Supporting Information for "Structural characterization of water ice condensed on silicate grains by infrared and Raman spectroscopy"

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Table S1 – Comparison of converged cell parameters with experimental and theoretical literature values for the forsterite crystal.

Author	Source[a]	a / Å	Error / %	b / Å	Error / %	c / Å	Error / %	Temperature /K
This study	Calculated, PBE functional	10,32	-	6,04	-	4,80	-	0
Gouriet *et al.,* 2019[1]	Calculated, PW91 functional	10,27	0,44	6,03	0,20	4,79	0,13	0
Hernandez *et al.,* 2014[2]	Calculated, PW91 functional	10,28	0,31	6,02	0,37	4,79	0,06	0
Durinck *et al.,* 2005[3]	Calculated, PW91 functional	10,28	0,33	6,04	0,02	4,79	0,06	0
Brodholt, 1997[4]	Calculated, PW91 functional	10,15	1,63	5,96	1,38	4,71	1,83	0
Xue *et al.,* 2017[5]	Calculated, PBE functional	10,20	1,09	5,99	0,92	4,76	0,73	0
Noel *et al.,* 2006[6]	Calculated, B3LYP functional	10,25	0,63	6,01	0,53	4,79	0,13	0
Song *et al.,* 2019[7]	Calculated, PZ81 functional	10,11	2,04	5,94	1,70	4,73	1,33	0
McKeown *et al.,* 2010[8]	Calculated, LDA functional	10,00	3,13	5,87	2,88	4,69	2,37	0
Stangarone, 2017[9]	Calculated, WC1LYP functional	10,18	1,31	5,98	1,08	4,77	0,62	0
	Calculated, WC1LYP functional, includes thermal effects	10,18	1,30	5,98	1,08	4,77	0,62	0
		10,24	0,73	6,00	0,62	4,78	0,32	300
		10,34	0,23	6,05	0,15	4,81	0,22	1000
Brodholt *et al.,* 1996[10]	Calculated, PZ81 functional	9,99	3,27	6,07	0,53	4,64	3,30	0
Fujino *et al.,* 1981[11]	Experimental	10,19	1,22	5,98	1,07	4,75	0,90	-
Smyth *et al.,* 2006[12]	Experimental	10,20	1,14	5,98	1,00	4,76	0,86	1873
Hazen, 1976[13]	Experimental	10,19	1,18	5,98	1,02	4,75	0,89	296
		10,18	1,32	5,98	1,10	4,75	1,05	77
Guyot *et al.,* 1996[14]	Experimental	10,19	1,21	5,98	1,00	4,75	0,93	298
Downs *et al.,* 1996[15]	Experimental	10,20	1,15	5,98	0,99	4,76	0,82	298
Kirfel *et al.,* 2005[16]	Experimental	10,19	1,21	5,98	1,07	4,75	0,93	298

[a] PBE – Perdew, Burke and Ernzerhof Generalized Gradient Approximation (GGA) functional[17], PW91 – Perdew and Wang[18] GGA functional; B3LYP – Becke 3-parameter Lee-Yang-Parr[19, 20] hybrid functional; PZ81 – Perdew and Zunger[21] Local Density Approximation (LDA) functional; WC1LYP – Wu and Cohen, 1 parameter, Lee-Yang-Parr[20, 22] hybrid functional.

Table S2 - Comparison of converged cell parameters with experimental and theoretical literature values for the enstatite crystal.

Author	Source[a]	a / Å	Error / %	b / Å	Error / %	c / Å	Error / %	Temperature /K
This study	Calculated, PBE functional	5,24	-	18,43	-	8,94	-	0
Stangarone *et al.*, 2016[23]	Calculated, WC1LYP functional	5,23	0,36	18,29	0,79	8,86	0,87	0
Demichelis *et al.,* 2012[24]	Calculated, PBE0 functional	5,19	0,98	18,24	1,02	8,83	1,26	0
Balan et *al.,* 2012[25]	Calculated, PBE functional	5,18	1,24	18,43	0,01	8,84	1,14	0
Stangarone, 2017[9]	Calculated, WC1LYP functional	5,18	1,27	18,21	1,20	8,81	1,46	0
	Calculated, WC1LYP functional, includes thermal effects	5,23	0,25	18,32	0,58	8,87	0,79	0
		5,24	0,15	18,31	0,67	8,88	0,64	300
		5,25	0,06	18,39	0,24	8,90	0,48	1000
Yang and Ghose, 1995[26]	Experimental	5,18	1,22	18,25	0,98	8,81	1,44	296
Ohashi, 1984[27]	Experimental	5,18	1,24	18,23	1,12	8,81	1,45	298
Ghose *et al.,* 1986[28]	Experimental	5,18	1,26	18,24	1,06	8,82	1,39	298
Morimoto and Koto, 1969[29]	Experimental	5,18	1,27	18,21	1,20	8,81	1,46	298

