

Table S1 Unit cell parameters refined from neutron diffraction data (in bold the more accurate data collections that were used to refine structural parameters). Space group is P2₁/n over all pressure range.

a(Å)	b(Å)	c(Å)	β(°)	V(Å ³)	P (GPa)	R _{wp} (%)
6.141(1)	3.6042(6)	12.086(3)	106.53(2)	256.4(1)	0.0001	3.503
6.127(3)	3.550(1)	12.051(7)	106.47(3)	251.4(2)	0.2	7.150
6.116(3)	3.497(1)	12.015(7)	106.59(4)	246.3(2)	0.4	7.133
6.084(1)	3.4189(4)	11.960(2)	106.56(1)	238.45(7)	1.0	2.925
6.0253(8)	3.3256(3)	11.875(1)	106.524(7)	228.12(4)	1.8	1.611
5.979(2)	3.2790(6)	11.817(3)	106.40(2)	222.2(1)	2.4	4.066
5.922(2)	3.223(1)	11.746(5)	106.29(2)	215.2(1)	3.2	5.388
5.856(2)	3.1821(7)	11.712(4)	106.18(2)	209.6(1)	4.0	5.021
5.8357(7)	3.1648(3)	11.681(1)	106.139(7)	207.23(4)	4.3	1.682
5.792(2)	3.1344(7)	11.656(4)	106.19(2)	203.2(1)	5.1	5.333
5.766(2)	3.1059(7)	11.629(4)	106.20(3)	200.0(1)	5.8	5.212
5.7533(8)	3.0920(3)	11.6211(2)	106.28(1)	198.44(4)	6.2	1.850
5.740(2)	3.0778(7)	11.615(4)	106.34(2)	196.9(1)	6.5	5.423
5.7240(2)	3.0678(8)	11.599(5)	106.36(2)	195.4(1)	6.8	5.308
5.712(1)	3.0560(3)	11.589(2)	106.391(9)	194.1(5)	7.2	1.846
5.699(3)	3.0434(8)	11.575(6)	106.39(3)	192.6(2)	7.6	4.276

