Electronic Supplementary Information (ESI)

to

Geometries, stabilities and fragmental channels of neutral and charged sulfur clusters: S_n^Q (n = 3-20, Q = 0, ±1)

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In order to check the validity of the computational method, we first carry out calculations on S_2, S_2^-, S_2^+ and S_6 clusters with a large variety of theoretical methods (HF[1], MP2[2], B3LYP[3,4], PW91[3,5], PBE[6], B3P86[4] and B3PW91[3,5,7]) with the 6-311+G* basis set. The calculated results are summarized in Table S1. As evidenced from Table S1, B3P86 and B3PW91 give results in close agreement for both bond length (r) and vibrational frequency (ω) for the considered clusters, when compared to the experimental values. However, the calculated dissociation energy (D) and adiabatic ionization potential (AIP), adiabatic electron affinity (AEA) of S_2, vertical detachment energy (VDE) of S_2^- and AIP of S_6 at B3PW91 level are in better agreement with experimental values, with deviation less than 4%, 4%, 4%, 4% and 2%, respectively. On this basis, the B3PW91/6-311+G* method is selected for determination of the lowest-energy structures of sulfur clusters.

To further confirm the reliability of the B3PW91/6-311+G* method, the relative energies of conformers of neutral and charged S_8 are obtained by the DFT calculations and listed in Table S2, with those obtained by the MP2 calculations for comparison. Their geometries are depicted in Fig. S1. It can be seen that the B3PW91/6-311+G* method gives the exact ground-state structure of S_8. In addition, based on the isodesmic reaction,

$$(m / 8)S_8 \rightarrow S_m, \quad (m = 5, 6, 7) \tag{1}$$

the thermodynamic heats of formation for neutral S_8 are calculated. The results are listed in Table S3, along with the previously theoretical and experimental data for comparison. From Table S3, our calculated heats of formation for S_8 are in good agreement with the available theoretical and experimental results, which mean that the present theoretical method is reliable.

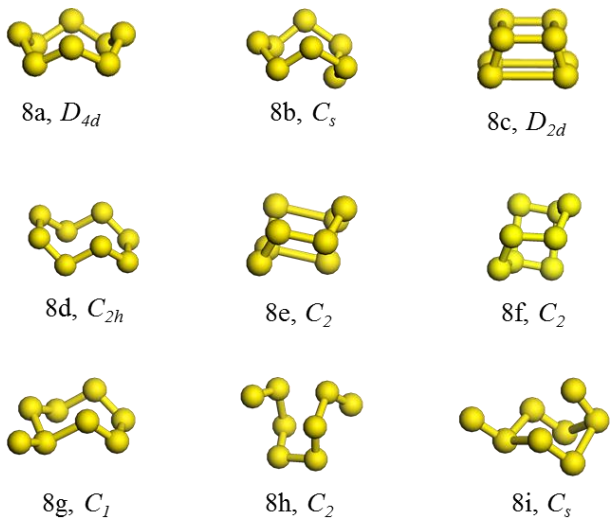

Fig. S1. The lowest-energy and low-lying structures of S_8 together with the point symmetry.

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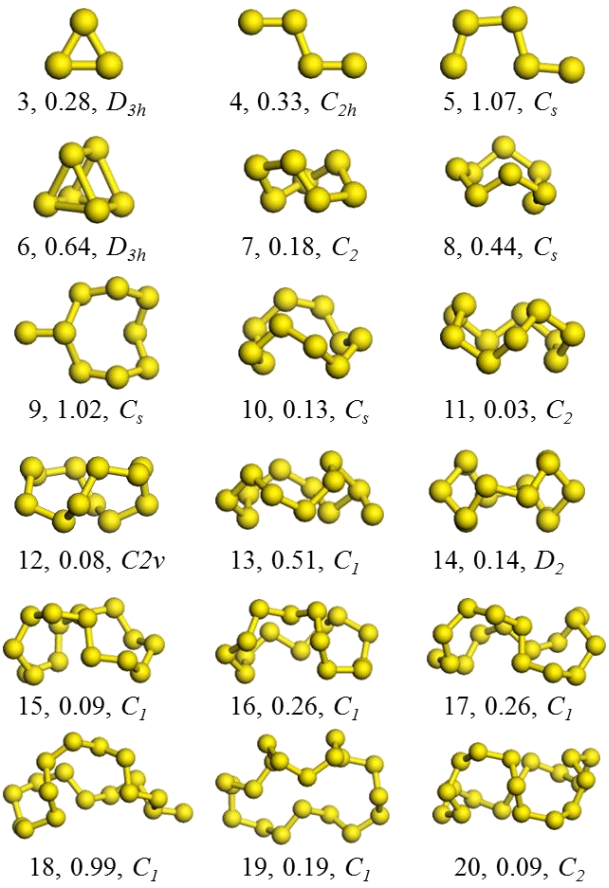

Fig. S2. The low-lying isomers of neutral S_n (n = 3-20) clusters together with the relative energy (eV) and point symmetry.

