Experimental report

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Proposal: 1-10-48 **Council:** 10/2020

Title: Accurate values for wavelength-dependent neutron scattering lengths of 141Pr, 152Sm, 154Sm, 153Eu and natEu

Research area: Methods and instrumentation

This proposal is a new proposal

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Samples: PrN
154SmN
EuN
152SmN
153EuN

Instrument	Requested days	Allocated days	From	To
D4	2	2	26/03/2021	30/03/2021
D20	1	1	04/06/2021	05/06/2021

Abstract:

The bound coherent neutron scattering length, bcoh(j), is one of the most basic properties of isotopes and of fundamental importance in many areas of neutron science. Tabulated scattering lengths have sometimes low accuracy (> 5% error) and / or rely on a single measurement only. The scope of this proposal is to make use of neutron powder diffraction for the determination of bcoh(j), in order to improve the accuracy of bcoh(j) values for some important isotopes, 141Pr, 152Sm, 154Sm, 153Eu and the natural isotopic mixture of europium, natEu. This will provide the user community with fundamentally important data needed for a plethora of neutron experiments. The determination relies on relative Bragg reflection intensities and is very robust as for systematic errors. To further minimize the latter, each sample (2.0 g 141PrN, 1.5 g 152SmN, 1.5 g 154SmN, 1.5 g 153EuN, 2.0 g natEuN) will be measured at two diffractometers, D20 and D4, using three wavelengths, which will also address the wavelength dependence of bcoh(natEu). Lanthanide nitrides LnN have been chosen because of their simple rocksalt-type crystal structure without free positional parameters.

Accurate values for wavelength-dependent neutron scattering lengths of ^{141}Pr, ^{152}Sm, ^{154}Sm, ^{153}Eu and ^{nat}Eu (experiment 1-10-48)

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Objectives: The bound coherent neutron scattering lengths, $b_{coh}(j)$, are one of the basic properties of any isotope *j* and many experimental neutron techniques in condensed matter as well as in nuclear physics rely on accurate values. Some of the reported scattering lengths in the widely used collections of scattering lengths [1, 2] have rather low accuracy (> 5% error) and / or rely on a single measurement only and thus clearly need redetermination. In this experiment the bound coherent neutron scattering lengths b_{coh} of the isotopes ^{203}Tl, ^{205}Tl, ^{nat}Tl, ^{141}Pr, ^{nat}Nd, ^{153}Eu and ^{nat}Eu were investigated via neutron Bragg diffraction on polycrystalline powders of cubic $^{203/205/nat}$TlBr, ^{141}PrN, ^{nat}NdN, ^{nat}EuN, hexagonal $^{141}PrCl_3$, $^{nat}NdCl_3$ and monoclinic $^{153}Eu_2O_3$ at room temperature. For extracting the b_{coh} values the Rietveld method was applied using the computer programs FULLPROF [3] and GSAS-II [4] in comparison.

Experimental Details: Due to isotope availabilities at the time of the experiment the initially foreseen isotopes ^{152}Sm and ^{154}Sm were replaced by ^{203}Tl, ^{205}Tl, ^{nat}Tl and ^{nat}Nd. For all samples thin-walled vanadium cylinders of 5 mm inner diameter were used as sample containers. Measurements were performed on the high-intensity diffractometers D20 and D4 in an evacuated vessel. In addition to the sample measurements data were collected for an empty container, the empty vessel and ^{10}B as a strong absorber, in order to obtain information about incoherent parts of scattering. Measurements on samples ^{203}TlBr, ^{205}TlBr, ^{nat}TlBr, ^{141}PrN and ^{nat}NdN were carried out with wavelengths of 70 pm (D4), 136 pm and 187 pm, measurements on $^{141}PrCl_3$ and $^{nat}NdCl_3$ at λ = 136 pm and 187 pm and measurements on ^{nat}EuN at λ = 70 pm. An attempt to investigate ^{nat}EuN at wavelength of 106 and 87 pm (both D20) did not lead to any reliable results. Furthermore, D4 beamtime were used to collect data of ^{6}Li, ^{7}Li, ^{nat}Li and ^{153}Eu at λ = 70 pm in addition to data at λ = 136, 159 and 187 pm obtained in a former experiment (1-10-46). Typical acquisition times were 30 min per sample for D20-data and 2-3 hours for D4-data.