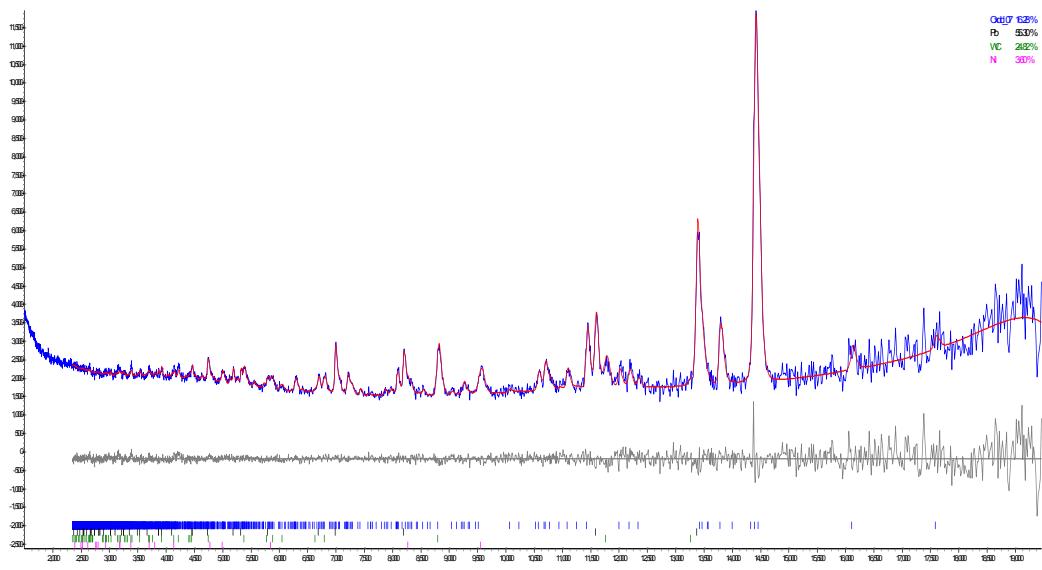


Figure S1 Observed and model refined neutron diffraction spectrum at 0.0001 GPa

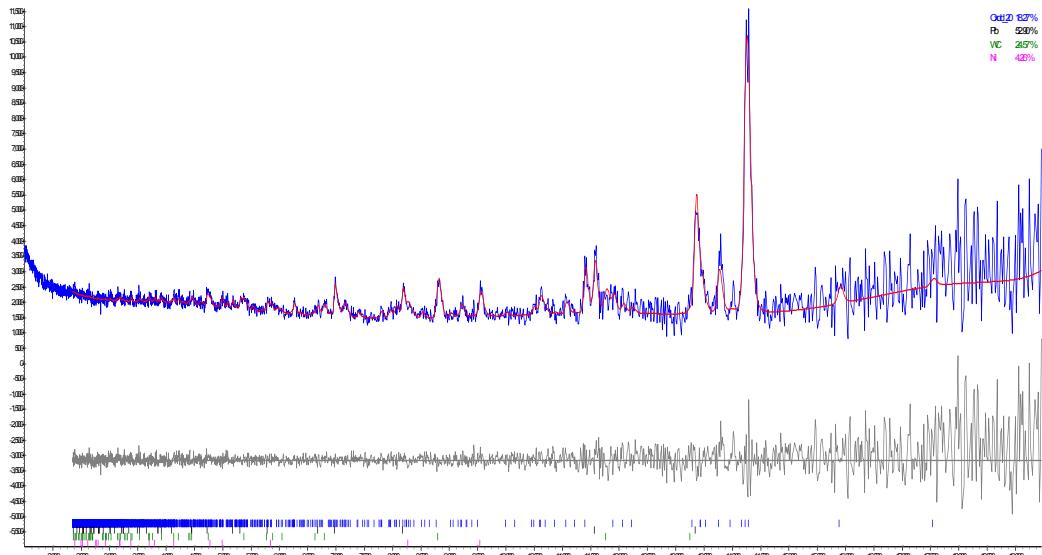


Figure S2 Observed and model refined neutron diffraction spectrum at 0.2 GPa

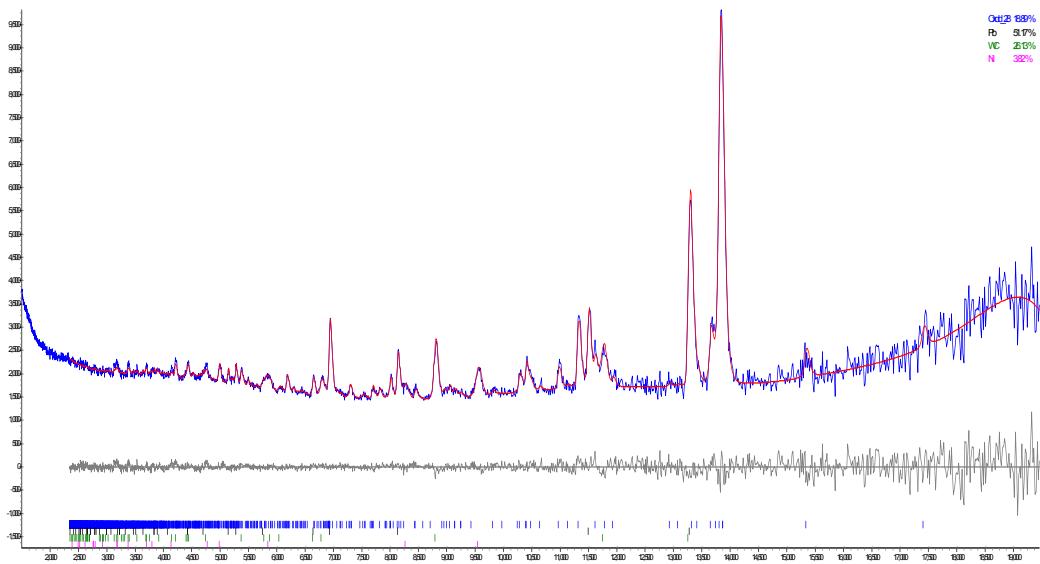


Figure S3 Observed and model refined neutron diffraction spectrum at 0.4 GPa

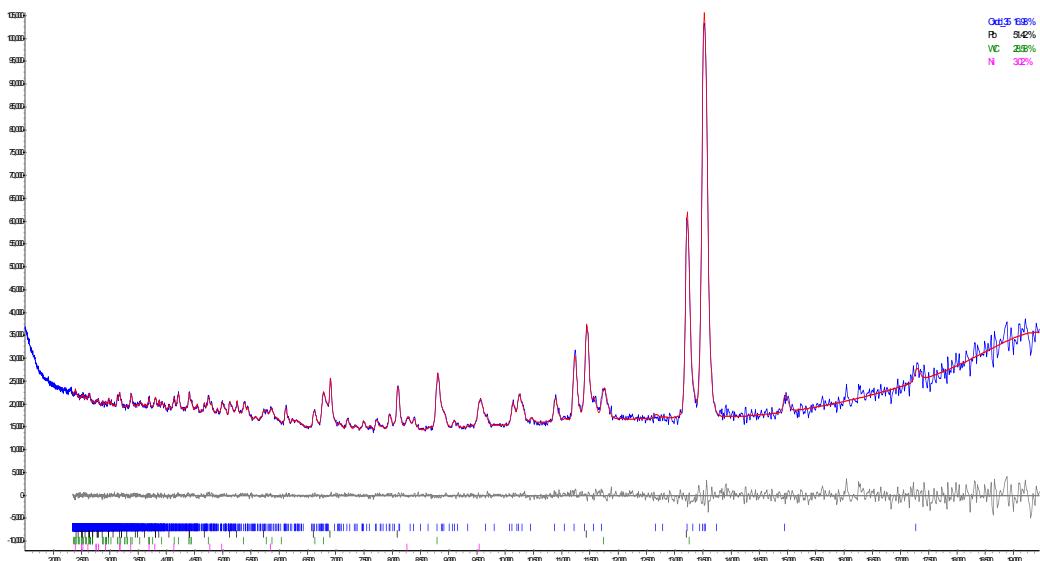


Figure S4 Observed and model refined neutron diffraction spectrum at 1.0 GPa