[a] PBE – Perdew, Burke and Ernzerhof GGA functional[17]; PBE0 – Hybrid functional based on the PBE exchange functional[30]; WC1LYP – Wu and Cohen, 1 parameter, Lee-Yang-Parr[20, 22] hybrid functional.

Table S3 - Comparison between the IR frequencies and the experimental data from literature for the forsterite crystal.

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}				
	This work, not scaled	This work, scaled[a]	Noel *et al.*, 2006[6]	Reynard, 1991[31]	Hofmeister, 1987[32]	Iishi, 1978[33]	Servoin and Piriou, 1973[34]	Sogawa *et al.*, 2006[35]
B1u	199	208	207	–	–	–	–	–
	264	276	278	274	276	274	274	276
	275	288	290	282	293	296	289	291
	285	298	313	–	309	–	–	305
	387	405	420	403	–	–	–	408
	399	417	428	411	412	423	415	415
	455	476	490	472	463	483	474	476
	483	505	514	501	502	502	501	502
	840	879	874	869	865	885	871	874
B2u	191	200	206	140	142	–	–	201
	262	274	275	275	268	280	276	275
	277	290	294	283	290	294	290	294
	306	320	322	341	345	352	350	320
	359	376	388	390	398	400	394	379
	383	401	412	415	418	421	416	403
	451	472	476	456	452	465	456	–
	483	505	513	503	504	510	506	502
	507	530	540	526	520	537	527	–
	577	604	614	–	–	–	–	604
	797	834	838	830	828	838	838	841
	920	962	962	867	865	882	875	958
	937	980	982	987	983	987	983	976
B3u	137	143	143	199	201	–	–	143
	262	274	277	274	275	274	274	277
	275	288	292	–	293	293	–	289
	330	345	350	316	319	320	318	348
	377	394	403	374	377	378	378	395
	397	415	432	397	405	403	404	416
	438	458	465	–	–	–	–	456
	486	508	517	498	505	498	498	506
	504	527	535	–	–	–	–	527
	598	626	638	601	601	601	601	–
	797	834	835	833	841	838	838	839
	839	878	870	952	950	957	957	873
	944	987	989	971	988	980	978	984

[a] Frequency scale factor used 1.046 calculated with least-squares linear fit (R^2=0.9998)

Table S4 - Comparison between the Raman frequencies and the experimental data from literature for the forsterite crystal.

Mode	Theoretical frequencies / cm^{-1}				Experimental frequencies / cm^{-1}				
	This work, not scaled	This work, scaled[a]	Noel *et al.*, 2006[6]	McKeown *et al.*, 2010[8]	Kolesov and Geiger, 2004[36]	Chopelas, 1991[37]	Iishi, 1978[33]	Servoin and Piriou, 1973[34]	McKeown *et al.*, 2010[8]
Ag	177	185	188	190	184	183	183	183	183
	219	230	234	227	227	226	227	227	227
	290	304	307	318	304	304	305	305	305
	312	327	329	337	329	329	329	329	329
	323	339	345	357	339	332	340	340	340
	402	421	425	436	421	422	424	424	423
	520	545	560	540	544	545	546	545	546
	577	605	618	606	608	608	609	609	609
	788	826	819	819	824	824	826	826	826
	820	859	856	849	856	856	856	856	856
	925	969	967	962	966	965	966	966	966
B1g	210	220	225	225	–	220	224	197	227
	243	255	260	259	–	274	260	265	275
	301	315	317	329	316	318	318	317	318
	336	352	367	365	–	351	–	–	374
	371	389	391	387	–	383	418	418	384
	416	436	442	447	434	434	434	434	434
	552	578	596	578	588	582	585	583	583
	600	629	645	629	–	632	632	632	632
	800	838	835	832	838	838	839	839	839
	828	868	866	859	866	866	866	866	866
	936	981	979	972	–	975	976	976	976
B2g	178	187	183	178	–	175	142	–	175
	276	289	253	250	243	242	244	244	244
	302	316	324	332	–	323	324	324	324
	356	373	373	374	–	365	368	365	368
	393	412	451	452	434	439	441	438	441
	563	590	608	580	588	586	588	585	588
	884	926	883	875	882	881	884	881	882
B3g	175	183	190	197	243	–	226	–	203
	232	243	303	289	–	286	272	–	272
	306	321	322	323	316	315	318	314	318
	348	365	381	387	373	374	376	374	376
	413	433	421	421	409	410	412	406	411
	558	585	609	587	588	592	595	591	595
	847	888	927	914	920	920	922	920	922