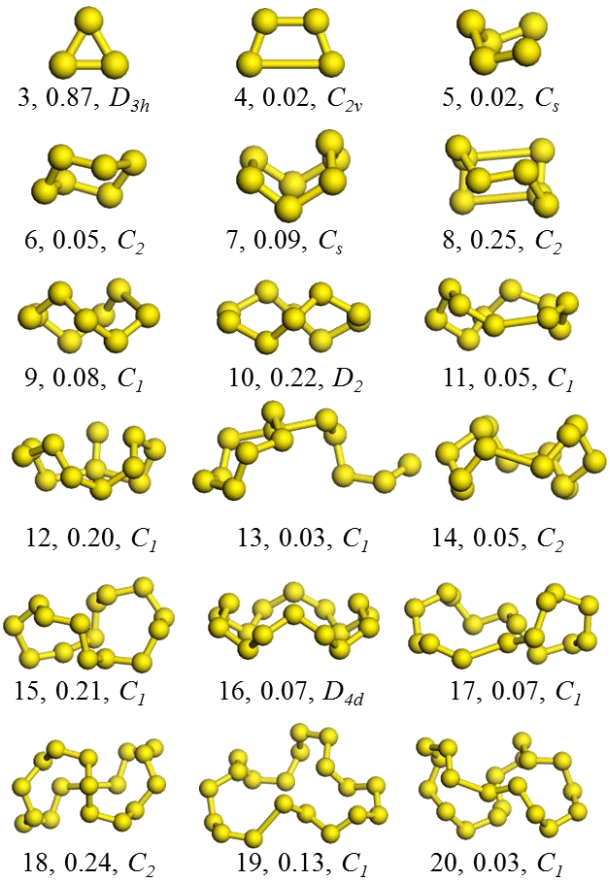

Fig. S3. The low-lying isomers of anionic S_n^- (n = 3-20) clusters together with the relative energy (eV) and point symmetry. S_3^- with D_{3h} symmetry has two imaginary frequencies.

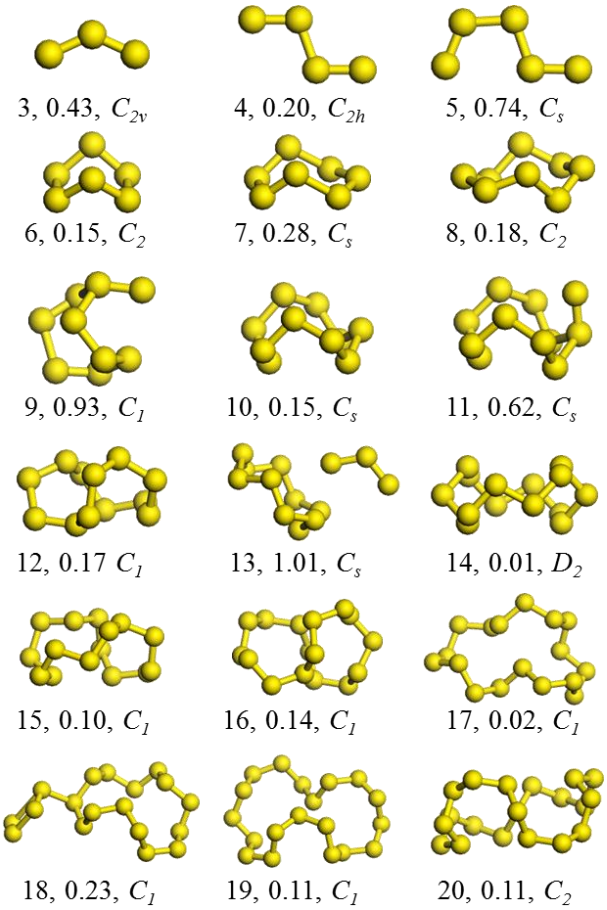

Fig. S4. The low-lying isomers of cationic S_n^+ (n = 3-20) clusters together with the relative energy (eV) and point symmetry.

Table S1. Calculated bond length r (Å), bond angle θ (°), dihedral angle ϕ (°), vibrational frequency ω (cm^{-1}), the dissociation energy D (eV), adiabatic ionization potential AIP (eV), vertical detachment energies VDE (eV) of S_2, S_2^-, S_2^+ and S_6 at different levels together with their corresponding experimental values.

	Property	HF	MP2	B3LYP	PW91	PBE	B3P86	**B3PW91**	**Experiment**
S_2	r	1.879	1.920	1.927	1.937	1.936	1.913	**1.914**	**1.889**[a]
	ω	792	678	684	665	667	707	**706**	**726**[a], **725**[b]
	D	1.72	1.69	4.08	4.61	4.61	4.32	**4.19**	**4.37**[a]
	AIP	9.41	9.53	9.67	9.59	9.54	10.25	**9.72**	**9.36**[c], **9.30**[d]
	AEA	1.02	1.04	1.79	1.70	1.66	2.30	**1.74**	**1.67**[e], **1.66**[f]
S_2^-	r	2.008	2.035	2.054	2.059	2.058	2.034	**2.035**	**2.005**[e]
	ω	624	553	537	526	528	562	**561**	**589**[g], **570**[e],
	VDE	1.27	1.36	1.97	1.86	1.82	2.48	**1.92**	**1.84**[b]
S_2^+	r	1.786	1.866	1.840	1.855	1.855	1.829	**1.831**	**1.825**[i],
	ω	951	705	805	774	777	830	**828**	**790**[a],**807**[k,l],
S_6	r	2.078	2.087	2.120	2.123	2.121	2.096	**2.099**	**2.068**[m]
	θ	102.9	102.8	103.1	103.2	103.2	102.9	**103.0**	**102.6**[m]
	ϕ	73.3	73.4	73.0	72.8	72.8	73.2	**73.1**	**73.8**[m]
	ω_1	174	167	159	156	156	163	**163**	**180**[n]
	ω_2	229	204	195	181	182	197	**197**	**203**[n]
	ω_3	278	267	252	229	231	261	**260**	**265**[n]
	ω_4	350	318	305	250	251	310	**309**	**312**[n]
	ω_5	508	396	307	293	294	334	**332**	**390**[n]
	ω_6	512	454	403	381	382	428	**426**	**451**[n]
	ω_7	513	458	428	418	420	448	**447**	**462**[n]
	ω_8	527	475	450	444	445	469	**468**	**477**[n]
	AIP	9.08	9.09	8.78	8.53	8.48	9.38	**8.82**	**9.00**[c]

[a] Refer. 8 [b] Refer. 9. [c] Refer. 10. [d] Refer. 11. [e] Refer. 12. [f] Refer. 13. [g] Refer. 14. [h] Refer. 15. [i] Refer. 16. [j] Refer. 17. [k] Refer. 18. [l] Refer. 19. [m] Refer. 20. [n] Refer. 21.