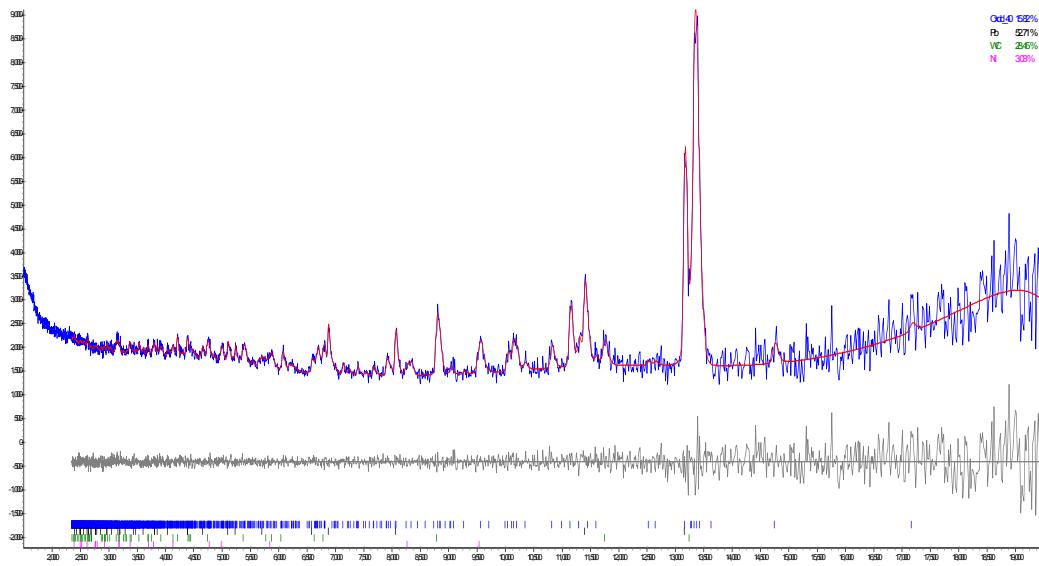


Figure S5 Observed and model refined neutron diffraction spectrum at 1.8 GPa

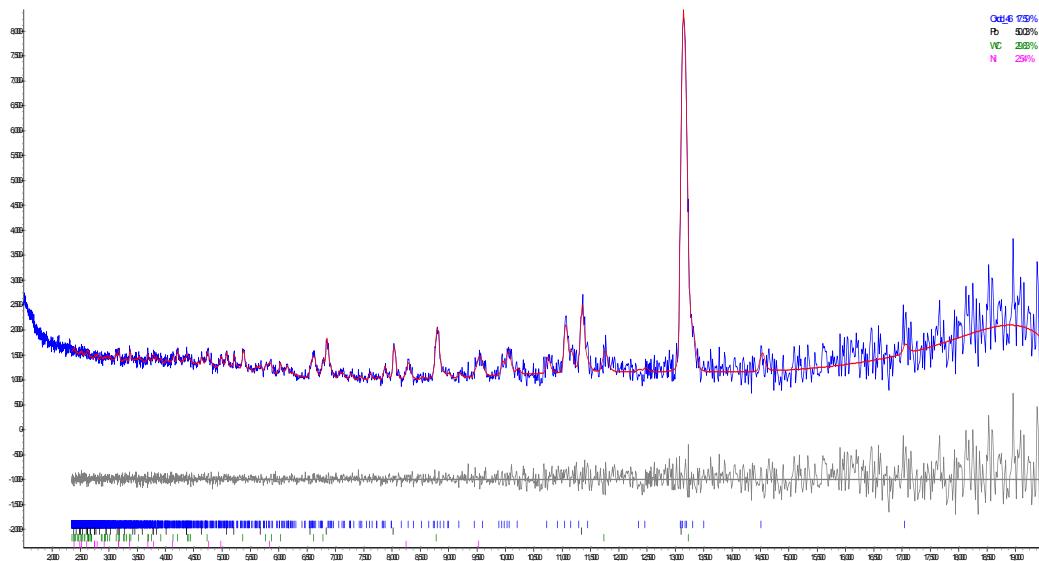


Figure S6 Observed and model refined neutron diffraction spectrum at 2.4 GPa

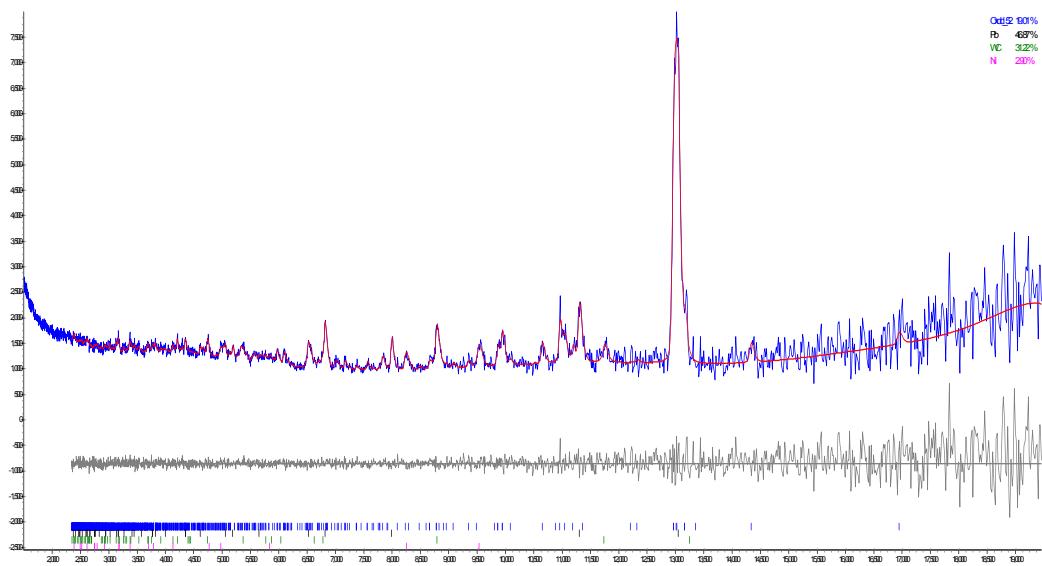


Figure S7 Observed and model refined neutron diffraction spectrum at 3.2 GPa

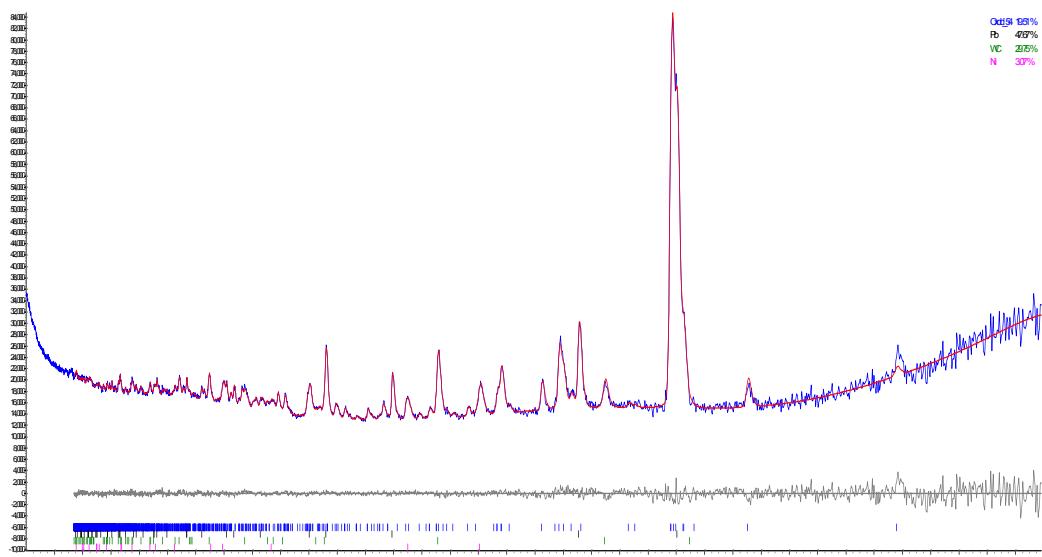


Figure S8 Observed and model refined neutron diffraction spectrum at 4.0 GPa

