Experiment Report

Proposal ID and Title: ES-1753 "Dehydration of lawsonite in cold subduction zones traced by experimental Quantum Crystallography methods"

Beamline	Scheduled Shifts	Session Start Date	Session End Date
ID27	9	21/11/2025	24/11/2025
Data DOI Link			
Local Contact(s)	MEZOUAR MOHAMED, PAKHOMOVA ANNA		

User Name	Attendance Mode
Ph.D. GAJDA, Roman	On-site
HUC, AGNIESZKA	On-site
Dr REJNHARDT, Piotr	On-site
Dr SLAWINSKI, Wojciech	On-site
Prof. WOZNIAK, Krzysztof	On-site

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Postal Address: ESRF - The European Synchrotron - User Office - CS 40220 - 38043 Grenoble Cedex 9 - France
Location : 71, avenue des Martyrs, 38043 Grenoble, France
Telephone: +33 (0)4 76 88 20 00

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The main aim of the project was to conduct a high-resolution, high-completeness single-crystal X-ray diffraction experiment to apply quantum crystallography methods and observe, in detail, complex rearrangements of water molecules and hydroxyl groups under high-pressure conditions. The second aim was to follow the detailed dehydration mechanism of lawsonite under cold subduction-zone conditions. Two measurements were made during the beamtime. The first one was carried out at high-pressure and room-temperature conditions in the range from 1.23 GPa to 16.08 GPa. For that experiment, we placed five differently oriented crystals of lawsonite and measured them separately to increase the completeness of the collected data. During the data collection, we were able to measure 10 pressure points for the first, orthorhombic phase, and 8 pressure points for the second, monoclinic phase. The completeness of the collected data was extremely high, and merging the 4 of 5 crystal data, we were able to reach 100% completeness for the orthorhombic phase at high-pressure conditions up to a resolution of 0.38 Å with very high redundancy (Table 1). After the phase transition, only 2 out of 5 crystals survived; however, merging both data, we were also able to reach very high completeness for a monoclinic system, around 70% up to a resolution of 0.5 Å (Table 2). The quality of the obtained data for both phases is extremely high and sufficient for applying quantum crystallography methods. In result, that experiment was fully successful.

Table 1. Statistics vs. resolution, along with redundancy for the data collected at 5.04 GPa.

Statistics vs resolution (taking redundancy into account) - point group symmetry: Cmmm

resolution (Å)	# kept	# theory	# unique	% complete	average redundancy	mean F2	mean F2/sig(F2)	Rint	RsigmaB
inf-0.85	2164	349	343	98.3	6.3	527681.11	167.42	0.044	0.004
0.85-0.66	2404	343	343	100.0	7.0	190910.91	85.34	0.047	0.008
0.66-0.58	2317	343	343	100.0	6.8	108390.29	57.32	0.052	0.012
0.58-0.52	2280	343	343	100.0	6.6	77869.32	46.21	0.056	0.015
0.52-0.48	2257	343	343	100.0	6.6	45597.13	33.38	0.060	0.021
0.48-0.45	2132	343	343	100.0	6.2	35800.76	27.56	0.064	0.026
0.45-0.43	2117	343	343	100.0	6.2	27634.79	22.79	0.073	0.030
0.43-0.41	1702	343	343	100.0	5.0	20156.36	17.58	0.070	0.041
0.41-0.39	1456	344	343	99.7	4.2	16180.96	14.06	0.077	0.052
0.39-0.38	1089	354	344	97.2	3.2	13210.51	11.84	0.079	0.067
inf-0.38	19918	3448	3431	99.5	5.8	117458.02	52.78	0.049	0.009

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Table 2. Statistics vs. resolution, along with redundancy for the data collected at 16.08 GPa.