Table S2. Calculated relative conformational energies of S_8 at different levels.

Isomers	Relative conformational energies						
	MP2	B3LYP	PW91	PBE	M062X	B3P86	B3PW91
8a	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8b	0.42	0.43	0.44	0.44	0.36	0.43	0.44
8c	1.64	0.63	0.28	0.30	0.78	0.62	0.65
8d	1.02	0.64	0.49	0.50	0.69	0.66	0.68
8e	2.33	0.71	0.34	0.35	0.96	0.69	0.72
8f	2.40	1.09	0.80	0.82	1.21	1.08	1.11
8g	2.20	1.13	0.83	0.84	1.34	1.18	1.19
8h	5.00	1.55	1.14	1.16	1.66	1.58	1.60
8i	6.87	1.68	1.06	1.08	2.36	1.74	1.77

Table S3. Calculated relative conformational energies of anionic S_8^- at different levels.

Isomers	Relative conformational energies						
	MP2	B3LYP	PW91	PBE	M062X	B3P86	B3PW91
8a	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8b	0.40	0.42	0.44	0.41	0.31	0.40	0.40
8c	0.98	0.48	0.36	0.34	0.39	0.44	0.46
8d	0.69	0.47	0.43	0.41	0.43	0.47	0.48
8e	0.11	0.05	0.12	0.09	0.22	0.03	0.04
8f	0.18	0.14	0.19	0.16	0.14	0.11	0.12
8g	0.23	0.30	0.44	0.41	0.11	0.33	0.32
8h	0.80	0.06	0.00	0.01	0.14	0.17	0.17
8i	0.98	0.59	0.21	0.21	1.26	0.77	0.77

Table S4. Calculated relative conformational energies of cationic S_8^+ at different levels.

Isomers	Relative conformational energies						
	MP2	B3LYP	PW91	PBE	M062X	B3P86	B3PW91
8a	8.10	9.67	9.66	9.54	9.90	10.93	9.80
8b	7.46	9.15	9.21	9.09	9.24	10.37	9.24
8c	0.00	0.00	0.00	0.00	0.00	0.00	0.00
8d	8.06	9.35	9.33	9.22	9.55	10.59	9.46
8e	7.92	9.87	9.66	9.55	10.11	11.08	9.96
8f	9.34	10.02	9.87	9.76	10.25	11.24	10.12
8g	7.78	9.82	9.81	9.69	10.03	11.09	9.95
8h	9.16	10.24	10.09	10.00	10.65	11.60	10.46
8i	11.39	10.45	10.01	9.91	11.30	11.87	10.73

Table S5. The heat of formation (298 K) of homocycle S_8.

Method	Heat of Formation (kcal/mol)		
	$S_8 \rightarrow 8/5\ S_5$	$S_8 \rightarrow 8/6\ S_6$	$S_8 \rightarrow 8/7\ S_7$
B3PW91	**18.5**	**9.2**	**7.1**
B3LYP[a]	17.8	8.8	6.3
MP2[a]	18.3	7.9	6.7
MP2[b]	18.2	7.2	6.5
Exp.[c]	12.8±3.4	6.26±0.33	5.77±0.31
Exp.[d]	14.3	6.2	5.7
Exp.[e]	10.9	6.1	5.9

[a] Refer. 22. [b] Refer. 23. [c] Refer. 24. [d] Refer. 25. [e] Refer. 26.

Table S6. The vibrational frequency of S_n (n = 3-20) clusters.

Clusters	Vibrational frequency
S_3	264, 582, 667
S_4	87, 213, 324, 328, 644, 672
S_5	93, 225, 279, 292, 323, 327, 418, 493, 501
S_6	163, 163, 197, 197, 261, 310, 332, 427, 427, 447, 447, 468
S_7	53, 127, 149, 167, 192, 233, 263, 288, 336, 354, 366, 441, 471, 512, 521
S_8	74, 74, 144, 144, 191, 191, 214, 239, 247, 247, 367, 399, 399, 452, 452, 455, 455, 463
S_9	49, 65, 80, 92, 143, 153, 173, 212, 214, 247, 251, 300, 372, 374, 408, 416, 441, 444, 446, 464, 471
S_{10}	41, 47, 65, 84, 98, 122, 137, 162, 212, 214, 223, 238, 244, 254, 357, 388, 389, 439, 448, 448, 453, 468, 475, 476
S_{11}	33, 41, 50, 63, 86, 108, 117, 142, 167, 180, 201, 215, 231, 243, 246, 271, 361, 369, 404, 413, 439, 446, 448, 451, 462, 471, 476
S_{12}	42, 42, 43, 43, 68, 77, 102, 144, 156, 156, 178, 178, 239, 239, 250, 250, 272, 288, 363, 384, 384, 422, 422, 443, 448, 449, 451, 451, 460, 460
S_{13}	33, 35, 48, 48, 62, 62, 76, 111, 124, 149, 166, 169, 180, 211, 231, 236, 237, 242, 253, 299, 353, 365, 388, 396, 419, 435, 443, 449, 450, 453, 464, 475, 484
S_{14}	18, 37, 42, 47, 58, 58, 66, 75, 122, 130, 149, 157, 168, 185, 185, 220, 230, 233, 240, 243, 254, 274, 359, 376, 376, 410, 414, 438, 442, 449, 450, 452, 455, 458, 463, 466
S_{15}	16, 22, 24, 31, 37, 59, 60, 62, 91, 93, 94, 123, 154, 156, 171, 193, 201, 212, 230, 237, 244, 244, 258, 264, 367, 368, 392, 395, 422, 424, 444, 445, 448, 452, 457, 459, 461, 472, 477
S_{16}	15, 15, 29, 29, 52, 52, 53, 53, 60, 74, 86, 128, 128, 161, 169, 169, 189, 189, 203, 203, 240, 240, 243, 243, 258, 263, 361, 374, 374, 403, 403, 430, 430, 445, 450, 450, 454, 454, 458, 458, 461, 461
S_{17}	16, 17, 20, 26, 33, 44, 47, 55, 58, 68, 72, 86, 112, 117, 152, 163, 175, 177, 181, 192, 219, 220, 235, 246, 251, 257, 264, 276, 362, 370, 386, 389, 414, 417, 437, 439, 441, 447, 448, 452, 454, 458, 465, 468, 473
S_{18}	11, 18, 23, 26, 32, 39, 43, 50, 55, 58, 67, 71, 108, 132, 134, 136, 154, 157, 175, 187, 208, 210, 217, 225, 228, 229, 237, 239, 248, 248, 360, 368, 375, 396, 396, 420, 423, 440, 443, 450, 451, 452, 456, 460, 462, 463, 464, 477
S_{19}	7, 14, 17, 23, 30, 33, 35, 45, 50, 54, 58, 69, 79, 85, 111, 117, 145, 146, 165, 173, 187, 188, 212, 225, 236, 243, 253, 256, 258, 263, 272, 295, 365, 367, 381, 386, 408, 409, 429, 432, 439, 441, 448, 449, 450, 451, 458, 458, 460, 465, 466
S_{20}	14, 16, 21, 23, 35, 38, 43, 47, 53, 61, 66, 68, 72, 87, 99, 115, 125, 145, 155, 160,169, 180, 189, 210, 216, 225, 228, 232, 234, 240, 244, 256, 258, 263, 361, 369, 372, 389, 393, 413, 414, 432, 434, 445, 448, 452, 453, 454, 457, 459, 460, 463, 464, 469

