Table S1. Hydrogen-bond and intermolecular contact interaction geometry.

D-H···A	D-H distance (Å)	H···A distance (Å)	D···A distance (Å)	D-H···A angle (°)
O3A – H1···O7A	0.80(2)	3.00(7)	3.475(5)	119(7)
O3B – H2···O7B	0.81(2)	2.43(7)	3.127(4)	143(10)

Table S2. Refined fractional atomic coordinates and isotropic displacement parameters for gedrite at selected pressures.

Atom	x	y	z	Site occupancy	U_{eq}
Gedrite at Ambient Pressure, *Pnma*					
a = 18.5838(5) Å, *b* = 17.8286(4) Å, *c* = 5.2780(1) Å					
Na	0.1152(4)	-0.25	0.841(1)	0.393(1)	0.030(1)
Mg1/Fe1	0.12430(4)	0.16089(5)	0.3744(1)	0.771(4)/0.228(4)	0.0075(2)
Al2/Mg2/Fe2	0.12465(5)	0.07248(5)	-0.1259(2)	0.541(4)/0.401(5)/0.055(4)	0.0067(2)
Mg3/Fe3	0.12430(6)	0.25	-0.1257(2)	0.747(6)/0.252(6)	0.0068(3)
Mg4/Fe4	0.11919(3)	-0.01449(3)	0.3693(1)	0.287(4)/0.712(4)	0.0111(1)
Si1A/Al1A	0.23175(4)	-0.16320(5)	-0.4475(1)	0.74(7)/0.25(7)	0.0055(3)
Si1B/Al1B	0.01989(4)	-0.16449(5)	0.2960(1)	0.60(7)/0.39(7)	0.0065(3)
Si2A	0.22784(4)	-0.07998(5)	-0.2003(1)	1	0.0059(1)
Si2B/Al2B	0.02638(4)	-0.07998(5)	-0.2003(1)	0.90(7)/0.09(7)	0.0072(3)
O1A	0.1795(1)	0.1592(1)	0.0369(4)	1	0.0107(3)
O1B	0.0695(1)	0.1584(1)	-0.2854(4)	1	0.0103(3)
O2A	0.1844(1)	0.0745(1)	-0.4398(4)	1	0.0091(3)
O2B	0.0629(1)	0.0745(1)	0.1902(4)	1	0.0111(3)
O3A	0.1806(1)	0.25	-0.4596(6)	1	0.0094(4)
O3B	0.0701(1)	0.25	0.2135(6)	1	0.0092(4)
O4A	0.1862(1)	0.0022(1)	0.0501(4)	1	0.0118(3)
O4B	0.0680(1)	-0.0045(1)	-0.2959(4)	1	0.0115(3)
O5A	0.1970(1)	-0.1113(1)	0.3226(4)	1	0.0132(4)
O5B	0.0541(1)	-0.1025(1)	0.0907(5)	1	0.0160(4)
O6A	0.2026(1)	-0.1315(1)	-0.1747(4)	1	0.0124(3)
O6B	0.0477(1)	-0.1444(1)	-0.4132(5)	1	0.0192(5)
O7A	0.2037(1)	-0.25	0.5219(7)	1	0.0140(5)
O7B	0.0452(1)	-0.25	0.2186(7)	1	0.0125(5)
H1	0.224(1)	0.25	-0.46(1)	1	0.02(2)
H2	0.028(2)	0.25	0.16(2)	1	0.06(3)
Gedrite at 10.6(5) GPa, *Pnma*					
a = 17.823 (3) Å, *b* = 17.427(1) Å, *c* = 5.1598(1) Å					
Na	0.123(4)	-0.25	0.851(7)	0.393(1)	0.023(9)
Mg1/Fe1	0.1240(6)	0.1631(3)	0.364(1)	0.771(4)/0.228(4)	0.006(1)
Al2/Mg2/Fe2	0.1236(7)	0.0754(4)	-0.135(1)	0.541(4)/0.401(5)/0.055(4)	0.009(1)
Mg3/Fe3	0.1234(9)	0.25	-0.134(1)	0.747(6)/0.252(6)	0.008(1)
Mg4/Fe4	0.1171(4)	-0.0122(2)	0.3598(7)	0.287(4)/0.712(4)	0.0084(8)
Si1A/Al1A	0.2295(8)	-0.1638(4)	-0.451(1)	0.74(7)/0.25(7)	0.007(1)
Si1B/Al1B	0.0263(7)	-0.1655(4)	0.305(1)	0.60(7)/0.39(7)	0.005(1)
Si2A	0.2274(8)	-0.0752(4)	0.044(1)	1	0.010(1)
Si2B/Al2B	0.0288(7)	-0.0810(4)	-0.194(1)	0.90(7)/0.09(7)	0.009(1)
O1A	0.180(1)	0.1622(9)	0.025(2)	1	0.007(3)
O1B	0.065(1)	0.158(1)	-0.295(3)	1	0.011(4)