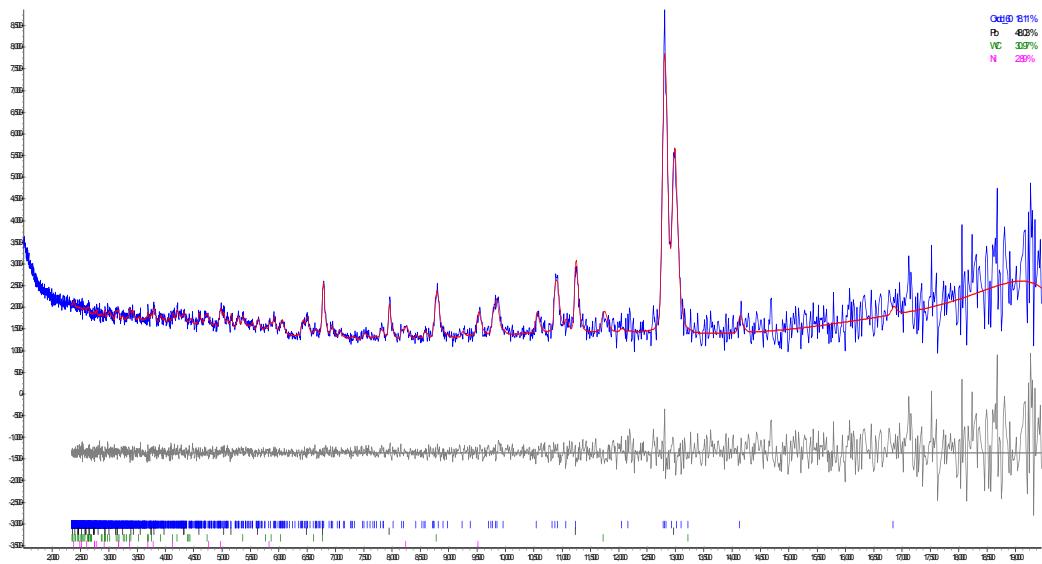


Figure S9 Observed and model refined neutron diffraction spectrum at 4.3 GPa

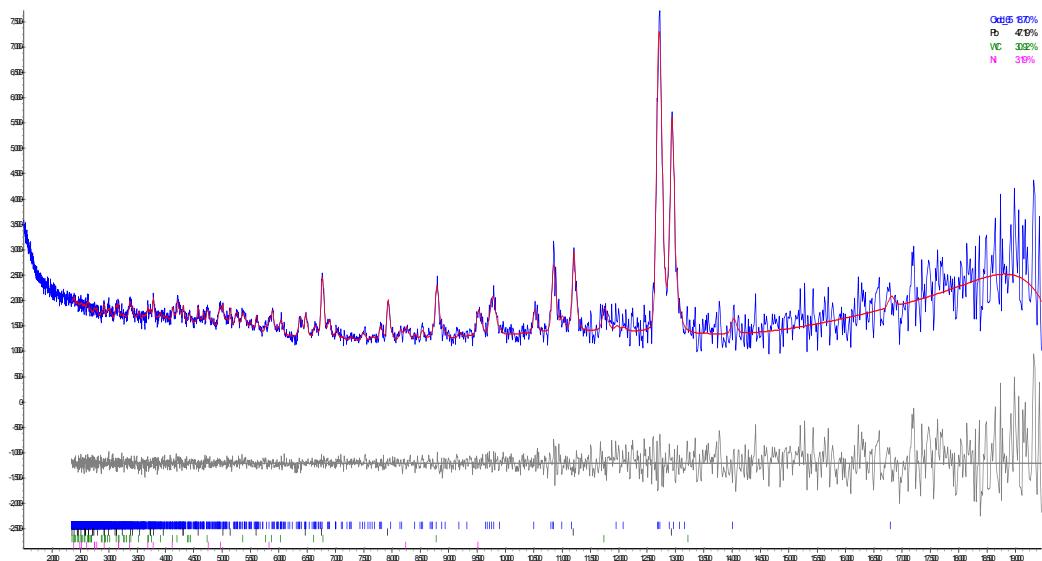


Figure S10 Observed and model refined neutron diffraction spectrum at 5.1 GPa

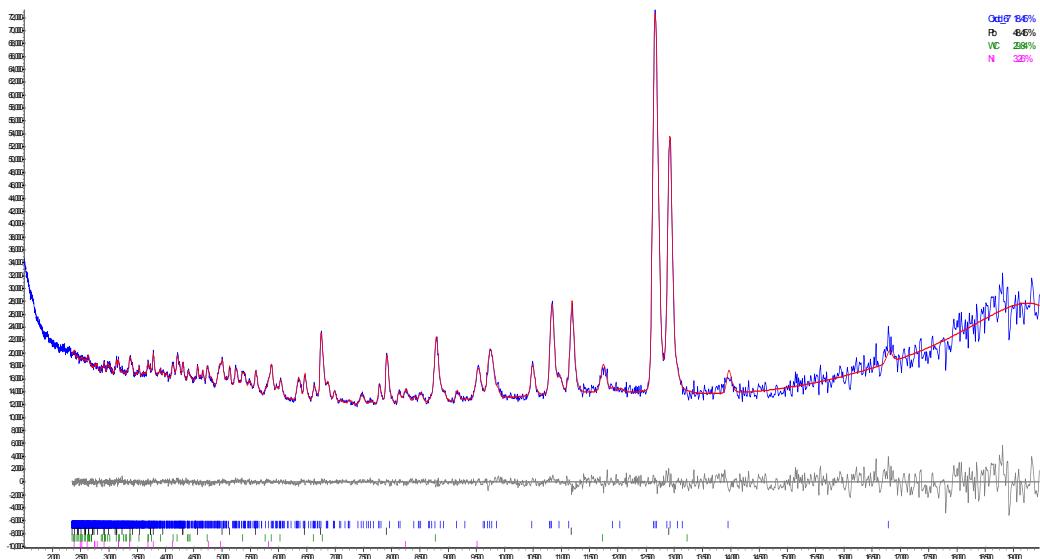


Figure S11 Observed and model refined neutron diffraction spectrum at 5.8 GPa

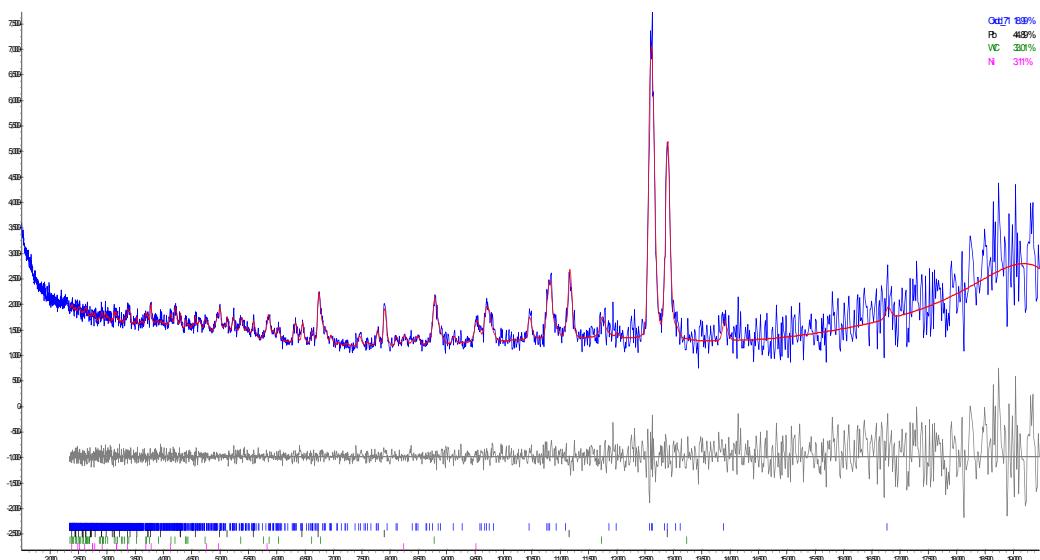


Figure S12 Observed and model refined neutron diffraction spectrum at 6.2 GPa