Table S7 The vibrational frequency of S_n^- (n = 3–20) clusters.

Clusters	Vibrational frequency
S_3^-	226, 510, 545
S_4^-	47, 110, 191, 415, 537, 560
S_5^-	43, 56, 121, 161, 226, 304, 417, 533, 550
S_6^-	35, 43, 79, 102, 192, 217, 245, 270, 414, 430, 532, 543
S_7^-	57, 65, 82, 139, 160, 163, 204, 206, 240, 330, 349, 421, 441, 517, 521
S_8^-	55, 59, 62, 84, 145, 162, 171, 195, 204, 226, 233, 347, 360, 410, 423, 436, 499, 500
S_9^-	46, 49, 50, 54, 73, 107, 136, 145, 161, 221, 243, 248, 284, 346, 354, 392, 415, 439, 443, 514, 515
S_{10}^-	34, 40, 52, 53, 68, 85, 115, 128, 149, 166, 181, 225, 249, 253, 280, 346, 349, 389, 410, 425, 437, 445, 483, 509,
S_{11}^-	28, 33, 36, 58, 64, 68, 80, 120, 150, 155, 165, 182, 188, 213, 232, 238, 259, 332, 337, 382, 408, 419, 431, 440, 454, 509, 511
S_{12}^-	24, 27, 33, 47, 56, 68, 75, 87, 108, 122, 144, 156, 174, 186, 222, 232, 239, 253, 274, 341, 357, 375, 403, 416, 431, 437, 447, 457, 480, 501
S_{13}^-	20, 25, 26, 43, 51, 57, 57, 65, 83, 111, 136, 142, 157, 168, 178, 206, 232, 236, 239, 243, 291, 339, 343, 366, 392, 414, 421, 433, 447, 452, 461, 504, 509
S_{14}^-	15, 28, 32, 37, 43, 48, 51, 66, 78, 88, 118, 146, 155, 160, 176, 177, 183, 218, 222, 227, 235, 238, 245, 337, 346, 371, 394, 412, 420, 432, 441, 447, 448, 459, 509, 513
S_{15}^-	15, 20, 25, 28, 33, 44, 48, 53, 62, 71, 78, 101, 117, 129, 143, 163, 175, 184, 217, 221, 238, 239, 261, 269, 283, 332, 349, 370, 387, 403, 414, 424, 436, 437, 445, 453, 454, 497, 504
S_{16}^-	13, 20, 22, 27, 36, 42, 45, 51, 62, 67, 74, 85, 121, 127, 139, 153, 166, 170, 179, 193, 211, 220, 226, 231, 234, 248, 259, 334, 347, 370, 384, 403, 413, 423, 435, 444, 448, 449, 451, 457, 486, 500
S_{17}^-	13, 15, 17, 20, 30, 38, 42, 43, 52, 55, 67, 77, 81, 95, 112, 134, 141, 163, 170, 177, 186, 198, 226, 226, 237, 241, 257, 260, 272, 335, 340, 371, 382, 398, 410, 420, 430, 437, 439, 440, 449, 461, 465, 492, 507
S_{18}^-	9, 13, 22, 25, 28, 32, 37, 41, 53, 59, 62, 67, 81, 97, 104, 121, 123, 141, 156, 163, 170, 182, 201, 202, 217, 229, 235, 240, 251, 257, 265, 285, 287, 362, 370, 385, 405, 422, 438, 442, 444, 449, 451, 460, 467, 486, 560, 619
S_{19}^-	13, 17, 19, 24, 28, 32, 39, 47, 53, 56, 61, 72, 74, 82, 89, 112, 124, 146, 155, 163, 170, 173, 176, 194, 207, 217, 222, 228, 233, 236, 242, 245, 262, 279, 340, 352, 374, 385, 397, 416, 422, 433, 436, 445, 447, 450, 453, 455, 464, 509, 517
S_{20}^-	12, 17, 22, 25, 28, 32, 35, 38, 44, 49, 52, 54, 58, 67, 74, 88, 98, 127, 135, 154, 157, 164, 167, 177, 184, 191, 215, 229, 231, 238, 242, 245, 258, 267, 283, 346, 353, 362, 370, 388, 400, 410, 421, 430, 437, 440, 442, 446, 449, 449, 457, 460, 509, 520

Table S8. The vibrational frequency of S_n^+ (n = 3–20) clusters.