O2A	0.183(1)	0.0769(9)	-0.452(3)	1	0.010(3)
O2B	0.061(1)	0.077(1)	0.179(3)	1	0.013(4)
O3A	0.185(2)	0.25	-0.473(4)	1	0.009(5)
O3B	0.064(2)	0.25	0.197(4)	1	0.010(6)
O4A	0.191(1)	0.007(1)	0.038(3)	1	0.011(4)
O4B	0.066(1)	-0.002(1)	-0.299(3)	1	0.018(5)
O5A	0.193(1)	-0.108(1)	0.317(3)	1	0.012(4)
O5B	0.058(1)	-0.0967(9)	0.101(2)	1	0.005(3)
O6A	0.197(1)	-0.137(1)	-0.167(3)	1	0.013(4)
O6B	0.054(1)	-0.150(1)	-0.391(3)	1	0.023(5)
O7A	0.191(2)	-0.25	0.494(4)	1	0.011(6)
O7B	0.055(2)	-0.25	0.203(4)	1	0.006(5)
β-Gedrite at 21(1) GPa, $P2_1/m$					
a = 17.514 (3) Å, b = 17.077(1) Å, c = 4.9907(2) Å, β = 82.882(6)°					
Na	0.381(4)	0.25	0.360(7)	0.393(1)	0.02(1)
Na'	0.870(3)	0.25	0.107(5)	0.393(1)	0.001(5)
Mg1/Fe1	0.1251(6)	0.1635(3)	0.368(1)	0.771(4)/0.228(4)	0.004(1)
Mg1'/Fe1'	0.3759(6)	0.8351(3)	0.8272(9)	0.771(4)/0.228(4)	0.006(1)
Al2/Mg2/Fe2	0.1260(7)	0.0755(4)	0.871(1)	0.541(4)/0.401(5)/0.055(4)	0.007(1)
Al2'/Mg2'/Fe2'	0.3757(8)	0.9236(4)	0.327(1)	0.541(4)/0.401(5)/0.055(4)	0.011(1)
Mg3/Fe3	0.1249(8)	0.25	0.864(1)	0.747(6)/0.252(6)	0.003(1)
Mg3'/Fe3'	0.6214(9)	0.25	0.680(1)	0.747(6)/0.252(6)	0.010(1)
Mg4/Fe4	0.1234(4)	0.9844(2)	0.3774(6)	0.287(4)/0.712(4)	0.0066(7)
Mg4'/Fe4'	0.3864(4)	0.0077(2)	0.8355(6)	0.287(4)/0.712(4)	0.0051(8)
Si1A/Al1A	0.2246(8)	0.8345(4)	0.456(1)	0.74(7)/0.25(7)	0.009(1)
Si1A'/Al1A'	0.2752(7)	0.1645(4)	0.052(1)	0.74(7)/0.25(7)	0.006(1)
Si1B/Al1B	0.0251(7)	0.8342(4)	0.321(1)	0.60(7)/0.39(7)	0.009(1)
Si1B'/Al1B'	0.4703(7)	0.1670(4)	0.815(1)	0.60(7)/0.39(7)	0.007(1)
Si2A	0.2253(8)	0.9221(4)	-0.040(1)	1	0.010(1)
Si2A'	0.2762(7)	0.0778(4)	0.544(1)	1	0.008(1)
Si2B/Al2B	0.0283(7)	0.9197(4)	0.821(1)	0.90(7)/0.09(7)	0.006(1)
Si2B'/Al2B'	0.4684(8)	0.0817(4)	0.312(1)	0.90(7)/0.09(7)	0.008(1)
O1A	0.183(1)	0.1614(9)	0.032(3)	1	0.011(3)
O1A'	0.319(1)	0.8366(1)	0.475(3)	1	0.017(4)
O1B	0.068(1)	0.1599(9)	0.702(2)	1	0.007(3)
O1B'	0.434(1)	0.8391(8)	0.177(2)	1	0.008(3)
O2A	0.184(1)	0.0758(8)	0.562(2)	1	0.005(3)
O2A'	0.315(1)	0.9217(9)	-0.010(2)	1	0.008(3)
O2B	0.060(1)	0.076(1)	0.178(3)	1	0.018(4)
O2B'	0.437(1)	0.917(1)	0.660(3)	1	0.014(4)
O3A	0.192(2)	0.25	0.539(3)	1	0.003(4)
O3A'	0.681(2)	0.25	1.018(3)	1	0.008(4)
O3B	0.062(2)	0.25	0.192(4)	1	0.007(4)
O3B'	0.560(3)	0.25	0.326(4)	1	0.015(5)
O4A	0.192(1)	0.001(1)	0.063(3)	1	0.019(4)
O4A'	0.311(1)	0.996(1)	0.492(3)	1	0.012(4)
O4B	0.064(1)	0.9983(9)	0.702(2)	1	0.006(3)
O4B'	0.438(1)	-0.0002(9)	0.186(2)	1	0.009(3)
O5A	0.196(1)	0.848(1)	0.145(3)	1	0.020(4)
O5A'	0.310(1)	0.0994(9)	0.844(3)	1	0.010(3)
O5B	0.053(1)	0.9053(9)	0.136(2)	1	0.007(3)
O5B'	0.442(1)	0.0944(9)	0.613(3)	1	0.013(4)