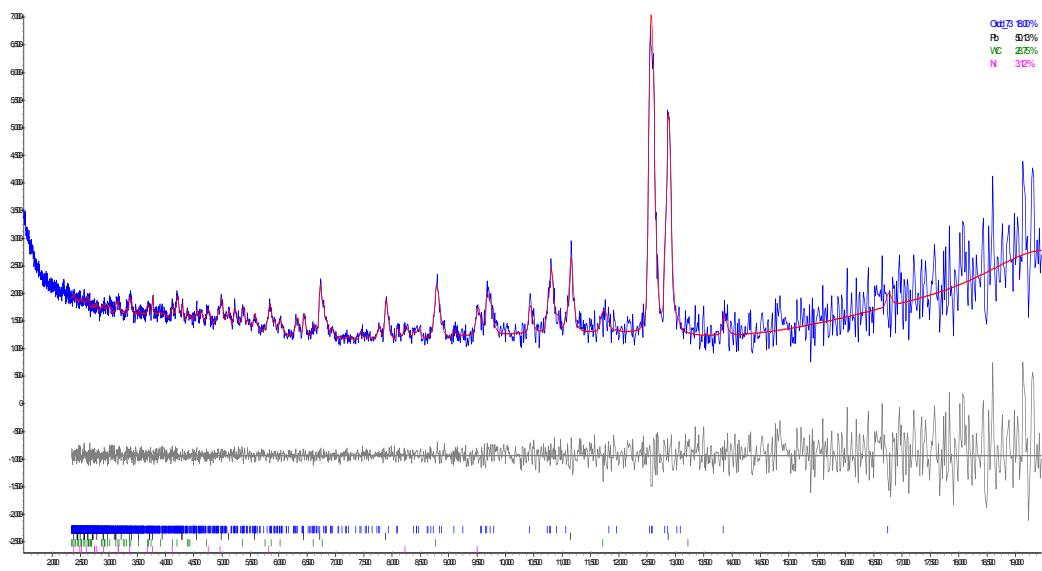


Figure S13 Observed and model refined neutron diffraction spectrum at 6.5 GPa

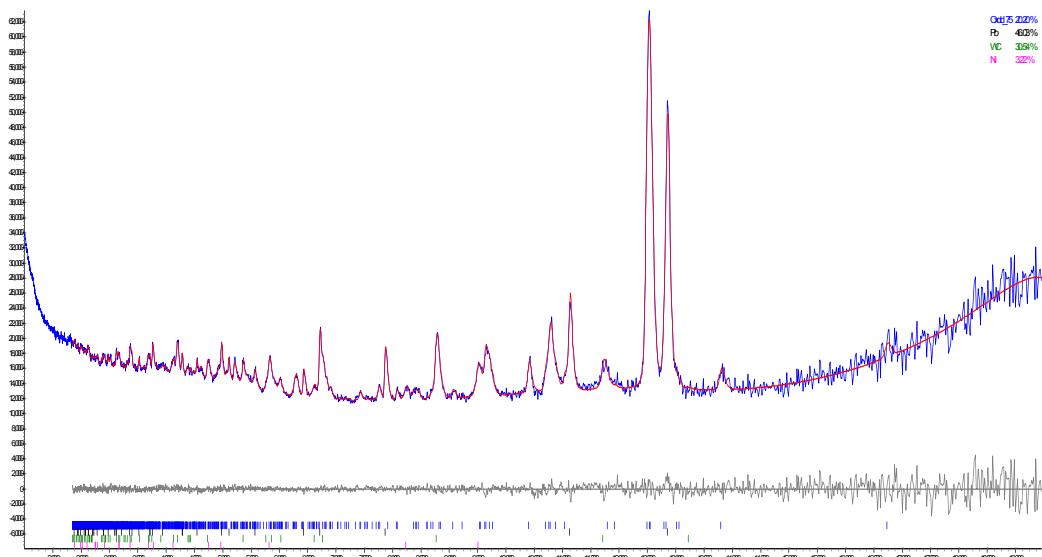


Figure S14 Observed and model refined neutron diffraction spectrum at 6.8 GPa

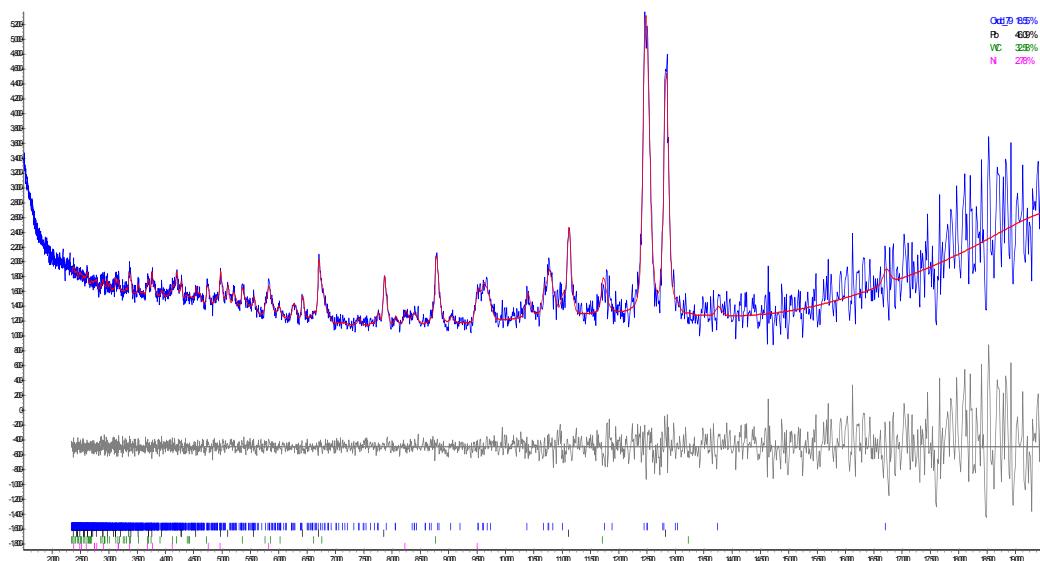


Figure S15 Observed and model refined neutron diffraction spectrum at 7.2 GPa

Table S2 Unit cell parameters of **1 β (N)** predicted from PDF calculations (0 K simulation, hydrogenated phase).