Clusters	Vibrational frequency
S_3^+	227, 425, 641
S_4^+	74, 152, 246, 300, 690, 726
S_5^+	34, 218, 274, 308, 331, 351, 446, 472, 508
S_6^+	76, 76, 198, 198, 257, 288, 341, 423, 423, 444, 444, 445
S_7^+	40, 102, 141, 144, 179, 217, 232, 277, 312, 337, 417, 418, 465, 468, 480
S_8^+	35, 82, 86, 106, 135, 148, 169, 197, 234, 281, 299, 361, 372, 393, 494, 511, 524, 546
S_9^+	31, 59, 71, 84, 115, 152, 162, 183, 204, 211, 226, 279, 344, 355, 384, 394, 440, 458, 465, 475, 499
S_{10}^+	33, 64, 71, 73, 86, 120, 134, 155, 199, 210, 210, 223, 236, 240, 340, 362, 381, 418, 442, 454, 459, 468, 473, 490
S_{11}^+	32, 39, 49, 53, 66, 101, 113, 138, 162, 174, 184, 192, 217, 243, 255, 269, 309, 332, 379, 397, 434, 442, 449, 457, 464, 465, 490
S_{12}^+	28, 28, 34, 41, 64, 69, 98, 111, 140, 143, 152, 176, 194, 213, 228, 239, 260, 276, 331, 353, 355, 405, 410, 436, 439, 449, 457, 458, 458, 465
S_{13}^+	5, 30, 41, 43, 54, 56, 70, 101, 105, 122, 153, 163, 174, 203, 221, 224, 233, 241, 243, 278, 290, 322, 357, 384, 419, 427, 444, 448, 450, 453, 457, 463, 473
S_{14}^+	18, 26, 28, 32, 35, 67, 68, 79, 82, 94, 114, 134, 155, 179, 183, 192, 199, 213, 219, 225, 235, 245, 343, 357, 360, 373, 410, 442, 443, 446, 461, 462, 471, 481, 482, 491
S_{15}^+	12, 16, 28, 29, 41, 49, 59, 61, 66, 92, 101, 125, 143, 145, 157, 177, 179, 180, 187, 201, 217, 219, 245, 247, 326, 338, 357, 366, 400, 430, 438, 443, 449, 454, 456, 463, 464, 473, 486
S_{16}^+	18, 18, 26, 26, 43, 43, 55, 56, 56, 75, 87, 128, 128, 156, 161, 161, 169, 169, 177, 177, 191, 191, 227, 227, 247, 252, 339, 352, 352, 381, 381, 418, 418, 443, 453, 456, 459, 459, 460, 460, 463, 463
S_{17}^+	17, 21, 30, 39, 42, 45, 52, 66, 70, 86, 103, 118, 124, 131, 148, 151, 166, 189, 198, 206, 212, 217, 227, 238, 244, 257, 274, 321, 336, 357, 370, 394, 406, 432, 440, 447, 449, 453, 454, 459, 462, 466, 473, 480
S_{18}^+	23, 25, 31, 35, 40, 42, 51, 53, 56, 58, 76, 77, 96, 116, 132, 137, 157, 157, 165, 182, 189, 190, 215, 221, 227, 232, 234, 245, 246, 247, 331, 347, 362, 380, 392, 414, 424, 441, 442, 449, 449, 454, 456, 457, 461, 465, 467, 506
S_{19}^+	12, 14, 14, 23, 24, 32, 37, 46, 48, 48, 58, 71, 75, 94, 111, 115, 138, 142, 149, 170, 174, 179, 200, 204, 216, 225, 231, 242, 244, 255, 271, 280, 339, 348, 361, 364, 384, 400, 423, 426, 439, 444, 447, 449, 450, 452, 455, 456, 457, 459, 460
S_{20}^+	10, 13, 15, 21, 28, 31, 32, 35, 41, 45, 54, 57, 64, 77, 92, 106, 119, 119, 139, 150, 156, 168, 174, 191, 197, 205, 210, 221, 226, 233, 241, 244, 259, 288, 340, 349, 355, 370, 377, 393, 413, 423, 435, 443, 445, 449, 451, 452, 456, 459, 463, 469, 469, 477

TABLE S9. The bond number (N) and bond length (r) of S_n (n = 3–20) clusters.

Sys.	N	r (Å)	Sys.	N	r (Å)	Sys.	N	r (Å)
S_3	2	1.937	S_{14}	2	2.107	S_{19}	2	2.094
S_4	1	2.200		2	2.080		2	2.091
	2	1.921		2	2.092		2	2.081
S_5	2	2.112		2	2.091		2	2.101
	2	2.052		2	2.090		2	2.079
	1	2.226		2	2.096		2	2.091
S_6	6	2.099		2	2.078		2	2.088
S_7	2	2.079	S_{15}	2	2.069		2	2.087
	2	2.013		2	2.083		1	2.091
	2	2.143		2	2.092		2	2.091
	1	2.236		2	2.092	S_{20}	1	2.100
S_8	8	2.088		2	2.081		1	2.084
S_9	2	2.112		2	2.111		1	2.086
	2	2.073		2	2.086		1	2.098
	2	2.085		1	2.130		1	2.093
	1	2.099	S_{16}	16	2.090		1	2.094
	2	2.096	S_{17}	2	2.093		1	2.079
S_{10}	2	2.076		2	2.088		1	2.100
	4	2.106		2	2.096		1	2.108
	4	2.083		2	2.083		1	2.078
S_{11}	1	2.080		2	2.087		1	2.105
	1	2.085		2	2.086		1	2.081
	1	2.090		2	2.099		1	2.089
	1	2.107		2	2.092		1	2.101
	1	2.085		1	2.082		1	2.094
	1	2.099	S_{18}	2	2.085		1	2.083
	1	2.123		2	2.101		1	2.084
	1	2.087		2	2.089		1	2.097
	1	2.088		2	2.091		1	2.085
	1	2.071		2	2.094		1	2.083
	1	2.085		2	2.086			
S_{12}	12	2.090		2	2.088			
S_{13}	2	2.102		2	2.091			
	2	2.084		2	2.099			
	2	2.101						
	2	2.092						
	2	2.046						
	2	2.082						
	1	2.177						

TABLE S10. The bond number (N) and bond length (r) of S_n^- (n = 3–20) clusters.

