Supporting Information for

Pre-Accretion Formation of Sulfuric Acid in Interstellar Ice Analogs: Implications for Sulfate in Ryugu and Bennu

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MATERIALS & METHODS

Photoionization Reflectron time-of-flight mass spectrometry

Cryogenic ices were prepared in an ultra-high vacuum chamber[1-2] maintained at 8×10^{-11} Torr by a turbomolecular pump (Osaka, TG1200M) in series with a second turbomolecular pump (Osaka, TG420M), backed by a multistage roots pump (Leybold, Ecodry 40 Plus). Gases were deposited onto a polished silver substrate affixed to a high-purity copper cold finger interfaced with a two-stage helium refrigerator (Sumitomo Heavy Industries, RDK-415E). The substrate was maintained at 5 K, as monitored by a cryogenic temperature sensor (Lake Shore, DT-470) and controlled by a programmable temperature controller (Lake Shore, Model 336) with a precision of ± 0.1 K.

Sulfur dioxide (SO_2; Air Liquide; 99.999%) and water (H_2O; Shanghai Macklin Biochemical Technology Co.; HPLC grade) were introduced into the chamber through a glass capillary array aimed at the target silver wafer at pressures of 1.5×10^{-9} and 2.8×10^{-8} Torr, respectively. The deuterated experiment used deuterium oxide (D_2O; Shanghai Aladdin Biochemical Technology Co.; 99.9 atom % D) with the same pressure ratio as the non-isotopically labeled experiments. Deposition was monitored by laser interferometry using a He–Ne laser (CVI Melles Griot; 25-LHP-230, 632.8 nm) at an incidence angle of 2° and a photodiode (Thorlabs, SM1PD1A) connected to a picoammeter (Keithley 6485).[3] Using the combined average refractive index of each ice component, the ice thickness was calculated to be 980 ± 80 nm (Table S2).

After deposition, irradiation was conducted isothermally at 5 K for 1 h at currents of 100 nA (low dose) and 1000 nA (high dose) using an electron gun (SPECS GmbH, EQ 22/35) producing 5 keV electrons. Using CASINO v2.42[4] and the combined average density of each ice component, the 99th percentile penetration depth was calculated to be 700 ± 100 nm (Table S3), which is smaller than the ice thickness derived from interferometry, thereby preventing interactions between the electrons and the silver–ice interface (Table S3). These electrons simulate secondary electrons produced from galactic cosmic rays (GCRs) as they penetrate interstellar grains in cold molecular clouds up to an age of 5×10^7 years.[5-6]

After irradiation, temperature-programmed desorption (TPD) was initiated by warming the silver target from 5 to 330 K at a rate of 1.0 K min^{-1}. Material subliming from the wafer surface was subsequently ionized by synchrotron-generated vacuum ultraviolet (VUV) light produced by

an undulator at the BL03U beamline of the National Synchrotron Radiation Laboratory. The resulting ions were accelerated, separated, and collected using a pulsed reflectron time-of-flight mass spectrometer (Jordan TOF Products, Inc.) operated with a 2.5 μs pulse width generated by a pulse delay generator (Quantum Composers, 9528) at a repetition rate of 10 kHz. The signal was processed by an amplifier-discriminator (Advanced Research Instruments Corporation) and collected using a multichannel scalar (FAST ComTec, MCS8). Each resulting spectrum contained 0.6 million sweeps binned at a width of 3.2 ns, corresponding to a 60 s data collection time.

Computations

The adiabatic ionization energies (IEs) and relative energies are computed with CCSD(T)-F12/cc-pVTZ-F12 single point energies from the MOLPRO quantum chemical software (Table S4).[7-10] These are based on B3LYP/aug-cc-pVTZ optimized geometries and harmonic zero-point vibrational energies (ZPVEs) computed with Gaussian16 (Table S5 and S6).[11-15] This approach is known to produce accuracies within 1 kJ mol^{-1} of pure CCSD(T)-F12/cc-pVTZ-F12 values in most instances.[16-17] Several conformers of the various isomers have been explored, but the table only reports the unique structures. Additionally, further CCSD(T)-F12/cc-pVTZ-F12 optimizations report a second minimum for the cation of H_2SO_4. The IE computed from the lowest energy neutral and cation for this higher-level approach is the same as that produced using the CCSD(T)-F12/cc-pVTZ-F12 single-point energy from the B3LYP/aug-cc-pVTZ optimized geometry, 11.53 eV.

Table S1. Summary of detected mass-to-charge in the 12.10 eV high dose experiment along with possible identities.

Mass-to-Charge (*m/z*)	**Possible Cation Formula**
34	H_2S; H_2O_2
66	H_2S_2
80	S_2O, SO_3^*
98	H_2SO_4
128	S_4, S_3O_2, S_2O_4
142	H_2S_4O, $H_2S_3O_3$, $H_2S_2O_5$
160	S_5, S_4O_2, S_3O_4
192	S_6, S_5O_2, S_4O_4, S_3O_6
208	S_6O, S_5O_3, S_4O_5, S_3O_7
240	S_7O, S_6O_3, S_5O_5, S_4O_7
256	S_8, S_7O_2, S_6O_4, S_5O_6, S_4O_8

*****12.10 eV cannot ionize sulfur trioxide (12.80 ± 0.04 eV) in this experiment; however, isomers of SO_3 could be energetically available for ionization.

