Experimental report

20/05/2021

Proposal: 1-10-46 **Council:** 4/2020

Title: Accurate neutron scattering lengths of 6Li, 148Sm, 154Sm, 153Eu, 156Gd, and 203Tl

Research area: Other...

This proposal is a new proposal

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Samples: Gd2O3
Eu2O3
Sm2O3
LiF
Tl2O3

Instrument	Requested days	Allocated days	From	To
D20	1	1	27/09/2020	28/09/2020
D2B	1	1	16/02/2021	17/02/2021

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, a technique that has been underexploited for the determination of bcoh(j), in order to improve the accuracy of bcoh(j) values for some important isotopes, 6Li, 148Sm, 154Sm, 153Eu, 156Gd, and 203Tl, and thereby provide the user community with fundamentally important data needed for a plethora of neutron experiments. The determination relies on relative Bragg reflection intensities without the need to normalize to an absolute scale. The packing density of samples, exact dimensions or weight – important parameters for other techniques of bcoh(j) determination - do not play any role either. To exclude systematic errors in the diffraction data used for Rietveld refinements, each sample (0.5 g 203Tl2O3, 1 g 6LiF, 1.5 g 148Sm2O3, 1.5 g 154Sm2O3, 1.5 g 153Eu2O3, 1.5 g 156Gd2O3) will be measured at two diffractometers, D2b and D20.

Accurate neutron scattering lengths of ^{6}Li, ^{148}Sm, ^{154}Sm, ^{153}Eu, ^{156}Gd, and ^{203}Tl (experiment 1-10-46)

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Objectives: The bound coherent neutron scattering length, $b_{coh}(j)$, is a basic property of any isotope *j* and of fundamental importance to all areas of neutron science. 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 ^{6}Li, ^{7}Li, ^{nat}Li and ^{153}Eu were investigated via neutron Bragg diffraction on polycrystalline powders of cubic $^{6/7/nat}$LiF 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 ^{148}Sm, ^{154}Sm, ^{156}Gd and ^{203}Tl were replaced by ^{7}LiF and ^{nat}LiF. For all samples thin-walled vanadium cylinders of 6 mm inner diameter were used as sample containers. Measurements were performed on the high-intensity diffractometer D20 in an evacuated vessel without radial collimator. 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. All measurements were carried out with the wavelengths of both 136 pm and 187 pm. Measurements for samples and empty container were also repeated on the high-resolution diffractometer D2B with wavelength of 159 pm under ambient conditions using an automatic sample changer.

Results and Discussion: Rietveld refinement was performed based on neutron powder diffraction data of ^{6}LiF, ^{7}LiF, ^{nat}LiF and $^{153}Eu_2O_3$ with FULLPROF [3] on the one hand side and GSAS-II [4] on the other hand side, using crystal structure models for LiF in space group type $Fm\bar{3}m$ (rocksalt type structure without free positional parameters) and for Eu_2O_3 in space group type $C2/m$ (Sm_2O_3 type structure). Background contributions were not subtracted in this analysis, but fitted with a polynomial function. 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 lithium and europium atoms, respectively.

Preliminary results taking the isotopic enrichment of samples into account reveal significant differences from former determinations (Tab. 1). Bound coherent neutron scattering length, $b_{coh}(j)$, for ^{6}Li not only indicate a scattering length more than 10% higher than the one previously reported [2], but also show a significant difference between results determined using GSAS-II [4] and FULLPROF [3]. This is attributed to the fact that the complex part b_{coh}'' of b_{coh} is neglected in the FULLPROF [3] code, but correctly considered in GSAS-II [4] and indicates that this issue becomes imported for Rietveld analysis of highly absorbing isotopes such as ^{6}Li. This finding is corroborated by the graphical representation of refinement results (Fig. 1, λ = 136 pm), which reveal intensity misfits for some reflections in refinements using FULLPROF

[3]. The same issue is found in refinements based on the data collected at wavelength of 187 pm and 159 pm (both not shown here).