Sys.	N	r (Å)	Sys.	N	r (Å)		N	r (Å)	Sys.	N	r (Å)	Sys.	N	r (Å)
S_3^-	2	2.026	S_{12}^-	1	2.027	S_{15}^-	1	2.020	S_{17}^-	1	2.022	S_{19}^-	1	2.195
S_4^-	1	2.009		2	2.117		1	2.135		1	2.123		1	2.085
	2	2.168		1	2.100		1	2.097		1	2.100		1	2.118
S_5^-	2	2.139		1	2.106		1	2.092		1	2.098		1	2.098
	2	2.008		1	2.087		1	2.097		1	2.091		1	2.094
S_6^-	2	2.008		1	2.100		1	2.078		1	2.093		1	2.089
	2	2.081		1	2.092		1	2.102		1	2.093		1	2.083
	1	2.192		1	2.102		1	2.089		1	2.095		1	2.106
S_7^-	2	2.016		1	2.073		1	2.097		1	2.086		1	2.140
	2	2.147		1	2.157		1	2.095		1	2.091		1	2.012
	2	2.095		1	2.031		1	2.091		1	2.088		1	2.022
S_8^-	1	2.101	S_{13}^-	1	2.023		1	2.110		1	2.095		1	2.099
	2	2.107		1	2.099		1	2.122		1	2.094		1	2.089
	2	2.114		1	2.098		1	2.024		1	2.101		1	2.092
	2	2.030		1	2.066	S_{16}^-	1	2.115		1	2.127		2	2.094
S_9^-	2	2.014		1	2.140		1	2.101		1	2.024		1	2.073
	2	2.150		1	2.094		1	2.114	S_{18}^-	1	2.052		1	2.095
	2	2.084		1	2.020		1	2.090		1	2.108	S_{20}^-	1	2.011
	2	2.101		1	2.072		1	2.026		1	2.092		1	2.092
S_{10}^-	1	2.087		1	2.147		1	2.115		1	2.095		1	2.088
	1	2.106		1	2.113		1	2.094		1	2.090		1	2.081
	1	2.101		1	2.116		1	2.095		1	2.092		1	2.097
	1	2.088		1	2.088		1	2.098		1	2.093		1	2.095
	1	2.100	S_{14}^-	1	2.109		2	2.089		1	2.086		1	2.085
	1	2.033		1	2.087		1	2.092		1	2.095		1	2.106
	1	2.123		1	2.083		1	2.100		1	2.096		1	2.088
	1	2.019		1	2.097		1	2.087		1	2.087		1	2.103
	1	2.140		1	2.093		1	2.031		1	2.098		1	2.080
S_{11}^-	1	2.019		1	2.019					1	2.063		1	2.149
	1	2.124		1	2.019					1	2.639		1	2.093
	1	2.110		1	2.127					1	1.989		1	2.089
	1	2.092		1	2.096					1	2.268		1	2.097
	1	2.100		1	2.089					1	1.953		1	2.107
	1	2.088		1	2.121								1	2.113
	1	2.092		1	2.100								1	2.022
	1	2.099		1	2.101								1	2.104
	1	2.139												
	1	2.021												

TABLE S11. The bond number (N) and bond length (r) of S_n^+ (n = 3–20) clusters.

Sys.	N	r (Å)	Sys.	N	r (Å)	Sys.	N	r (Å)	Sys.	N	r (Å)
S_3^+	2	2.029	S_{12}^+	4	2.085	S_{16}^+	16	2.087	S_{19}^+	1	2.092
	1	2.186		4	2.080	S_{17}^+	1	2.071		1	2.087
S_4^+	1	2.392		4	2.090		1	2.096		1	2.085
	2	1.878	S_{13}^+	1	2.096		1	2.086		1	2.084
S_5^+	2	2.070		1	2.085		1	2.096		1	2.088
	2	2.116		1	2.108		1	2.133		1	2.085
	1	2.055		1	2.080		1	2.064		1	2.093
S_6^+	6	2.082		1	2.064		1	2.065		1	2.083
S_7^+	2	2.113		1	2.078		1	2.088		1	2.084
	1	2.063		1	2.110		1	2.100		1	2.088
	2	2.065		1	2.087		1	2.083		1	2.088
	2	2.091		1	2.062		1	2.092		1	2.081
S_8^+	2	2.184		1	2.115		1	2.088		1	2.080
	2	2.022		1	2.096		1	2.120		1	2.099
	2	1.987		1	2.085		1	2.092		1	2.085
	1	2.233		1	2.086		1	2.097		2	2.087
	1	2.166	S_{14}^+	1	2.125		1	2.083		1	2.086
S_9^+	1	2.144		1	2.045		1	2.087		1	2.090
	2	2.045		1	2.085	S_{18}^+	2	2.084	S_{20}^+	1	2.067
	2	2.137		1	2.098		2	2.093		1	2.114
	2	2.054		1	2.085		2	2.078		1	2.063
	2	2.094		1	2.074		2	2.094		1	2.128
S_{10}^+	2	2.104		1	2.111		2	2.095		1	2.061
	4	2.072		1	2.083		2	2.082		1	2.100
	4	2.092		1	2.108		2	2.097		1	2.087
S_{11}^+	1	2.085		1	2.138		2	2.093		1	2.088
	1	2.095		1	2.058		2	2.086		1	2.084
	1	2.101		1	2.058					1	2.088
	1	2.073		1	2.070					1	2.096
	1	2.095		1	2.097					1	2.081
	1	2.080	S_{15}^+	3	2.083					1	2.089
	1	2.075		2	2.089					1	2.087
	1	2.075		2	2.092					1	2.089
	1	2.098		2	2.074					1	2.079
	1	2.108		2	2.111					1	2.098
	1	2.082		2	2.062					1	2.083
				1	2.112					1	2.082
				1	2.088					1	2.100

TABLE S12. The geometric symmetry, electronic state, total energy (E), binding energy per atom (E_b), and the HOMO–LUMO energy gaps (E_{gap}) of S_n clusters (n = 3-20).