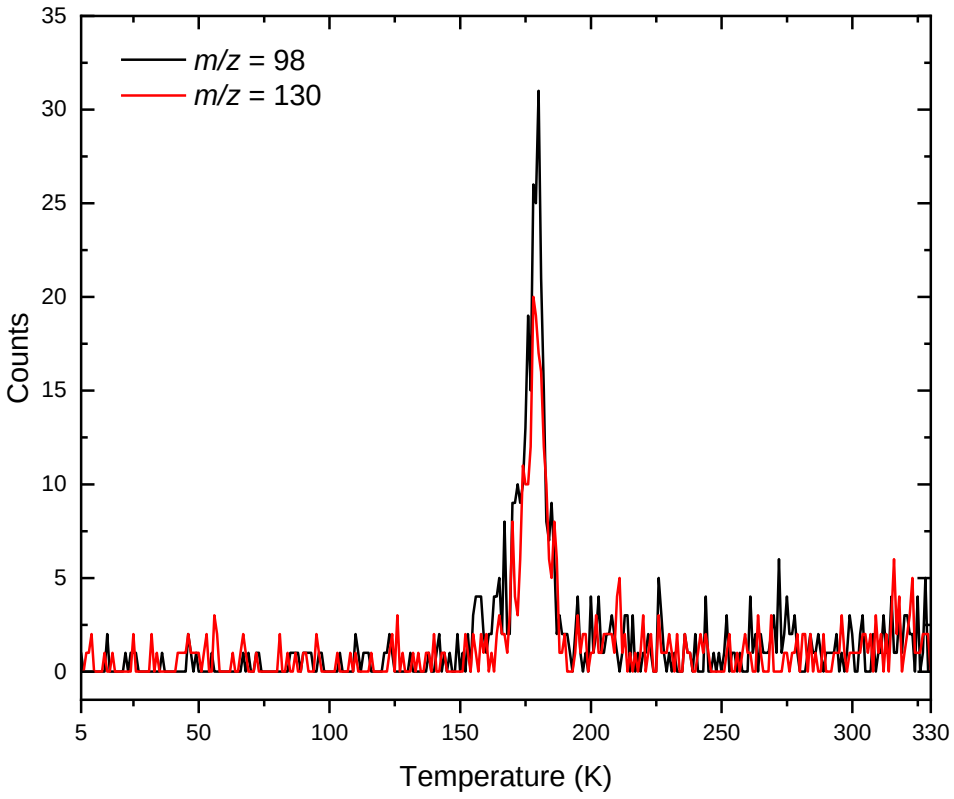


Figure S1. TPD-PI-ReToF-MS of *m/z* = 98 (red) and *m/z* = 130 (black) using 11.60 eV light. The sublimation event at 179 K shows possible fragmentation from *m/z* = 130 to *m/z* = 98.

Table S2. Physical parameters of each ice reagent used in these experiments.

Ice Component	Density ($g\ cm^{-3}$)	Refractive Index (n)	Reference
SO_2	1.395 ± 0.005	1.332 ± 0.005	[18]
H_2O	0.719 ± 0.005	1.234 ± 0.008	[19]

Table S3. Calculated Dose for SO_2 in cryogenic ice analogs utilizing CASINO v2.42 and the respective derived Molecular Cloud age.[4]

Experiment	Density (g cm^{-3})	Dose (eV molecule^{-1})	Average Ice Thickness (nm)	99% Penetration Depth (nm)	Derived Age of Molecular Cloud (years)
SO_2/H_2O (1:1) Low Dose	1.15	19 ± 3	980 ± 80	700 ± 100	5×10^6
SO_2/H_2O (1:1) High Dose	1.15	192 ± 29	-	-	5×10^7

Table S4. List of all molecules calculated here at the CCSD(T)-F12/cc-pVTZ-F12//B3LYP/aug-cc-pVTZ level of theory along with the adiabatic ionization energy and the relative energy to the global minimum of the H_2SO_4 isomer surface.

Identity	Adiabatic Ionization Energy (eV)	Relative Energy (kJ mol^{-1})
1a[a]	11.75	3
1b[b]	11.53	0
2	9.71	277
3a	10.10	296
3b	10.02	302
3c	9.85	306
3d	9.82	309
4	8.40	504
5a	9.69	589
5b	9.69	590
5c	9.66	590
5d	9.64	594
6a	10.61	591
6b	10.50	602
6c	10.23	604
6d	10.23	604
6e	10.18	609
6f	8.19	600
6g	8.16	598
6h	8.11	604
6i	8.09	605

[a]The cation minimum here is a C_2 structure, and the relative energy is for the difference in the two cations.

[b]The cation minimum here is a C_{2v} structure.

Table S5. Calculated coordinates for the H_2SO_4 isomer system including neutral and cations. All geometries are in angstroms (Å). Calculations utilized CCSD(T)-F12/cc-pVTZ-F12//B3LYP/aug-cc-pVTZ level of theory unless otherwise noted.

