Experimental report

19/09/2024

Proposal: 5-31-2991 **Council:** 10/2022

Title: The impact of temperature factors on the determination of bound coherent neutron scattering lengths using Bragg powder diffraction

Research area: Methods and instrumentation

This proposal is a new proposal

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Samples: 7LiF
141PrN
141PrCl3
141PrOCl

Instrument	Requested days	Allocated days	From	To
D1B	2	0		
D20	2	2	21/06/2023	23/06/2023

Abstract:

The bound coherent neutron scattering length, bcoh(j), is one of the most basic properties of a nucleus 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.
For their determination Bragg powder diffraction is a robust method with low susceptibility to systematic errors and a rather good precision of about 1%. But atomic displacement parameters are known to be correlated with the determination bcoh(j) in Rietveld refinements. Thus, the aim of this proposal is to ivestigate the incluence of temperaturfactors in a systematic way investigating selected compounds of different crystal structures at different temperatures.

Experimental report for 5-31-2991

The impact of temperature factors on the determination of bound coherent neutron scattering lengths using Bragg powder diffraction

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Objectives

The bound coherent scattering length $b_c(j)$ of a nucleus *j* may be determined using neutron powder diffraction (Bragg diffraction) in combination with Rietveld refinement. A typical source of uncertainty in these determinations is the correlation between different refined parameters. In particular the correlation between thermal displacement and occupancy factors, representing the scattering length in Rietveld refinement, is prone to have a significant impact. Therefore, in this experiment data for samples of different composition and crystal structures were recorded at different temperatures and different *Q* space ranges to explore the actual impact of different thermal displacement factors on refined scattering lengths.

This experiment, together with the previous experiments 1-10-46 and 1-10-48, 1-10-50 is part of the ILL PhD project entitled *"Determination of accurate bound coherent neutron scattering lengths, b_c, of lanthanide, cadmium, thallium and lithium isotopes"*. It aims at a systematic investigation of neutron scattering lengths lacking accuracy by taking advantage of neutron powder diffraction as primary determination method.

Experimental Details

$^{165}Ho_2O_3$, ^{165}HoN, $^{165}HoCl_3$, $^{141}PrCl_3$ and ^{7}LiF were measured on the high-intensity diffractometer D20 using wavelengths of 106, 136 and 187 pm at temperatures of 10, 100, 300 (not for ^{7}LiF) and 400 K. Samples enclosed in cylindric vanadium cans were mounted in a helium cooled cryo-furnace with neutron transparent aluminum windows. For each measurement the sample temperature was equilibrated to set temperature and then powder pattern of each wavelength were recorded for about 30 min.

Key results

Rietveld refinement was performed based on neutron powder diffraction data of the samples with *FULLPROF*[1] and *GSAS-II*[2], using appropriate crystal structure models and a Thompson-Cox-Hastings[3] like profile function. All parameters (i.e. scale, cell, shape, asymmetry, background, position and thermal displacement factors) were refined simultaneously. The background was fitted with a twelfth-degree Chebyshev polynomial function. Since scattering lengths $b_c(j)$ could not be refined directly in either software, the $b_c(j)$ values were obtained via refinement of the site occupation factors for the isotope of investigation and multiplying the results with the corresponding programs internal b_c values. For the non-cubic compounds $PrCl_3$ and $HoCl_3$ temperature factor anisotropic thermal displacement factors were refined.

^{7}LiF: A slight, but not significant trend towards lower b_c values for higher *B* values in the wavelength averaged mean is seen (figure 1). 136 pm data tend to produce slightly lower b_c values than the other wavelengths used, independent from the temperature. Former measurements on samples at ambient conditions without furnace environment and radial collimator result in a value of $b_c(^{7}Li)$ = -2,28(2) fm.

$^{141}PrCl_3$: The refined scattering length is independent from temperature (figure 2). Here, 136 pm data tend to produce slightly higher b_c values, independent from the temperature. Former measurements on samples at ambient conditions without furnace environment and radial collimator result in a value of $b_c(^{7}Li)$ = 4,44(4) fm. The slight differences with wavelengths may be attributed to imperfections in the radial collimator producing higher absorption at random angles.