Clusters	Symm.	State	E (Hartree)	E_b (eV)	E_{gap} (eV)
S_3	C_{2v}	$^1A_1$	-1194.46	2.20	2.76
S_4	C_{2v}	$^1A_1$	-1592.63	2.31	2.14
S_5	C_s	$^1A'$	-1990.82	2.45	3.64
S_6	D_{3d}	$^1A_{1g}$	-2389.00	2.54	4.37
S_7	C_s	$^1A'$	-2787.17	2.55	4.27
S_8	D_{4d}	$^1A_1$	-3185.35	2.60	4.73
S_9	C_2	^1A	-3583.51	2.56	4.27
S_{10}	D_2	^1A	-3981.68	2.56	3.92
S_{11}	C_1	^1A	-4379.85	2.56	4.07
S_{12}	D_{3d}	$^1A_{1g}$	-4778.03	2.59	4.37
S_{13}	C_2	^1A	-5176.18	2.56	4.04
S_{14}	C_s	$^1A'$	-5574.36	2.57	4.13
S_{15}	C_2	^1A	-5972.52	2.57	4.04
S_{16}	D_{4d}	$^1A_1$	-6370.70	2.58	4.06
S_{17}	C_2	^1A	-6768.86	2.58	4.07
S_{18}	C_2	^1A	-7167.03	2.57	3.97
S_{19}	C_2	^1A	-7565.20	2.58	4.07
S_{20}	C_1	^1A	-7963.37	2.57	4.12

TABLE S13. The geometric symmetry, electronic state, total energy (E), binding energy per atom (E_b), and the HOMO–LUMO energy gaps (E_{gap}) of S_n^- clusters (n = 3-20).

Clusters	Symm.	State	E (Hartree)	E_b (eV)	E_{gap} (eV)
S_3^-	C_{2v}	$^2B_1$	-1194.56	2.38	2.09
S_4^-	C_{2h}	^2B_g	-1592.73	2.44	1.75
S_5^-	C_2	^2B	-1990.90	2.46	1.15
S_6^-	C_2	^2A	-2389.07	2.50	1.03
S_7^-	C_s	$^2A"$	-2787.25	2.53	1.94
S_8^-	C_2	^2B	-3185.42	2.56	1.86
S_9^-	C_2	^2B	-3583.59	2.55	1.92
S_{10}^-	C_1	^2A	-3981.75	2.55	1.91
S_{11}^-	C_1	^2A	-4379.92	2.55	1.87
S_{12}^-	C_1	^2A	-4778.09	2.56	1.91
S_{13}^-	C_1	^2A	-5176.26	2.57	1.87
S_{14}^-	C_1	^2A	-5574.43	2.57	1.90
S_{15}^-	C_1	^2A	-5972.61	2.58	1.80
S_{16}^-	C_1	^2A	-6370.77	2.57	1.79
S_{17}^-	C_1	^2A	-6768.94	2.58	1.67
S_{18}^-	C_1	^2A	-7167.11	2.58	1.69
S_{19}^-	C_1	^2A	-7565.28	2.58	1.63
S_{20}^-	C_1	^2A	-7963.45	2.58	1.58

TABLE S14. The geometric symmetry, electronic state, total energy (E), binding energy per atom (E_b), and the HOMO–LUMO energy gaps (E_{gap}) of S_n^+ clusters (n = 3-20).

Clusters	Symm.	State	E (Hartree)	E_b (eV)	E_{gap} (eV)
S_3^+	C_{2v}	$^2B_1$	-1194.12	2.59	1.77
S_4^+	C_{2v}	$^2B_1$	-1592.31	2.74	1.95
S_5^+	C_s	$^2A''$	-1990.51	2.86	2.06
S_6^+	D_{3d}	$^2A_{1g}$	-2388.68	2.83	1.61
S_7^+	C_2	^2B	-2786.86	2.83	1.65
S_8^+	C_2	^2A	-3185.03	2.80	1.49
S_9^+	C_2	^2A	-3583.20	2.80	1.37
S_{10}^+	D_2	$^2B_3$	-3981.38	2.80	1.29
S_{11}^+	C_1	^2A	-4379.54	2.77	1.33
S_{12}^+	C_{2h}	^2B_u	-4777.72	2.76	1.09
S_{13}^+	C_1	^2A	-5175.88	2.74	0.88
S_{14}^+	C_1	^2A	-5574.06	2.74	0.92
S_{15}^+	C_2	^2B	-5972.23	2.73	0.76
S_{16}^+	D_{4d}	$^2B_2$	-6370.40	2.73	0.91
S_{17}^+	C_1	^2A	-6768.56	2.71	1.09
S_{18}^+	C_2	^2B	-7166.73	2.71	1.36
S_{19}^+	C_1	^2A	-7564.91	2.71	0.81
S_{20}^+	C_1	^2A	-7963.08	2.70	0.59

Table S15. Cartesian coordinates of ground state S_n (n = 3-20) clusters reported in Fig. 1.