1a/1b B3LYP

S	0.2072270321	0.0555263160	-0.1581187567
O	0.0277286637	0.3403050207	1.4163932974
H	0.2903823876	-0.4527225115	1.9075318303
O	-1.1907594235	-0.6519381738	-0.5289753991
H	-1.8459004169	0.0359227114	-0.7206375638
O	0.2489101215	1.3463726227	-0.7832467896
O	1.2109503807	-0.9552041796	-0.3306586934

1a/1b CCSD(T)-F12/cc-pVTZ-F12

S	0.0000000000	0.0000000000	0.1318098238
O	-1.0933884113	0.6022373963	0.7966504514
O	1.0933884113	-0.6022373963	0.7966504514
O	0.6301266115	1.0542009513	-0.8586102189
O	-0.6301266115	-1.0542009513	-0.8586102189
H	-0.0545947935	1.6865411392	-1.1127570817
H	0.0545947935	-1.6865411392	-1.1127570817

$1a^+$ C_2 CCSD(T)-F12/cc-pVTZ-F12

S	0.0000000000	-0.0000000000	-0.0364815631
O	-0.3950162132	1.0751262339	-0.9239043649
O	0.3950162132	-1.0751262339	-0.9239043649
O	1.1845835335	0.3030597788	0.8631755083
O	-1.1845835335	-0.3030597788	0.8631755083
H	1.0521796696	0.9910510445	1.5441640624
H	-1.0521796696	-0.9910510445	1.5441640624

$1b^+$ C_2 B3LYP

S	0.0123587568	0.0033115189	-0.0094300020
O	-0.1108904299	-0.1106523143	1.5186584496
H	0.7259672645	0.0502621201	2.0039514359
O	-1.4602219150	-0.3103259701	-0.3198653793
H	-1.6807728449	-0.3061015045	-1.2754139022
O	0.4795849100	1.3141647114	-0.4946648015
O	0.9825130033	-0.9223967558	-0.6209478751

$1b^+$ C_{2v} CCSD(T)-F12/cc-pVTZ-F12

S	0.0000000000	0.0000000000	-0.0415378816
O	0.0000000000	1.1301830438	-0.9570464385
O	0.0000000000	-1.1301830438	-0.9570464385
O	1.1279272941	0.0000000000	0.9630713422
O	-1.1279272941	0.0000000000	0.9630713422
H	2.0208901412	0.0000000000	0.5649715236

H	-2.0208901412	0.0000000000	0.5649715236

2

O	-0.0743583954	-0.0271870421	0.0097466739
S	-0.0136991235	-0.0167743227	1.4684081050
O	1.2903290645	0.1862315419	2.0728478066
O	-0.8484935937	2.7218854485	-0.2728364330
O	-0.7916535849	2.5906109179	1.1650566009
H	-0.0325995731	3.1538175501	1.3761958313
H	-0.5815726159	1.8262157284	-0.5529948400

2^+

O	0.0154845346	0.0072444672	-0.0000965960
S	-0.0063989304	-0.0027505668	1.5668533392
O	1.3043280829	0.0022444107	2.1561203987
O	2.4182817505	2.2032111266	2.6931165887
O	3.4571918726	1.7779382760	3.3591911775
H	3.9276876989	2.5781980430	3.6837849710
H	0.8906121459	-0.0317336937	-0.4435739429

3a

S	-0.0011261693	-0.0010575483	-0.0008803051
O	-0.0012643762	0.0009034889	1.6978518744
O	1.3592759410	0.0038447772	2.1842450082
H	1.6130979359	-0.9263179055	2.0674786082
O	0.7166334859	1.4362820669	-0.2761869641
H	1.6549457712	1.3789614414	-0.0260730092
O	0.9465348478	-1.0448224397	-0.3940003584

$3a^+$

S	0.2093862539	-0.7215202167	-0.4201218896
O	-0.1457051001	-0.1978284276	1.6539990396
O	0.9446554401	0.3314015479	2.0879451739
H	1.6222737463	0.3044226672	1.3303403949
O	-0.6096309779	0.4785184220	-1.0339503972
H	-1.5325828399	0.2538645372	-1.2732753630
O	1.5823934776	-0.2797435300	-0.3384989586

3b

O	-0.0811980038	0.0784164700	-0.0834957253
S	-0.1644423285	0.1195356101	1.3739219882
O	1.3284965206	0.3348660661	1.9901803069
H	1.9770871169	-0.1505326754	1.4522657754
O	-0.1744976466	-1.5268245661	1.8669381929
O	-1.4220648456	-2.0741131524	1.3903473721
H	-1.1913236001	-2.3265587216	0.4823293019

$3b^+$

O	-0.3100256050	-0.1733808675	0.0499559467
S	-0.1669859132	0.3125824302	1.4083374957
O	1.3235978578	0.3217485807	1.8837855848
H	1.9908369919	-0.1060702695	1.3056216710
O	-0.7207178415	-1.6347733851	2.2244524871
O	-0.9824823975	-2.4046119482	1.2230833624
H	-0.8621658796	-1.8607055099	0.3772506646