a(Å)	b(Å)	c(Å)	β (°)	V(Å ³)	P (GPa)
9.810	4.875	5.278	99.272	249.1	0
9.683	4.879	5.210	100.686	241.9	0.5
9.576	4.909	5.128	102.396	235.4	1
9.429	4.924	5.017	104.574	225.4	2
9.303	4.959	4.909	106.559	217.1	3
9.341	4.789	4.949	105.976	212.8	4
9.278	4.792	4.885	107.108	207.6	5
9.254	4.734	4.863	107.718	202.9	6
9.215	4.711	4.827	108.391	198.9	7
9.177	4.693	4.794	109.012	195.2	8
9.134	4.628	4.751	109.717	189.1	10

Table S3 Unit cell parameters of **1 β (CT)** predicted from PDF calculations (0 K simulation, hydrogenated phase).

a(Å)	b(Å)	c(Å)	β (°)	V(Å ³)	P (GPa)
10.998	5.567	3.884	87.269	237.5	0.5
10.982	5.591	3.804	87.884	233.4	1
10.410	5.490	3.964	95.053	225.7	2
9.861	5.296	4.197	100.990	215.1	3
9.605	5.189	4.289	103.152	208.2	4
9.450	5.135	4.321	104.394	203.1	5
9.296	5.088	4.358	105.460	198.6	6
9.215	4.711	4.827	108.391	198.9	7
9.177	4.693	4.794	109.012	195.2	8
9.026	4.966	4.365	107.676	186.4	10