Statistics vs resolution (taking redundancy into account) - point group symmetry: P2/m (b-unique)

resolu-tion(Å)	# kept	# theory	# unique	% complete	average redundancy	mean F2	mean F2/sig(F2)	Rint	RsigmaB
inf-1.10	355	265	186	70.2	1.9	428131.95	64.98	0.018	0.010
1.09-0.87	340	250	186	74.4	1.8	153498.02	31.97	0.032	0.019
0.87-0.76	357	258	186	72.1	1.9	136826.69	29.46	0.043	0.024
0.76-0.69	367	258	186	72.1	2.0	78312.70	20.60	0.046	0.032
0.69-0.63	353	268	186	69.4	1.9	54475.03	15.58	0.060	0.045
0.63-0.60	354	276	186	67.4	1.9	46018.17	14.43	0.075	0.053
0.60-0.56	341	275	186	67.6	1.8	41221.83	12.70	0.064	0.056
0.56-0.54	342	249	186	74.7	1.8	31689.52	10.80	0.086	0.075
0.54-0.52	316	259	186	71.8	1.7	32168.92	10.90	0.069	0.066
0.52-0.50	343	271	190	70.1	1.8	20906.97	8.32	0.131	0.093
inf-0.50	3468	2630	1864	70.9	1.9	103666.41	22.19	0.038	0.025

The second experiment was carried out at both high-pressure and high-temperature conditions. We used an external heating device (vacuum chamber) to have a higher diamond anvil cell opening angle. The experiment was partially successful since we were not able to reach the dehydration process for lawsonite. The maximal temperature, which we obtained, was 400 ˚C, because at 500 ˚C we melted Kapton windows from the vacuum chamber, and we had to stop the whole experiment. Nevertheless, the completeness of the data at 5 GPa and 400 ˚C is sufficiently high for quantum crystallography approaches, and as a result, we will obtain the first experimental charge density redistribution for the compound when both external stimuli (temperature and pressure) are applied.

Our preliminary results using Hirshfeld Atom Refinement (HAR) show that at ambient conditions, the hydrogen atom from the hydroxyl group is disordered into two separate positions with a half occupancy. However, above 4 GPa, the ordering of that hydrogen atom occurs, which probably has a crucial impact on the bulk modulus of lawsonite and seismic velocities at cold subduction zone environment (Fig. 1). The phase transition for lawsonite around 4 GPa was previously predicted by X-ray diffraction[1] and electrical conductivity[2] experiments, but it was never explained from the structural point of view.

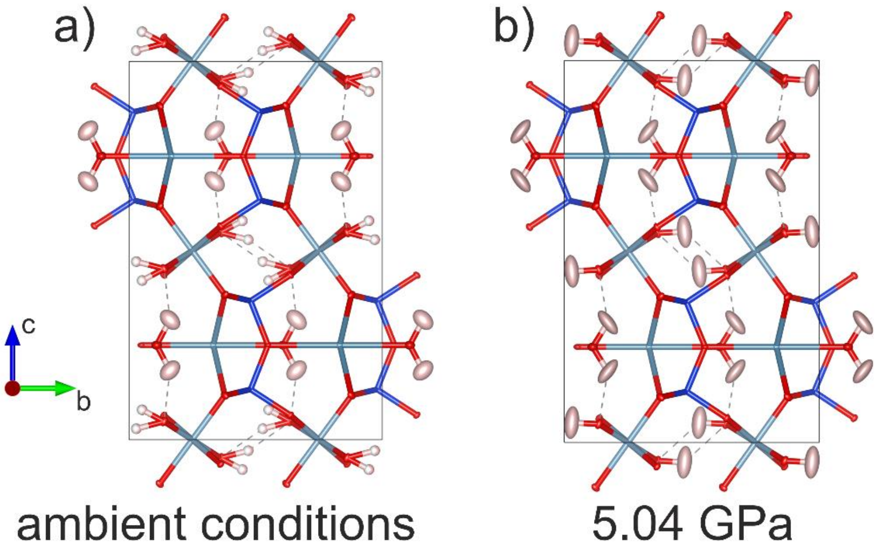

Figure 1. Crystal structure of lawsonite along *a* axis at ambient conditions (a) and at 5.04 GPa (b). For the structure at ambient conditions, the hydrogen atom from the hydroxyl group is disordered and at 5.04 GPa the same hydrogen atom is fully ordered.

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1. Ballaran, T. B. & Angel, R. J. Equation of state and high-pressure phase transitions in lawsonite. *Eur. J. Mineral.* **15**, 241–246 (2003).
2. Pommier, A., Williams, Q., Evans, R. L., Pal, I. & Zhang, Z. Electrical Investigation of Natural Lawsonite and Application to Subduction Contexts. *J. Geophys. Res. Solid Earth* **124**, 1430–1442 (2019).

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