3c

O	-0.0169059638	0.1821436023	0.0260033433
S	-0.1078333392	0.0406723350	1.4748380936
O	1.4371207398	0.1526214195	2.0311562937
H	2.0250782734	0.0546803548	1.2635413323
O	-0.4049159249	-1.5774946014	1.8601701045
O	0.5298551044	-2.4700069815	1.2049944214
H	1.0937166002	-2.7097100530	1.9558501705

3c^{+}

O	-0.0174074000	-0.0361057126	0.0184901507
S	0.0076928459	0.0044635909	1.4485141795
O	1.4629915191	0.0112351655	2.0299891894
H	2.1952324180	-0.1055033286	1.3877670369
O	-0.2994095556	-2.2257866818	1.9433541771
O	-0.8614552454	-2.7610669012	0.8894949242
H	-1.0086871702	-3.7090839205	1.1167560232

3d

O	0.2348455734	0.2145139556	-0.2202688293
S	-0.0865952686	0.2744885452	1.1974134356
O	1.3092039605	0.3384358696	2.0433338606
H	1.9942601437	-0.1528731941	1.5586145932
O	-0.3531164332	-1.3458284398	1.7171895146
O	-1.6187969992	-1.7370992752	1.1299791314
H	-2.1749988951	-1.7927227163	1.9209244133

3d^{+}

O	0.2299469793	0.1624902863	-0.2159577002
S	0.1884190156	0.6447280594	1.1305286066
O	1.5000764580	0.3301648022	1.9285574431
H	2.1657212753	-0.2325893878	1.4785670694
O	-0.9779155539	-1.0574676008	2.1561325537
O	-1.6443961106	-1.6682337836	1.2097245128
H	-2.1570499823	-2.3801776306	1.6596336341

4

O	0.0094235122	0.0194188689	0.0161014522
S	-0.0049855389	0.0161167873	1.6694053244

O	1.3787084860	-0.0700596375	2.1344240391
O	1.6216597788	2.2630970115	2.0964921148
O	0.2736370167	2.3043630453	1.9270496229
H	0.1330987927	2.5414076163	0.9913774273
H	0.9355440382	0.0064884086	-0.2794622137

4^{+}

O	0.0000000000	0.0000000000	0.0000000000
S	0.0000000000	0.0000000000	1.5672015400
O	1.3189119866	0.0000000000	2.1380499208
O	2.4566483687	2.1972084362	2.6430118073
O	3.5033396100	1.7688870699	3.2947080552
H	3.9839894182	2.5678270954	3.6075916935
H	0.8686836393	-0.0471447405	-0.4552312086

5a

O	0.0160345141	-0.2687470489	-0.0796865019
O	0.0772948644	-0.0441515657	1.4072650175
S	1.6037584805	0.0959699336	1.9347650132
O	1.9248333013	1.6834924290	1.8286607169
O	2.5738093945	2.0496242778	0.5373472973
H	1.7976352055	2.3376151132	0.0327055990
H	-0.4609435412	-1.1116596133	-0.0925552276

5a^{+}

O	0.0635753693	-0.1603099260	-0.0568291082
O	0.0159332469	0.0195756501	1.3428667794
S	1.5521955817	0.1339064495	1.9186230724
O	1.1949320143	0.3127191400	3.4383467227
O	1.0050633826	1.7573061952	3.7382389107
H	1.4850851727	1.8088821654	4.5882496669
H	-0.8850765440	-0.3113381374	-0.2456773767

5b

O	-0.0004187191	-0.0400919531	-0.0168138082
O	0.1579846649	-0.1268668149	1.4755904394
S	1.6983166407	0.0615010436	1.9456556322
O	1.9675951269	1.6595819912	1.9222629314
O	1.4601583687	2.3435655235	3.1493896138
H	0.6733365032	2.7738774377	2.7821943048
H	-0.3349153531	-0.9327665754	-0.1894493665

5b^{+}

O	-0.0479440251	0.0937959367	0.2486010570
O	0.5066966191	-0.3887262269	1.5473026799
S	2.0063903826	0.0399915796	1.7045251003
O	1.9952081837	1.5842942908	2.2600982613

O	0.7201072558	2.1189191195	2.4963014088
H	0.9682156331	3.0038624094	2.8367111067
H	-0.5266168170	-0.7133364567	-0.0247098670

5c

O	0.0316080531	0.0055858518	-0.1020250066
O	0.0980835287	0.0386802290	1.3958460888
S	1.6195081343	-0.0668028184	1.9486274251
O	1.9799536170	-1.6481085768	1.9789953601
O	1.4914755503	-2.3209752103	3.2271863989
H	0.8103322203	-2.8957115453	2.8480708877
H	-0.4089038716	-0.8477158030	-0.2278714072

5c^{+}

O	0.1122464686	-0.8383605323	0.3087721261
O	0.3617051363	0.2538151827	1.1809815970
S	1.6936336863	0.0945096725	2.0933039857
O	2.0547788482	-1.4312985833	2.1739238865
O	1.0129726993	-2.1951979341	2.9044624618
H	1.1326698600	-3.0648457523	2.4742761401
H	-0.7459494667	-0.5536699261	-0.0668904502