Results and Discussion: Rietveld refinement was performed based on neutron powder diffraction data of the samples, summarized in table 1, with FULLPROF [3] on the one hand and GSAS-II [4] on the other hand, using appropriate crystal structure models. All parameters (i.e. scale, cell, shape, asymmetry, background, position and temperature factors) were refined simultaneously. Background were fitted with a sixth-degree polynomial function, whereby D4 data were background subtracted using empty can and empty vessel data. Since scattering lengths $b_{coh}(j)$ could not be refined directly in either software, the $b_{coh}(j)$ values were obtained via refinement of the site occupation factors for the isotope of investigation, respectively. Values obtained for ^{205}Tl are in good agreement with each other and indicating that $b_{coh}(^{205}Tl)$ is about 2% lower than the one previously reported [2]. Values for ^{203}TlBr are close to previous one in average, but have a huge spread with uncertainties an order of magnitude higher than for $b_{coh}(^{205}Tl)$. This is attributed to the similarity $b_{coh}(^{nat}Br)$ = 6.795(15) [2] and $b_{coh}(^{203}Tl)$ which

causes a high correlation between the scale factor and occupancy in the refinement (Fig. 1), making their determination correspondingly imprecise.

Table 1: Bound coherent neutron scattering lengths, $b_{coh}(j)$, in fm as extracted from Rietveld refinement using GSAS-II (GS) [4] and FULLPROF (FP) [3] for three wavelengths and literature values. Grey values from former experiment 1-10-46 for comparison. For all values isotopic enrichment of samples is already taken into account.

sample	$b_{coh}(j)$ [1]	$b_{coh}(j)$ [2]	D4		D20			
			FP 70 pm	GS 70 pm	FP 136 pm	GS 136 pm	FP 187 pm	GS 187 pm
^{203}TlBr	--	6.99(16)	7.7(3)	7.4(5)	7.1(5)	6.5(6)	6.8(4)	6.9(2)
^{205}TlBr	--	9.52(7)	9.38(9)	9.25(13)	9.30(8)	9.32(8)	9.38(7)	9.40(7)
^{nat}TlBr	8.76(2)	8.776(5)	8.90(8)	8.70(11)	8.67(7)	8.50(8)	8.82(6)	8.67(7)
^{141}PrN	4.58(5)	4.58(5)	4.39(2)	4.38(2)	4.45(2)	4.44(2)	4.46(3)	4.44(2)
$^{141}PrCl_3$			--	--	4.35(2)	4.44(3)	4.41(2)	4.48(2)
^{nat}NdN	7.80(7)	7.69(5)	7.67(8)	7.75(9)	7.91(6)	7.95(8)	7.99(5)	7.98(7)
$^{nat}NdCl_3$			--	--	7.63(3)	7.98(3)	7.79(3)	8.02(4)
$^{153}Eu_2O_3$	8.22(12)	8.22(12)	8.10(4)	8.21(4)	8.17(3)	8.30(4)	8.30(3)	8.28(4)
^{nat}EuN	6.73(3)	7.22(2)	5.91(5)	5.83(6)	--	--	--	--
^{6}LiF	2.0(1)	2.0(1)	2.26(1)	2.25(1)	2.38(2)	2.27(2)	2.70(3)	2.27(3)
^{7}LiF	-2.22(2)	-2.22(2)	-2.28(2)	-2.26(2)	-2.28(1)	-2.30(1)	-2.28(1)	-2.27(1)
^{nat}LiF	-1.90(3)	-1.90(3)	-1.95(2)	-1.96(2)	-1.94(1)	-1.96(1)	1.90(1)	-1.94(1)

In the case of ^{141}Pr values for b_{coh} are quite coherent both for different wavelength lengths and for different compounds as well as for FULLPROF [3] and GSAS-II [4]. The values are lower than the latest value reported in [1] and [2] of 4.58(5) pm, but confirming and older value also given in [1] of 4.45 fm. Similar results were obtained for simple rock salt type PrN as well as for hexagonal $PrCl_3$ as shown in Fig. 2. More structural degrees of freedom (atomic positions in the latter compound) did not decrease precision of b_{coh} determination in a significant way.

Uncertainties obtained for $b_{coh}(^{nat}Nd)$ based on NdN are larger than their ^{141}Pr analogues, while these obtained from trichlorides are comparable. This is attributed to the same effect as for ^{203}TlBr since values for $b_{coh}(^{nat}Nd)$ and $b_{coh}(^{nat}N)$ = 9.36(2) fm are quite similar. In addition, $b_{coh}(^{nat}Nd)$ obtained by refinements on $^{nat}NdCl_3$ data with GSAS-II [4] tend to be significantly higher than by refinement using FULLPROF [3], the reason is still under investigation. Rietveld fits are quite similar to their ^{141}Pr analogues and thus not shown here. In the case of ^{153}Eu, b_{coh} values are comparable to those obtained in former experiment (1-10-46) at higher wavelengths and thus to the tabulated one of 8.22 fm [2]. Finally, values obtained for lithium isotopes at 70 pm are in very good agreement to that obtained at higher wavelengths before.

