

All the physical quantities involved, namely N, λ, and t have been experimentally determined. The number of uranium atoms per unit volume N is given by $N = \rho N_A/M$, where N_A is the Avogado number, M the atomic mass of natural uranium, and ρ the volumic mass. ρ has been measured with an Archimedes balance. The value obtained is $\rho_U = 19.025 \pm 0.008$ g/cm^3. This value is very close to the value calculated from the lattice parameter measurement ($\rho_U = 19.047$ g/cm^3). The uranium was a 99.99% high-purity metal furnished by the Commissariat à l'Energie Atomique and it has been obtained by electrorefining followed by remelting under vacuum. The uranium platelet was optically polished with diamond paste. The thickness was measured with a comparator device and the parallelism checked ($t = 3156 \pm 1$ μm). The angular positioning of the sample with respect to the neutron path was measured by an optical method and led to an effective thickness $t = 3597 \pm 1.3$ μm. The neutron wavelength has been measured with the interferometer silicon crystal used as a spectrometer. The value obtained is $\lambda = 1.8389 \pm 0.0006$ Å. Thus the neutron coherent scattering length of natural uranium is

$$b_U = (8.417 \pm 0.005) \times 10^{-13} \text{ cm}.$$

The best values obtained using Bragg diffraction[11,12] are slightly higher. The most obvious reason for the discrepancy is the lack of very precise knowledge of the stoichiometry of the oxides. The value obtained by the Christiansen filter method is too large and it should be noted that this was also the case for other nuclides.[7] A rough measurement of $b_{^{235}U}$ has been done[12]; the value obtained is $b_{^{235}U} = (9.8 \pm 0.6) \times 10^{-13}$ cm. Since the content in ^{235}U of natural uranium is very low it is possible to deduce with good accuracy the coherent scattering length of ^{238}U isotope alone, which is

$$b_{^{238}U} = (8.407 \pm 0.007) \times 10^{-13} \text{ cm}.$$

In fact, several terms contribute to the atomic scattering length[1,13]:

$$b = b_{NF} - Zb_F + Zf(\sin\theta/2\lambda)b_{ne}, \quad (7)$$

where b_{NF} is the nuclear force scattering length,

b_F is the Foldy term due to the interaction of the neutron with the electrostatic potential of a bound electronic charge, Z is the atomic number, b_{ne} is the neutron-electron scattering length, and $f(\sin\theta/2\lambda)$ is the electronic form factor. For forward scattering $f=1$. Using the value $b_F = -1.468\times10^{-3}$ fm calculated by Foldy[14] and the value $b_{ne} = (-1.38\pm0.02)\times10^{-3}$ fm measured by Koester, Nistler, and Waschkowski,[13] one obtains a nonnuclear term $b_e = 8.28\times10^{-3}$ fm whose value is greater than the experimental error on the measured b value. Then the nuclear force scattering length for uranium is given by $b_{NF} = (8.409\pm0.0062)\times10^{-13}$ cm.

In conclusion, the very accurate determination of the neutron coherent scattering length of natural uranium makes it possible to obtain an accurate determination of the neutron coherent scattering length of the isotope ^{238}U. We intend to continue these fundamental constant measurements in the actinide series, in particular for ^{235}U and ^{232}Th (for which contradictory results exist).

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Monte Carlo Study of the Isotropic-Nematic Transition in a Fluid of Thin Hard Disks

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The first numerical determination of the thermodynamic isotropic-nematic transition in a simple three-dimensional model fluid, viz., a system of infinitely thin hard platelets, is reported. Thermodynamic properties were studied with use of the constant-pressure Monte Carlo method; Widom's particle-insertion method was used to measure the chemical potential. The phase diagram is found to differ considerably from predictions of a second-virial ("Onsager") theory. Virial coefficients up to the fifth were computed; b_5 is found to be negative.

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Beginning with the work of Onsager[1] a great variety of theoretical models have been proposed that aim to link the thermodynamic properties of liquid-crystal-forming fluids to the intermolecular interactions of the constituent molecules.[2] In assessing the relative merits of these models, direct comparison with experiments on real liquid crystals has often been less than conclusive because disagreement between theory and experiment could be blamed on the use of unrealistic models for the intermolecular interactions, rather than on fundamental deficiencies of the theoretical approach used. For lack of exact results there is clearly a great need for numerical