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

j	[2]	*D20*				*D2B*	
		GS 136 pm	**GS 187 pm**	**FP 136 pm**	**FP 187 pm**	**GS 159 pm**	**FP 159 pm**
^{6}**Li**	2.0(1)	2.23(2)	2.28(3)	2.37(3)	2.74(4)	2.31(6)	2.41(7)
^{7}**Li**	-2.22(2)	-2.29(1)	-2.24(1)	-2.26(1)	-2.27(1)	-2.26(1)	-2.33(1)
^{nat}**Li**	-1.90(3)	-1.94(1)	-1.92(1)	-1.94(1)	-1.90(1)	-1.97(2)	-1.94(2)
^{153}**Eu**	8.22(12)	8.30(4)	8.28(4)	8.17(3)	8.27(3)	8.25(6)	8.15(6)

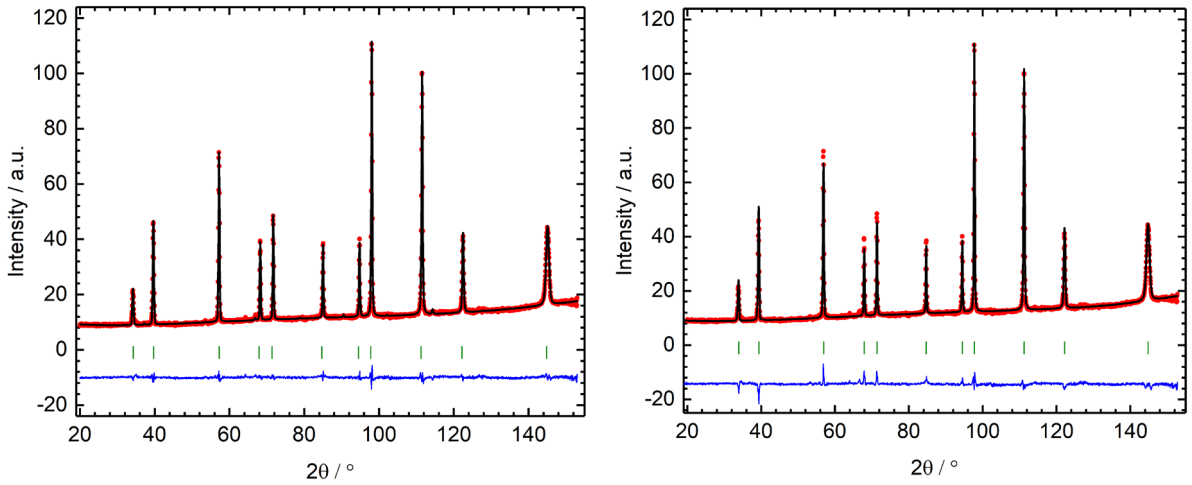

Figure 1: Neutron powder diffraction data taken at D20 (λ = 136 pm) of ^{6}LiF and Rietveld refinement with GSAS-II [4] (**left**) and FULLPROF [3] (**right**); measured – red, calculated – black, difference (measured-calculated) – blue, Bragg marker – green

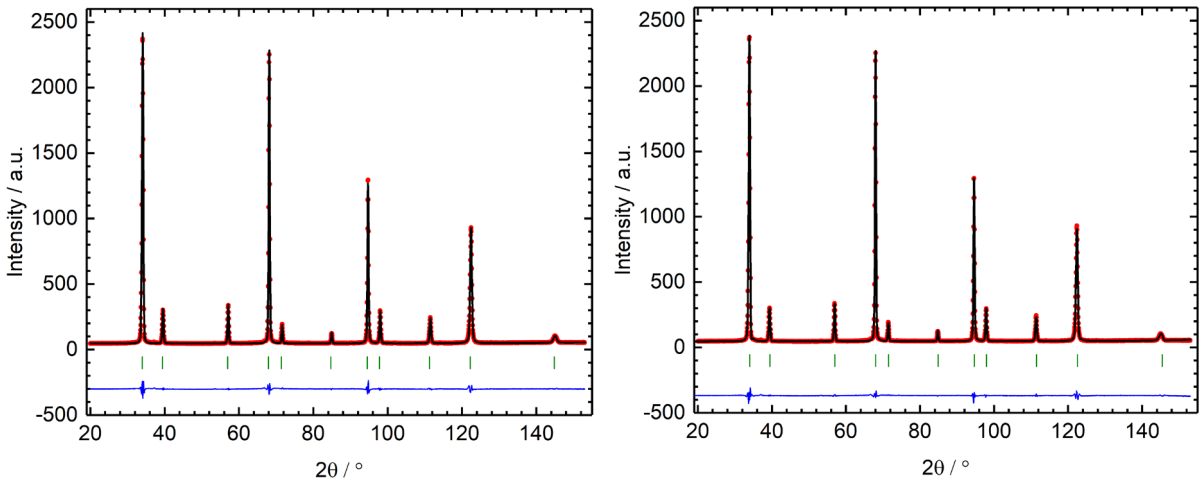

Figure 2: Neutron powder diffraction data taken at D20 (λ = 136 pm) of ^{7}LiF and Rietveld refinement with GSAS-II [4] (**left**) and FULLPROF [3] (**right**); measured – red, calculated – black, difference (measured-calculated) – blue, Bragg marker – green