5d

O	0.0079244976	-0.2571176043	-0.0956722343
O	0.0710888106	0.0179095193	1.3792837669
S	1.5927870928	0.0426521685	1.9269730601
O	2.0453498280	1.5923424955	1.8284823290
O	2.7144989047	1.9015730516	0.5200885723
H	3.5491993147	2.2598864686	0.8571090302
H	-0.5761503161	-1.0300016783	-0.0813677588

5d^{+}

O	-0.3089619459	-0.2868732272	0.0287914826
O	0.2340862248	-0.1610069119	1.4077885904
S	1.8042875988	-0.1311898192	1.3536290158
O	1.9877041053	1.4925717385	1.1668656032
O	3.3656022388	1.7919743201	1.0943965735
H	3.3316589249	2.7703461637	1.0911785142
H	-1.0096790143	-0.9485778432	0.1922469856

6a

S	-0.0453110610	-0.1764676393	0.2069002042
H	0.1658858517	0.3583600712	1.4628079614
O	1.3063511527	-0.5507136705	-0.2722364118
O	-0.4881987654	1.2973108308	-0.4412791261
O	0.5841580075	2.2753081079	-0.1407709867
O	1.4518803941	2.3059544647	-1.2385220667

H	2.0020229749	1.5145955730	-1.0896714758

6a^{+}

S	0.0022180121	0.0008725303	0.0003591156
H	-0.0051714389	0.0015980159	1.3759702045
O	1.3316145744	0.0044582246	-0.5318580064
O	-1.2360887622	-0.4894701160	-0.5424990508
O	-1.8958842905	-2.8326332455	-0.7339335804
O	-2.8140526351	-2.7190907618	-1.6452139735
H	-3.1849431121	-3.6211212863	-1.7885885060

6b

S	0.1046263232	0.2453009443	0.2680306088
H	0.2937184584	-0.5345311311	1.3933146054
O	1.4442759665	0.7180709254	-0.1152583755
O	-0.4185975828	-1.0973204862	-0.6115035693
O	0.3713851134	-2.3071840794	-0.3238050599
O	1.3043861428	-2.4789200367	-1.3581467321
H	0.7900746271	-2.9601531849	-2.0267989547

6b^{+}

S	0.0022180121	0.0008725303	0.0003591156
H	-0.0051714389	0.0015980159	1.3759702045
O	1.3316145744	0.0044582246	-0.5318580064
O	-1.2360887622	-0.4894701160	-0.5424990508
O	-1.8958842905	-2.8326332455	-0.7339335804
O	-2.8140526351	-2.7190907618	-1.6452139735
H	-3.1849431121	-3.6211212863	-1.7885885060

6c

S	-0.0839085340	0.1202832302	0.0174114168
H	0.2385091656	-0.1487581358	1.3337817052
O	1.1847779712	0.5582934043	-0.6005856590
O	-0.4410812448	-1.4629106527	-0.3139187954
O	0.7091182570	-2.4175806706	-0.1497248440
O	1.2398188226	-2.2987679120	1.1381041294
H	2.0345995455	-1.7594048682	0.9808403572

6c^{+}

S	-0.2167887988	0.3911237653	0.0017627649
H	-0.1980469401	0.1497390933	1.3677254972
O	1.1191824213	0.8564276055	-0.3241511407
O	-0.3697669368	-1.7439174427	-0.1924916345
O	0.6774551580	-2.4718298280	-0.1227508240
O	1.5613141263	-1.9799760900	0.8801325538
H	2.3084849533	-2.6104127081	0.7956810934

6d

S	-0.0839085317	-0.1202832302	0.0174114072
H	0.2385091748	0.1487581440	1.3337816924
O	1.1847779821	-0.5582933900	-0.6005856632
O	-0.4410812458	1.4629106504	-0.3139187955
O	0.7091182551	2.4175806749	-0.1497248360
O	1.2398188154	2.2987679044	1.1381041367
H	2.0345995333	1.7594048513	0.9808403686

6d^{+}

S	-0.2167887992	-0.3911237527	0.0017627474
H	-0.1980469840	-0.1497391264	1.3677254884
O	1.1191824364	-0.8564275664	-0.3241511353
O	-0.3697669533	1.7439174536	-0.1924915761
O	0.6774551562	2.4718298217	-0.1227508160
O	1.5613141493	1.9799760810	0.8801325428
H	2.3084849779	2.6104126939	0.7956810588

6e

S	0.2766972978	0.1063354348	0.3497151856
H	1.0187647330	-0.2809024077	1.4496363226
O	1.1802293598	0.0575330444	-0.8097194533
O	-0.5329379933	-1.3880730807	0.5846159942
O	-1.7420735364	-1.3649696596	-0.2080666919
O	-2.7973858169	-0.9158121918	0.6241695074
H	-2.8406081169	0.0313684091	0.4155574454

6e^{+}

S	0.0000000000	0.0000000000	0.0000000000
H	0.0000000000	0.0000000000	1.3872843200
O	1.3988357461	0.0000000000	-0.3860076286
O	-0.4817733462	-2.0583075502	0.3892427975
O	-1.2935190718	-2.6804020023	-0.3735937768
O	-2.3698797386	-1.8305651362	-0.7523797345
H	-2.8841588780	-2.4305031226	-1.3345716355