O6A	0.194(1)	0.9040(9)	0.650(3)	1	0.009(3)
O6A'	0.303(1)	0.1481(8)	0.364(2)	1	0.005(3)
O6B	0.057(1)	0.844(1)	0.645(3)	1	0.016(4)
O6B'	0.441(1)	0.1556(9)	0.121(3)	1	0.008(3)
O7A	0.806(2)	0.25	0.452(3)	1	0.003(4)
O7A'	0.312(2)	0.25	0.963(4)	1	0.010(5)
O7B	0.944(3)	0.25	0.793(4)	1	0.017(6)
O7B'	0.438(3)	0.25	0.692(4)	1	0.021(6)

Table S3. Anisotropic displacement parameters for gedrite at ambient pressure.

Atom	**U_{11}**	**U_{22}**	**U_{33}**	**U_{23}**	**U_{13}**	**U_{12}**
Na	0.031(3)	0.023(3)	0.036(3)	0	0.021(3)	0
Mg1/Fe1	0.0080(3)	0.0082(3)	0.0063(3)	0.0009(2)	0.0009(2)	0.0002(2)
Al2/Mg2/Fe2	0.0063(3)	0.0068(3)	0.0070(3)	0.0006(2)	0.0004(2)	0.0001(2)
Mg3/Fe3	0.0076(4)	0.0061(4)	0.0066(4)	0	-0.0000(3)	0
Mg4/Fe4	0.0115(2)	0.0125(2)	0.0094(2)	0.0020(1)	0.0039(1)	0.0034(1)
Si1A/Al1A	0.0045(3)	0.0071(4)	0.0049(3)	-0.0005(2)	-0.004(2)	0.0007(2)
Si1B/Al1B	0.0046(3)	0.0070(4)	0.0078(4)	0.0009(2)	-0.0001(2)	-0.0005(2)
Si2A	0.0045(2)	0.0088(3)	0.0043(2)	-0.0012(2)	-0.0002(2)	0.0009(2)
Si2B/Al2B	0.0047(3)	0.0088(4)	0.0080(4)	0.0025(2)	0.0014(2)	-0.0002(2)
O1A	0.0062(7)	0.0155(9)	0.0104(8)	0.0030(7)	0.0024(6)	0.0023(6)
O1B	0.0075(7)	0.0171(9)	0.0062(7)	0.0018(7)	-0.0010(6)	-0.0025(6)
O2A	0.0051(7)	0.0131(8)	0.0091(7)	0.0012(7)	0.0001(6)	0.0008(6)
O2B	0.0062(7)	0.0111(8)	0.0161(9)	0.0051(7)	-0.0014(7)	0.0003(6)
O3A	0.007(1)	0.011(1)	0.009(1)	0	0.0013(9)	0
O3B	0.008(1)	0.010(1)	0.009(1)	0	-0.0005(9)	0
O4A	0.0090(8)	0.0109(8)	0.0154(9)	-0.0035(7)	-0.0003(7)	0.0026(6)
O4B	0.0112(8)	0.0147(9)	0.0085(8)	0.0035(7)	0.0017(7)	0.0008(7)
O5A	0.0077(8)	0.023(1)	0.0083(8)	0.0041(8)	0.0008(6)	0.0000(7)
O5B	0.0081(8)	0.019(1)	0.020(1)	0.0105(9)	0.0015(7)	0.0014(7)
O6A	0.0082(7)	0.018(1)	0.0104(8)	-0.0085(7)	0.003(6)	-0.0008(7)
O6B	0.0115(9)	0.021(1)	0.025(1)	-0.011(1)	-0.0045(9)	0.0046(8)
O7A	0.011(1)	0.010(1)	0.020(1)	0	0.000(1)	0
O7B	0.011(1)	0.011(1)	0.014(1)	0	-0.000(1)	0