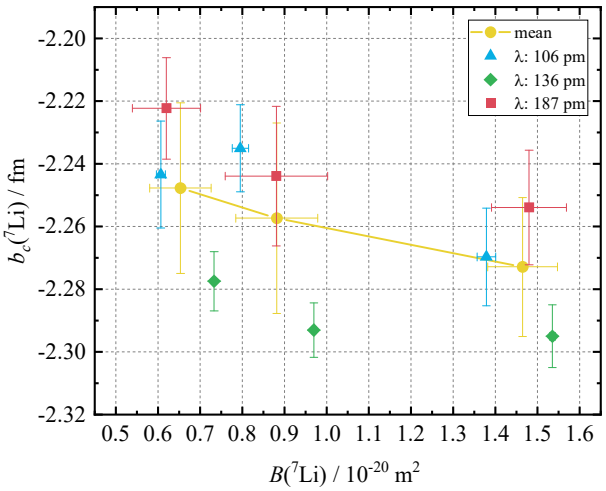

Figure 1: Refined b_c values for ^{7}Li plotted against the refined temperature factor B for the ^{7}Li crystallographic site in ^{7}LiF. Values from different refinement programs have been averaged.

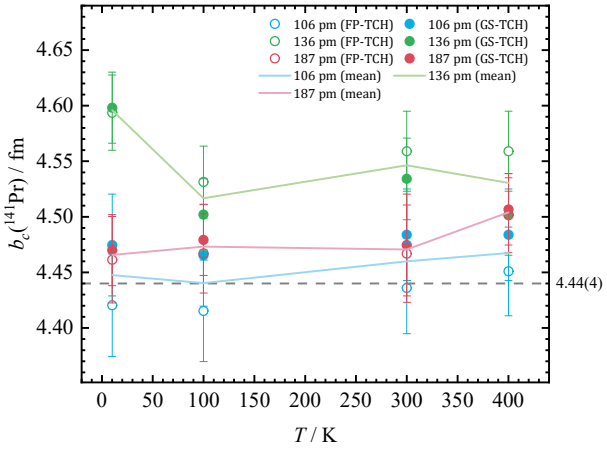

Figure 2: Refined b_c values for ^{141}Pr plotted against the sample temperature during the measurements of ^{141}PrCl$_3$. Dashed line indicates the results from former results for ^{141}Pr obtained at ambient conditions.

^{165}Ho$_2$O$_3$: There may be a slight, but not significant trend towards higher b_c values with higher temperature factors of Ho$_1$ position (figure 3). Figure 4 in addition shows a separation of values more with respect to the neutron wavelength, than to the sample temperature. The overall scattering of values is slightly higher than for Li and Pr results.

^{165}HoCl$_3$: Refined b_c values are independent of temperature, but are lower than for Ho$_2$O$_3$. At 100 K and 10 K a second phase appears which can be described using a high-pressure modification model [4] (figure 5). This modification was not yet described as a low temperature phase of HoCl$_3$.

^{165}HoN: The similarity of $b_c(^{165}\mathrm{Ho})$ and $b_c(^{nat}\mathrm{N})$ in combination with commutative crystal structure leads to non-reliable results in refinements. However, the compound was found to show ferromagnetic ordering as described in literature [5].

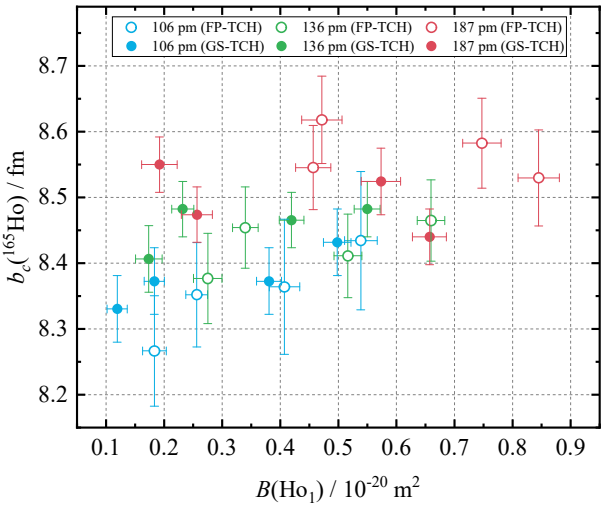

Figure 3: Refined b_c values for ^{165}Ho plotted against the refined temperature factor B for the Ho$_1$ crystallographic site in ^{165}Ho$_2$O$_3$. FP-TCH and GS-TCH represent the results obtained by the two refinement programs used.