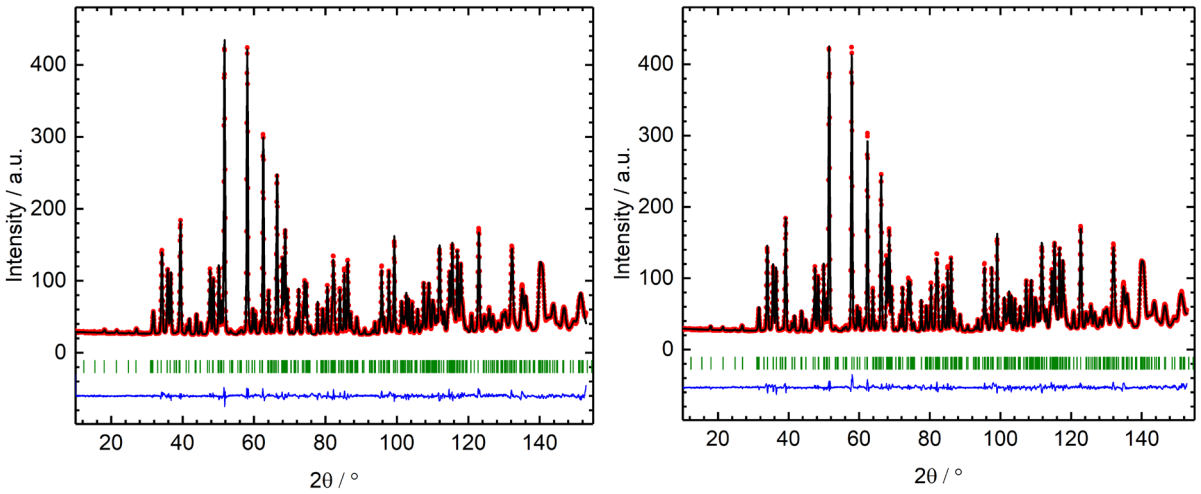

Figure 3: Neutron powder diffraction data taken at D20 (λ = 136 pm) of monoclinic $^{153}Eu_2O_3$ and Rietveld refinement with GSAS-II [4] (**left**) and FULLPROF [3] (**right**); measured – red, calculated – black, difference (measured-calculated) – blue, Bragg marker – green

Rietveld refinements for ^{7}Li and ^{nat}Li with low and moderate neutron absorption could be performed both with GSAS-II [4] and FULLPROF [3] leading to comparable results (Tab. 1, Fig. 2). This supports the above made interpretation of intensity misfits for ^{6}LiF due to neglecting of b_{coh}''.

In the case of ^{153}Eu, b_{coh} values are close to the tabulated one of 8.22 fm [2]. This is in contrast to a former redetermination using cubic bixbyite type $^{153}Eu_2O_3$ (space group type $Ia\bar{3}$) which yielded 8.85 pm [5]. The cause of this difference is still under investigation. The monoclinic Sm_2O_3 type modification used in this experiment (1-10-46) has more free positional parameters for potential correlation with the site occupancy factor as compared to the formerly used bixbyite type modification [5]. In view of this discrepancy, it will be expedient to check the obtained values with measurements on a crystal structure having no free positional parameters, e. g. EuN in the rocksalt type, in the future. Rietveld refinements performed both with GSAS-II [4] and FULLPROF [3] yield comparable results (Tab. 1, Fig. 3), since there is almost no highly absorbing ^{151}Eu in the sample and thus b_{coh}'' is considered to be negligible. The results on the data collected at λ = 136 pm are very similar (not shown here).

Data collected on D2B instrument lead to comparable results as shown in Tab. 1, for both Li and Eu samples, but have lower precision due to lower counting rates and more challenging reflex profiles.

Literature

[1] L. Koester, H. Rauch, E. Seymann, *At. Data Nucl. Data Tables* **1991**, *49*, 65–120
[2] V. F. Sears, *Neutron News* **1992**, *3*, 26–37
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[4] B. H. Toby, R. B. von Dreele, *J. Appl. Crystallogr.* **2013,** *46*, 544–549
[5] H. Kohlmann, C. Hein, R. Kautenburger, T. C. Hansen, C. Ritter, S. Doyle, *Z. Kristallogr.* **2016**, *231*, 517–523