6f

S	0.0810468976	0.0180211484	0.7208388911
H	0.9932422278	-0.9289816337	1.1581419862
O	0.5759700789	0.5425324246	-0.5685062373
O	-0.9348462412	-1.3183423780	0.4341709310
O	-2.1801610339	-0.7271912478	-0.0553888176
O	-2.1822118874	-0.8617655032	-1.4558038045
H	-1.6221677222	-0.1202290170	-1.7462248506

6f^{+}

S	-0.8235540363	0.2345547627	-0.0775796175
H	-0.4023232359	0.8484868246	1.0700098351

O	-1.3410896135	-1.0657650173	0.1634022965
O	-1.1001118904	1.1587601493	-1.1192661436
O	1.1907554762	-0.2483772001	-0.5506632157
O	1.9585652049	0.0926005483	0.4546343399
H	2.8751120950	-0.1625210674	0.1869365054

6g

S	-0.0111526788	0.1192424501	-0.0109539846
H	0.4712668581	-0.1004661248	1.2650402513
O	1.1029032218	0.7158475183	-0.7656571346
O	-0.1680275886	-1.5473482541	-0.2796232805
O	1.0287783994	-2.3028573581	0.1017096470
O	0.9463404222	-2.5572221653	1.4868711336
H	0.3721016029	-3.3398453305	1.5221774034

6g^{+}

S	-0.7625239545	-0.0397925433	0.2358085876
H	-0.7694820106	-0.0949905707	1.6002468928
O	-0.7077812777	1.2934758397	-0.2538432056
O	-1.2899064198	-1.2201059883	-0.3525650387
O	1.2928415438	-0.5716610042	0.2679528253
O	1.9673597978	0.3596143480	-0.3529174132
H	2.9010353210	0.0340439189	-0.3796846482

6h

S	0.0032813720	0.0351478371	0.0059605070
H	0.0214040722	-0.0403075572	1.3879652252
O	1.4090534766	0.0222415260	-0.4266038012
O	-0.6258564597	-1.5678747842	0.1098687500
O	-1.1642273885	-1.9072537883	-1.1869805555
O	-2.5265149601	-1.5191199691	-1.2019757054
H	-2.9622854647	-2.2870625083	-0.7984216633

6i

S	0.0679316962	0.1353884604	0.4710287565
H	0.6730609609	-0.6553188150	1.4319041032
O	1.0697461421	0.3244155001	-0.5893789317
O	-0.9030080909	-1.2625972352	0.2031875534
O	-2.1267497068	-0.8434624288	-0.4463216618
O	-1.9462626221	-0.9597151506	-1.8440013576
H	-2.1341887311	-1.8992219406	-2.0005859392

6i^{+}/6h^{+}

S	0.0007460093	-0.0014825971	0.0007461309
H	-0.0233549360	-0.0130035229	1.3660376719
O	1.3234292610	0.0031995700	-0.5196951811
O	-1.2128028890	-0.4789226312	-0.5621059371

O	-0.4467797352	2.0734013229	0.0548264049
O	0.4953901484	2.7135586140	-0.5853839623
H	0.2072721892	3.6597452465	-0.5989707092

Table S6. Harmonic frequencies (cm^{-1}) derived from calculated geometries in Table S5.

1a/1b

Frequencies --	235.0939	312.3841	356.5171
Frequencies --	418.4536	476.7328	521.4762
Frequencies --	530.8595	786.8256	840.5288
Frequencies --	1150.6592	1163.3670	1185.3128
Frequencies --	1430.4745	3746.5118	3750.6863

$1a^+$

Frequencies --	223.32	310.87	324.17
Frequencies --	372.44	438.34	484.76
Frequencies --	512.67	756.33	894.50
Frequencies --	1024.08	1066.82	1089.87
Frequencies --	1275.03	3651.45	3662.64

$1b^+$

Frequencies --	176.9444	343.1412	347.1037
Frequencies --	367.4550	412.6843	457.2864
Frequencies --	482.0877	805.6176	867.3937
Frequencies --	932.8333	1119.3757	1156.0283
Frequencies --	1196.8501	3604.1219	3613.7750

2

Frequencies --	54.6446	85.0458	123.7062
Frequencies --	182.8795	228.9813	335.7637
Frequencies --	483.3076	525.0713	954.0180
Frequencies --	1145.4772	1313.7708	1343.7048
Frequencies --	1477.6996	3596.8874	3732.2187

2^+

Frequencies --	32.1543	53.5423	72.2435
Frequencies --	103.5959	211.6217	279.4344
Frequencies --	429.6797	506.1824	867.6817
Frequencies --	1041.7041	1187.6978	1288.0125
Frequencies --	1420.7249	3516.6805	3581.3366

3a

Frequencies --	103.4567	179.1826	282.6846
Frequencies --	327.7310	422.5686	442.7315
Frequencies --	565.7888	602.0776	767.7465
Frequencies --	903.0722	1113.9461	1218.3346
Frequencies --	1395.5632	3687.2324	3700.8557