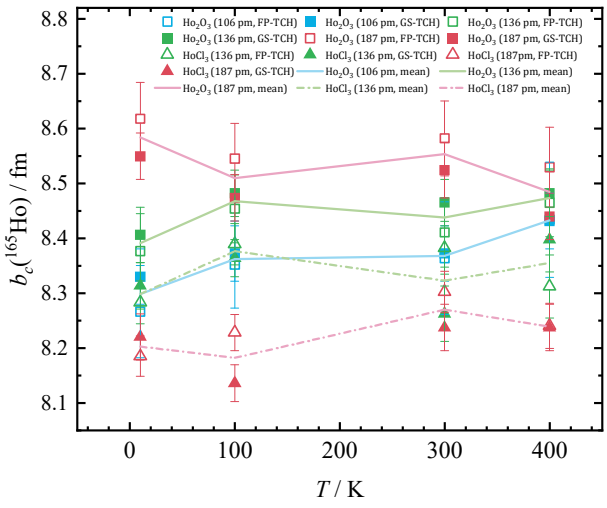

Figure 4: Refined b_c values for ^{165}Ho plotted against the sample temperature during the measurements of ^{165}HoCl$_3$ and ^{165}Ho$_2$O$_3$. FP-TCH and GS-TCH represent the results obtained by the two refinement programs used.

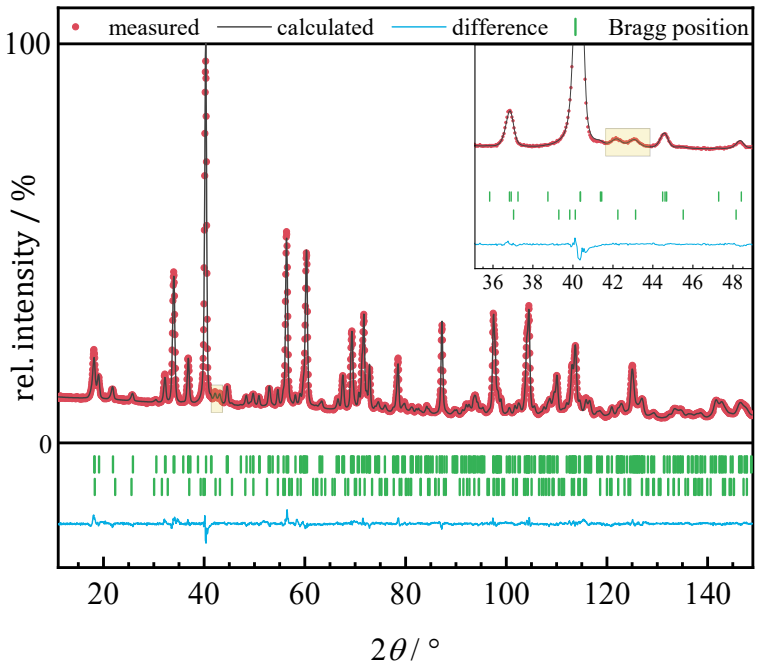

Figure 5: Rietveld refinement (FP-TCH) of the crystal structure of $^{165}HoCl_3$, based on neutron diffraction data collected on D20 at T = 10 K and $\lambda \approx 187$ pm. The enlarged section shows the reflections of the presumed low-temperature phase of $HoCl_3$, which can be modelled by an orthorhombic high-pressure modification (*Cmcm*). Bragg positions from top to bottom: $HoCl_3$ (*C*2/*m*), $HoCl_3$ (*Cmcm*).

Conclusion

Temperature does not have a significant impact on the determination bound coherent neutron scattering lengths. The correlation between the neutron scattering length (represented by the occupancy factor) and the thermal displacement parameter is rather affecting the values uncertainties than the values themself.

Literature

[1] J. Rodríguez-Carvajal, *Physica B* **1993**, *192*, 55–69. **DOI:** 10.1016/0921-4526(93)90108-I.

[2] B. H. Toby, R. B. von Dreele, *J. Appl. Crystallogr.* **2013**, *46*, 544–549. **DOI:** 10.1107/S0021889813003531.

[3] P. Thompson, D. E. Cox, J. B. Hastings, *J. Appl. Crystallogr.* **1987**, *20*, 79–83. **DOI:** 10.1107/S0021889887087090.

[4] H. P. Beck, E. Gladrow, *Z. Anorg. Allg. Chem.* **1983**, *502*, 178–184. **DOI:** 10.1002/zaac.19835020724.

[5] H. R. Child, M. K. Wilkinson, J. W. Cable, W. C. Koehler, E. O. Wollan, *Phys. Rev.* **1963**, *131*, 922–931. **DOI:** 10.1103/PhysRev.131.922.