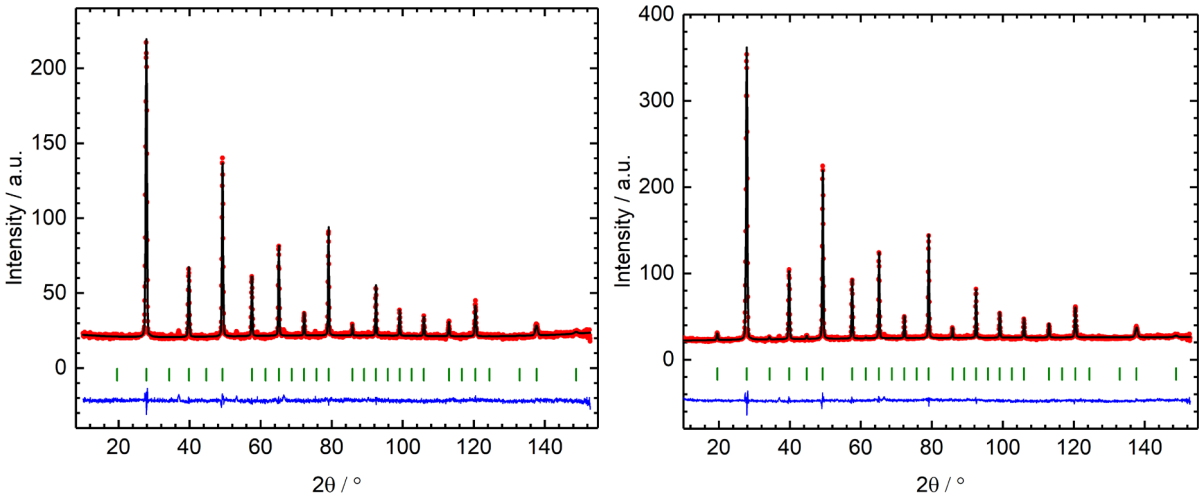

Figure 1: Neutron powder diffraction data taken at D20 (λ = 136 pm) of ^{203}TlBr **(left)** and ^{205}TlBr **(right)** and Rietveld refinement with FULLPROF [3]; measured – red, calculated – black, difference (measured-calculated) – blue, Bragg marker – green

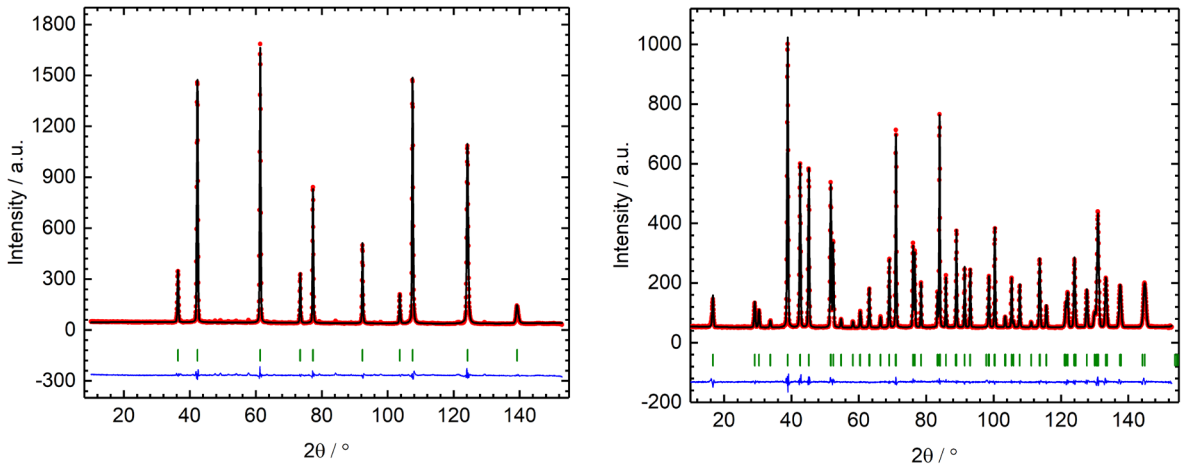

Figure 2: Neutron powder diffraction data taken at D20 (λ = 187 pm) of ^{141}PrN **(left)** and $^{141}PrCl_3$ **(right)** and Rietveld refinement with FULLPROF [3]; measured – red, calculated – black, difference (measured-calculated) – blue, Bragg marker – green

Literature

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