3a$^+$

Frequencies --	131.0349	199.3367	220.1628
Frequencies --	340.3656	421.2341	474.8937
Frequencies --	511.0016	684.9159	886.2115
Frequencies --	1046.8902	1244.3039	1280.4146
Frequencies --	1484.1909	2980.6932	3618.3521

3b

Frequencies --	106.7528	197.3576	261.2590
Frequencies --	353.2794	379.7741	437.5508
Frequencies --	490.7541	676.4583	778.4029
Frequencies --	931.8300	1112.5938	1227.7855
Frequencies --	1399.5291	3690.4793	3711.3772

3b$^+$

Frequencies --	134.4819	182.7858	211.2937
Frequencies --	330.3912	421.3978	476.3923
Frequencies --	511.8895	671.4344	879.6456
Frequencies --	1053.3187	1233.8910	1269.1359
Frequencies --	1478.0163	3043.7288	3590.2082

3c

Frequencies --	118.0763	193.4392	228.2412
Frequencies --	368.5160	385.5781	422.9625
Frequencies --	573.2828	593.9847	748.2338
Frequencies --	896.4797	1111.9289	1241.5909
Frequencies --	1380.3524	3699.5526	3731.3911

3c$^+$

Frequencies --	31.1016	117.1755	156.0269
Frequencies --	271.5275	306.6015	408.1562
Frequencies --	459.4719	527.3876	880.6464
Frequencies --	1054.1318	1214.0675	1331.8625
Frequencies --	1428.2491	3510.8373	3592.7799

3d

Frequencies --	102.3399	203.4268	269.7308
Frequencies --	312.1908	371.1122	426.0258
Frequencies --	474.8582	672.2910	773.2549
Frequencies --	929.1088	1115.9082	1248.3074
Frequencies --	1380.1948	3691.6043	3736.7800

3d$^+$

Frequencies --	30.3075	117.2603	155.9779

Frequencies -- 271.6546 306.9996 408.1608
Frequencies -- 459.5734 527.6561 880.5834
Frequencies -- 1054.3157 1214.2321 1331.7807
Frequencies -- 1428.3080 3510.6280 3592.7015

4

Frequencies -- 63.8869 157.0220 172.3078
Frequencies -- 291.8564 298.7215 396.0947
Frequencies -- 463.8291 678.7086 769.4561
Frequencies -- 1005.0241 1087.9958 1220.4979
Frequencies -- 1326.5068 3593.0138 3700.8182

4^{+}

Frequencies -- 31.8564 53.6804 72.2222
Frequencies -- 103.5585 211.5266 280.1608
Frequencies -- 429.6990 506.3993 867.5765
Frequencies -- 1041.8119 1187.8285 1287.8500
Frequencies -- 1420.8083 3516.1252 3581.1552

5a

Frequencies -- 105.0749 131.4689 169.7809
Frequencies -- 241.6517 362.1941 391.8997
Frequencies -- 444.1047 641.5110 698.9933
Frequencies -- 772.5293 785.2089 1308.7128
Frequencies -- 1336.8097 3730.6512 3746.9883

5a^{+}

Frequencies -- 67.9006 102.4318 217.8698
Frequencies -- 243.8415 254.0491 309.5820
Frequencies -- 422.5309 710.4226 773.9411
Frequencies -- 840.9846 909.9744 1384.4962
Frequencies -- 1452.9104 3628.3966 3651.4165

5b

Frequencies -- 109.2275 155.2277 193.9271
Frequencies -- 214.2241 295.6304 444.4318
Frequencies -- 464.6929 642.0062 691.4322
Frequencies -- 771.1373 777.4249 1312.8648
Frequencies -- 1327.3221 3739.4873 3744.0302

5b^{+}

Frequencies -- 59.2203 95.9129 192.5930
Frequencies -- 230.8520 289.7427 321.9494
Frequencies -- 498.7468 655.6430 737.3494

Frequencies -- 836.1720 893.4483 1378.6730
Frequencies -- 1455.6405 3614.8077 3653.5728

5c

Frequencies -- 113.6641 129.5440 160.5288
Frequencies -- 189.8103 290.8714 441.4571
Frequencies -- 469.8272 635.6254 684.7752
Frequencies -- 769.7596 774.0016 1313.7470
Frequencies -- 1314.5494 3745.9405 3748.4836

5c$^+$

Frequencies -- 78.9635 110.8998 157.8124
Frequencies -- 233.8051 257.7636 327.5657
Frequencies -- 508.8979 702.3648 744.9757
Frequencies -- 833.6973 870.3085 1382.4988
Frequencies -- 1445.0790 3627.6200 3652.5072

5d

Frequencies -- 118.4458 134.6993 150.8106
Frequencies -- 185.1268 361.1202 383.3985
Frequencies -- 433.6218 654.4137 700.5244
Frequencies -- 775.0557 782.8508 1318.9985
Frequencies -- 1324.9947 3742.5746 3743.8392

5d$^+$

Frequencies -- 67.6993 102.1984 217.8044
Frequencies -- 243.6584 254.1174 309.6130
Frequencies -- 422.5229 710.1725 774.3202
Frequencies -- 840.8881 910.2597 1384.5582
Frequencies -- 1452.8763 3628.6229 3651.5486