[a] Frequency scale factor used 1.048 calculated with least-squares linear fit (R^2=0.9998)

Table S5 - Comparison between the IR frequencies and the experimental data from literature for the enstatite crystal.

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}
	This work, not scaled	This work, scaled[a]	Demichelis *et al.*, 2012[24]	Demichelis *et al.*, 2012[24]
B1u	126	130	137	140
	134	139	147	147
	162	168	179	-
	183	189	196	197
	215	222	229	-
	224	232	238	238
	260	269	284	282
	276	285	300	305
	286	296	313	-
	306	316	331	328
	330	341	355	354
	332	343	358	-
	367	379	395	-
	391	404	414	408
	407	421	439	-
	436	451	464	463
	441	456	471	-
	459	475	486	-
	484	500	511	504
	515	533	548	537
	549	568	585	570
	620	641	662	656
	657	679	706	695
	837	865	868	850
	891	921	940	928
	930	962	989	978
	958	991	1022	1008
	1002	1036	1053	1043
	1084	1121	1153	1134

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}
	This work, not scaled	This work, scaled[a]	Demichelis *et al.*, 2012[24]	Demichelis *et al.*, 2012[24]
B2u	83	86	87	-
	142	147	145	-
	171	177	191	194
	193	200	202	204
	219	226	237	-
	247	255	266	-
	264	273	288	-
	277	286	297	293
	285	295	306	303
	300	310	318	317
	307	317	329	326
	324	335	353	351
	347	359	378	371
	386	399	413	406
	404	418	431	421
	416	430	446	436
	429	444	459	449
	481	497	509	504
	504	521	536	523
	518	536	554	545
	537	555	571	556
	613	634	653	645
	642	664	689	681
	837	865	868	-
	894	924	946	-
	895	925	950	940
	964	997	1027	1013
	1009	1043	1067	1055
	1104	1142	1171	1152

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}
	This work, not scaled	This work, scaled[a]	Demichelis *et al.*, 2012[24]	Demichelis *et al.*, 2012[24]
B3u	107	111	115	-
	183	189	201	-
	205	212	223	221
	212	219	223	224
	231	239	243	242
	255	264	274	271
	280	290	297	295
	313	324	339	336
	323	334	351	-
	327	338	358	350
	362	374	383	379
	379	392	407	397
	395	408	419	414
	414	428	441	434
	442	457	471	463
	453	468	482	475
	462	478	488	-
	466	482	495	-
	490	507	519	-
	522	540	559	550
	534	552	578	568
	702	726	736	725
	715	739	757	746
	849	878	902	890
	874	904	924	-
	887	917	929	919
	970	1003	1026	1013
	978	1011	1035	1021
	1028	1063	1086	1072

[a] Frequency scale factor used 1.034 calculated with least-squares linear fit (R^2=0.9977)

Table S6 - Comparison between the Raman peak positions and the experimental data from literature for the enstatite crystal.