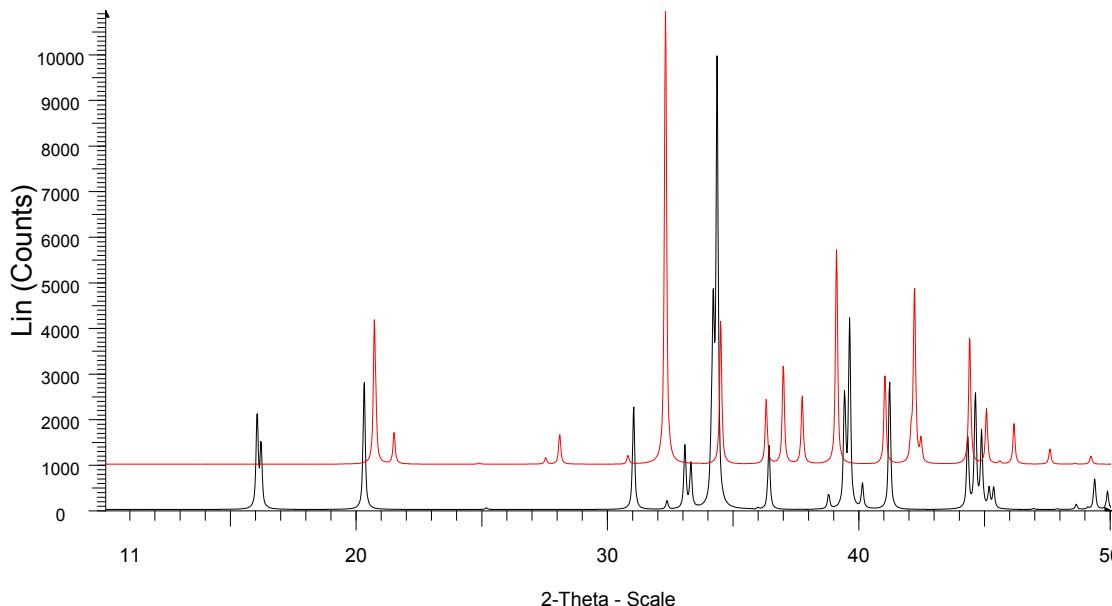


Figure S16 Simulated powder pattern, CuK α radiation, for the CT configuration of **1 α** (black) and **1 β** (red) polymorphs at ca. 10 GPa.

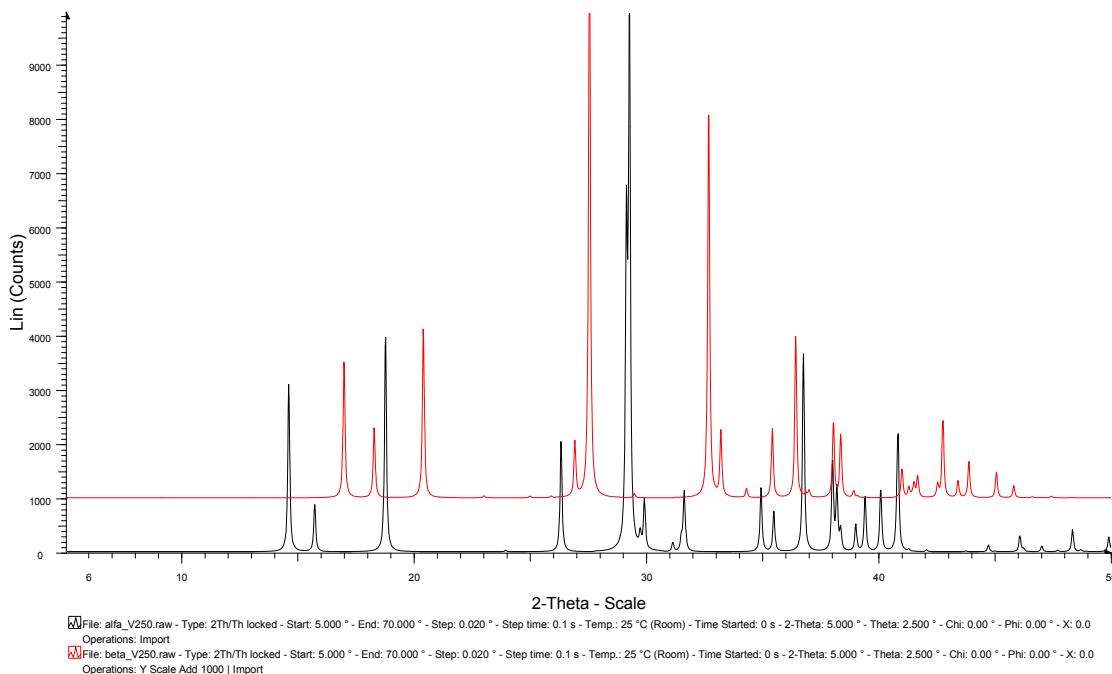


Figure S17 Simulated powder pattern, CuK α radiation, for the N configuration of **1 α** (black) and **1 β** (red) polymorphs at room conditions.