6a

Frequencies -- 110.7036 168.4028 286.8251
Frequencies -- 401.6739 550.9976 561.0174
Frequencies -- 574.6837 714.9108 948.8484
Frequencies -- 999.2192 1087.2708 1142.7013
Frequencies -- 1460.0738 2367.7303 3628.5442

6a$^+$

Frequencies -- 30.8535 64.4719 86.8914
Frequencies -- 124.9313 223.2043 320.3494
Frequencies -- 399.8285 498.4886 989.4983
Frequencies -- 1093.5288 1209.1995 1410.9512
Frequencies -- 1436.4578 2418.1953 3491.2697

6b

Frequencies --	88.6229	114.6059	266.6581
Frequencies --	382.2128	437.7214	534.4943
Frequencies --	557.1352	710.4615	938.5273
Frequencies --	988.2575	1095.9522	1182.8484
Frequencies --	1422.8163	2361.5889	3690.4043

6b^{+}

Frequencies --	30.8535	64.4719	86.8914
Frequencies --	124.9313	223.2043	320.3494
Frequencies --	399.8285	498.4886	989.4983
Frequencies --	1093.5288	1209.1995	1410.9512
Frequencies --	1436.4578	2418.1953	3491.2697

6c

Frequencies --	36.2602	115.9182	247.9272
Frequencies --	375.4808	497.7063	549.8263
Frequencies --	614.0918	682.7881	948.6107
Frequencies --	999.0828	1107.9308	1160.3897
Frequencies --	1415.0347	2363.3825	3658.2360

6c^{+}

Frequencies --	49.1204	88.0059	117.5213
Frequencies --	257.1829	269.4399	356.7282
Frequencies --	576.4416	705.7973	828.9232
Frequencies --	1074.3433	1132.0126	1246.1185
Frequencies --	1432.8647	2384.4748	3605.4030

6d

Frequencies --	36.2602	115.9182	247.9272
Frequencies --	375.4808	497.7063	549.8263
Frequencies --	614.0918	682.7881	948.6107
Frequencies --	999.0828	1107.9308	1160.3897
Frequencies --	1415.0347	2363.3825	3658.2360

6d^{+}

Frequencies --	49.1204	88.0059	117.5213
Frequencies --	257.1829	269.4399	356.7282
Frequencies --	576.4416	705.7973	828.9232
Frequencies --	1074.3433	1132.0126	1246.1185
Frequencies --	1432.8647	2384.4748	3605.4030

6e

Frequencies --	85.8707	121.9762	251.8670
Frequencies --	351.0979	443.3404	528.6825
Frequencies --	618.3355	741.9512	930.2131
Frequencies --	987.9240	1075.9535	1184.2655
Frequencies --	1399.8882	2370.1994	3695.1346

6e$^+$

Frequencies --	21.9053	91.1483	140.0095
Frequencies --	199.3905	287.8701	370.9229
Frequencies --	577.3273	739.0476	838.7609
Frequencies --	1075.1551	1142.6506	1246.5088
Frequencies --	1436.8469	2383.3286	3602.0359

6f

Frequencies --	72.9786	153.0209	280.0887
Frequencies --	385.3367	488.4188	522.3561
Frequencies --	658.4731	728.0808	942.0905
Frequencies --	1009.5876	1043.1709	1158.7265
Frequencies --	1446.1818	2349.7398	3656.4306

6f$^+$

Frequencies --	-8.4449	181.7050	201.8262
Frequencies --	238.5486	408.8212	493.4160
Frequencies --	513.5766	807.3641	1028.8480
Frequencies --	1188.8109	1214.2643	1430.6565
Frequencies --	1505.7936	2486.5612	3490.6217

6g

Frequencies --	75.1505	151.3676	237.5216
Frequencies --	411.3791	480.8373	523.8053
Frequencies --	548.5510	705.9397	931.3088
Frequencies --	994.8206	1085.0164	1180.4788
Frequencies --	1404.7475	2369.9449	3688.6154

6g$^+$

Frequencies --	47.1000	162.2527	215.7234
Frequencies --	342.1381	376.6264	468.1012
Frequencies --	523.6642	794.3531	1031.7626
Frequencies --	1184.6980	1224.6273	1430.9888
Frequencies --	1503.7397	2505.5952	3481.5650

6h

Frequencies --	82.9569	122.4511	258.4409

Frequencies --	358.0404	446.6147	538.7807
Frequencies --	615.6953	738.6218	930.1469
Frequencies --	991.0934	1077.3837	1183.4627
Frequencies --	1404.7457	2355.8283	3697.3471

6i

Frequencies --	80.6266	105.2587	290.5287
Frequencies --	347.4994	416.3250	526.5522
Frequencies --	628.3322	736.2080	927.8748
Frequencies --	991.1249	1084.3426	1183.4869
Frequencies --	1410.7416	2357.5961	3697.7897

6i^{+}/6h^{+}

Frequencies --	45.7584	162.2387	215.4240
Frequencies --	342.1844	376.7585	468.1345
Frequencies --	523.6512	794.4718	1031.7363
Frequencies --	1184.7022	1225.1325	1431.2601
Frequencies --	1503.7348	2505.7100	3481.5159

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