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}				Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}			
	This work, not scaled	This work, scaled[a]	Stangarone, 2017[9]	Lin, 2003	Stalder *et al.*, 2009[38]	Chopelas, 1999[39]	Zhang *et al.*, 2013[40]		This work, not scaled	This work, scaled[a]	Stangarone, 2017[9]	Lin, 2004[41]	Stalder *et al.*, 2009[38]	Chopelas, 1999[39]	Zhang *et al.*, 2013[40]
Ag	73	77	79	83	-	83	-	B1g	146	153	153	-	-	-	-
	118	124	127	134	134	134	137		153	160	161	162	-	-	-
	149	156	159	155	-	153	-		169	177	184	-	-	-	190
	182	191	193	-	-	-	201		183	192	195	197	198	197	197
	189	198	201	-	-	-	-		210	220	224	-	-	-	-
	194	204	206	207	206	-	208		230	241	245	-	-	-	-
	223	234	236	238	239	237	240		270	283	283	-	-	-	-
	248	260	270	-	-	261	261		283	297	299	-	-	-	-
	262	275	277	278	-	-	272		293	307	311	-	-	-	-
	287	301	305	302	303	302	303		319	335	341	-	-	-	343
	304	319	325	323	-	-	-		326	342	348	-	-	-	-
	318	334	336	-	-	-	-		359	377	379	-	-	-	374
	323	339	343	344	344	343	345		371	389	395	-	-	-	-
	367	385	387	384	385		381		380	399	406	-	-	-	-
	383	402	406	402	403	402	403		403	423	422	-	-	-	-
	403	423	426	422	423	422	424		437	458	458	-	-	-	448
	427	448	451	446	446	446	445		445	467	468	-	-	-	-
	438	459	458	459	-	457	459		469	492	489	-	-	-	-
	460	483	481	-	-	487	488		495	519	522	-	-	-	-
	506	531	536	525	527	519	528		507	532	531	-	-	-	-
	522	548	552	552	554	553	555		554	581	594	595	581	-	-
	532	558	561	-	-	-	-		565	593	610	-	-	-	-
	631	662	668	664	665	665	665		704	738	735	-	-	-	-
	652	684	693	686	687	687	688		722	757	759	-	-	-	-
	837	878	866	851	853	886	851		849	891	886	-	-	-	-
	888	932	924	927	928	927	930		881	924	919	-	-	-	-
	894	938	933	935	937	937	-		909	954	945	-	-	-	-
	966	1013	1012	1011	1013	1014	-		982	1030	1023	-	-	-	-
	989	1037	1027	1033	-	-	-		1003	1052	1043	-	-	-	-
	1003	1052	1039	-	1035	1034	-		1020	1070	1067	-	-	-	-

[a] Frequency scale factor used 1.049 calculated with least-squares linear fit (R^2=0.9986)

Table S6 - Continuation.

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}			
	This work, not scaled	This work, scaled[a]	Stangarone, 2017[9]	Lin, 2004[41]	Stalder *et al.*, 2009[38]	Chopelas, 1999[39]	Zhang *et al.*, 2013[40]
B2g	78	82	81	107	-	-	-
	151	158	160	-	-	-	-
	156	164	164	-	-	-	-
	194	204	212	-	-	-	-
	221	232	238	-	-	239	241
	232	243	248	245	-	245	246
	269	282	285	-	-	-	-
	288	302	304	-	-	-	-
	294	308	311	-	-	-	-
	317	333	341	-	-	-	340
	341	358	359	-	-	-	-
	368	386	389	-	-	-	-
	376	394	401	-	-	-	-
	391	410	415	-	-	-	-
	406	426	426	-	-	-	-
	429	450	449	-	-	-	-
	452	474	474	473	-	-	479
	456	478	477	-	-	-	-
	473	496	498	-	-	-	-
	503	528	527	-	-	-	-
	524	550	553	-	-	-	-
	564	592	610	-	-	-	-
	713	748	749	751	-	-	753
	723	758	760	-	-	-	-
	856	898	894	-	-	-	-
	870	913	906	-	-	-	-
	908	952	943	-	-	-	-
	972	1020	1017	-	-	-	-
	987	1035	1025	-	-	-	-
	1022	1072	1063	-	-	-	-

Mode	Theoretical frequencies / cm^{-1}			Experimental frequencies / cm^{-1}			
	This work, not scaled	This work, scaled[a]	Stangarone, 2017[9]	Lin, 2004[41]	Stalder *et al.*, 2009[38]	Chopelas, 1999[39]	Zhang *et al.*, 2013[40]
B3g	98	103	108	115	-	115	-
	136	143	145	-	-	-	-
	146	153	158	-	-	-	-
	158	166	166	167	-	166	170
	184	193	196	-	-	-	-
	207	217	225	-	-	-	-
	223	234	233	-	-	-	-
	249	261	263	-	-	-	-
	277	291	296	-	-	-	287
	279	293	299	-	-	-	-
	297	312	318	-	-	-	-
	305	320	327	328	-	-	-
	346	363	364	-	-	-	-
	366	384	386	-	-	-	-
	388	407	412	-	-	-	-
	398	418	422	-	-	-	-
	427	448	450	-	-	-	-
	446	468	470	-	-	-	-
	495	519	523	-	-	-	-
	510	535	542	-	541	540	542
	520	545	550	-	-	-	-
	537	563	566	580	-	580	582
	624	655	659	-	-	-	-
	647	679	688	-	-	-	-
	838	879	866	-	-	-	-
	893	937	932	-	-	-	-
	916	961	957	-	-	-	-
	975	1023	1024	-	-	-	-
	989	1037	1026	-	-	-	-
	1100	1154	1153	-	-	-	-

[a] Frequency scale factor used 1.049 calculated with least-squares linear fit (R^2=0.9986)

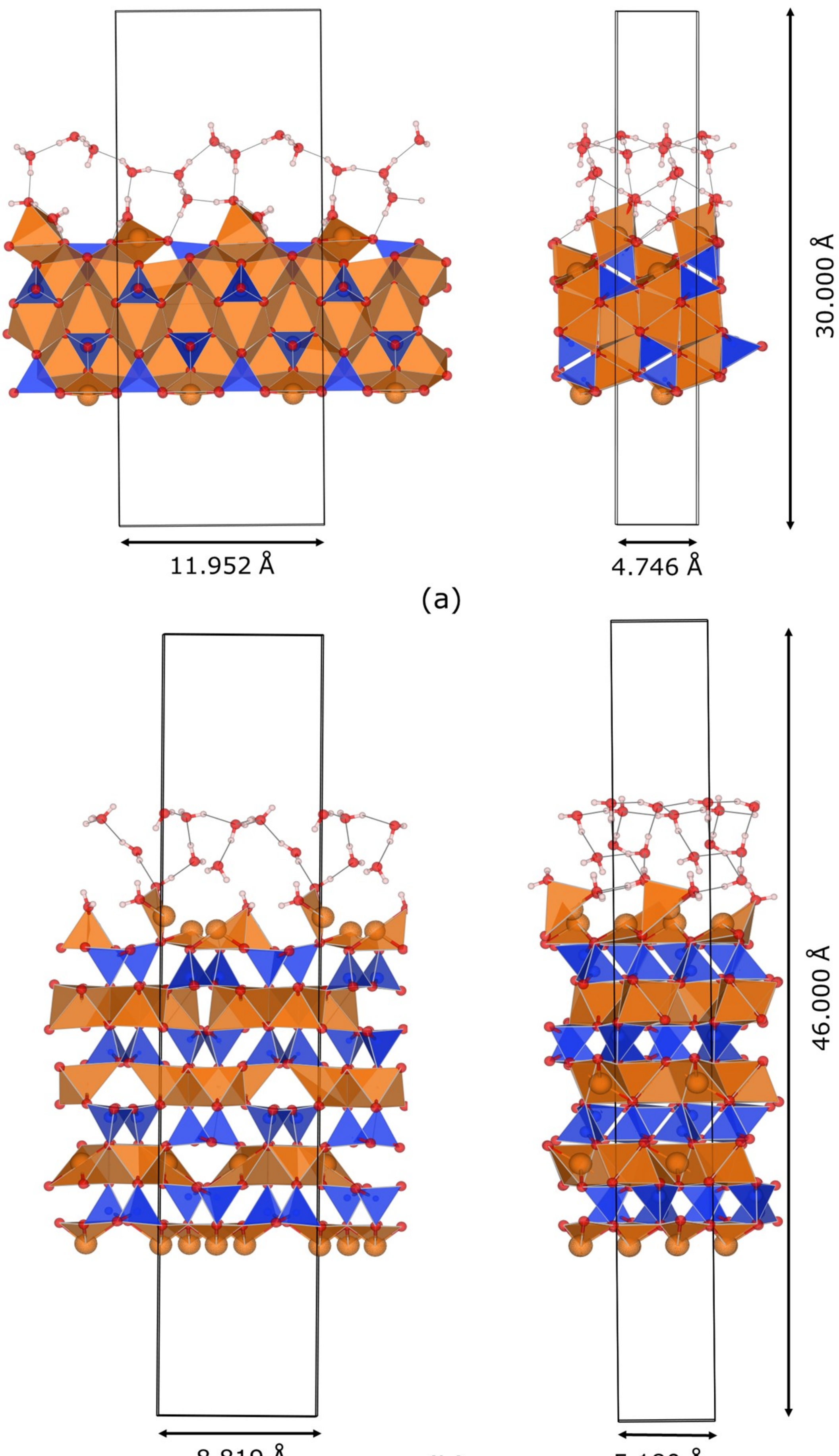

Figure S1 – Side view of the final equilibrium structures of forsterite+ice (a) and enstatite+ice (b). Si atoms in blue; Mg atoms in orange; O atoms in red; H atoms in white

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