S_3

S	0.00000000	0.00000000	0.66125300
S	0.00000000	1.66425700	-0.33062700
S	0.00000000	-1.66425700	-0.33062700

S_4

S	0.00000000	1.09996000	0.93175600
S	0.00000000	-1.09996000	0.93175600
S	0.00000000	-1.56734400	-0.93175600
S	0.00000000	1.56734400	-0.93175600

S_5

S	-1.06329700	-1.34152400	0.00000000
S	0.26582400	-0.67246000	1.49936700
S	0.26582400	1.34322200	1.11277200
S	0.26582400	1.34322200	-1.11277200
S	0.26582400	-0.67246000	-1.49936700

S_6

S	0.00000000	1.89639100	0.44947800
S	-1.64232300	0.94819600	-0.44947800
S	-1.64232300	-0.94819600	0.44947800
S	0.00000000	-1.89639100	-0.44947800
S	1.64232300	-0.94819600	0.44947800
S	1.64232300	0.94819600	-0.44947800

S_7

S	-0.86554200	-1.84378800	1.11800300
S	1.15515700	0.77456900	1.67371700
S	1.15515700	0.77456900	-1.67371700
S	-0.86554200	-1.84378800	-1.11800300
S	-0.86554200	0.06553300	1.75721300
S	1.15185500	2.00737300	0.00000000
S	-0.86554200	0.06553300	-1.75721300

S_8

S	0.00000000	2.40604400	0.49164000
S	-2.40604400	0.00000000	0.49164000
S	2.40604400	0.00000000	0.49164000
S	-1.70133000	1.70133000	-0.49164000
S	0.00000000	-2.40604400	0.49164000

S	1.70133000	-1.70133000	-0.49164000
S	-1.70133000	-1.70133000	-0.49164000
S	1.70133000	1.70133000	-0.49164000

S_9

S	0.00000000	2.68183500	0.56727200
S	0.07688900	1.04672000	-2.40102900
S	1.19840600	-1.82781700	-0.94784200
S	-1.15321900	-1.18789400	1.42597800
S	-1.19840600	1.82781700	-0.94784200
S	-0.07688900	-1.04672000	-2.40102900
S	0.00000000	0.00000000	2.71124200
S	1.15321900	1.18789400	1.42597800
S	0.00000000	-2.68183500	0.56727200

S_{10}

S	2.83477900	0.01396600	1.03776700
S	-2.83477900	0.01396600	-1.03776700
S	2.83477900	-0.01396600	-1.03776700
S	-2.83477900	-0.01396600	1.03776700
S	1.22986300	1.22758900	1.65815700
S	-1.22986300	1.22758900	-1.65815700
S	1.22986300	-1.22758900	-1.65815700
S	0.00000000	0.00000000	-2.80723800
S	0.00000000	0.00000000	2.80723800
S	-1.22986300	-1.22758900	1.65815700

S_{11}

S	0.17542200	2.90956800	0.65248300
S	1.41252300	-1.62426600	-1.38224600
S	-2.81911600	1.48293300	-0.38397100
S	1.46664900	1.44510400	1.36946100
S	-2.54604400	-1.73284500	0.79925200
S	-2.77471400	-0.53726300	-0.89651300
S	2.89986900	1.20771100	-0.17889300
S	-0.97556400	2.29535600	-0.97448200
S	0.57449900	-2.88421200	0.05964400
S	-0.54336700	-1.73018300	1.38816600
S	3.12984300	-0.83190200	-0.45290100

S_{12}

S	-1.97696900	2.17664200	1.69859300
S	0.00000000	0.00000000	3.39788400
S	1.97696900	-2.17664200	1.69859300

S	1.97696900	-2.17664200	-1.69859300
S	0.00000000	0.00000000	-3.39788400
S	-1.97696900	2.17664200	-1.69859300
S	0.00000000	-1.69266800	-2.17236800
S	0.00000000	-1.69266800	2.17236800
S	0.00000000	1.69266800	-2.17236800
S	2.52443800	-1.08951600	0.00000000
S	-2.52443800	1.08951600	0.00000000
S	0.00000000	1.69266800	2.17236800

S_{13}

S	1.25648200	1.20595600	2.17846700
S	1.89215800	-2.40127900	-0.90700600
S	1.72422400	-0.32864200	-1.20956100
S	-1.89215800	2.40127900	-0.90700600
S	0.00000000	3.09209100	-0.34083100
S	-1.08441800	-0.09182700	-3.10681300
S	-1.25648200	-1.20595600	2.17846700
S	0.24871100	2.99412200	1.72399400
S	0.00000000	-3.09209100	-0.34083100
S	-1.72422400	0.32864200	-1.20956100
S	1.08441800	0.09182700	-3.10681300
S	-0.24871100	-2.99412200	1.72399400
S	0.00000000	0.00000000	3.32350100

S_{14}

S	-0.50120900	-1.84449500	2.07390000
S	-2.14120100	1.17729500	-1.71574500
S	-2.14338000	-0.69987200	-2.63840600
S	2.25554500	1.59821200	1.69352700
S	1.03997100	1.74839000	0.00000000
S	2.25554500	1.59821200	-1.69352700
S	-2.14120100	1.17729500	1.71574500
S	1.11019100	-1.22818500	3.28354100
S	2.56766200	-0.44799900	-2.02529900
S	-2.14338000	-0.69987200	2.63840600
S	-0.50120900	-1.84449500	-2.07390000
S	-3.33518600	1.14170000	0.00000000
S	2.56766200	-0.44799900	2.02529900
S	1.11019100	-1.22818500	-3.28354100

S_{15}

S	-0.06546200	2.47241700	-3.19739600
S	2.15013100	-2.07230600	2.69264200

S	-2.15013100	2.07230600	2.69264200
S	0.06546200	-2.47241700	-3.19739600
S	1.95418200	-3.03565800	-0.46975200
S	0.00000000	0.00000000	4.31046200
S	0.81253100	-0.68838400	-3.93245800
S	-0.81253100	0.68838400	-3.93245800
S	0.00000000	2.57161300	-1.11788200
S	-2.53315500	1.44114900	0.73517900
S	-0.13639200	1.71780300	3.13443600
S	0.13639200	-1.71780300	3.13443600
S	2.53315500	-1.44114900	0.73517900
S	0.00000000	-2.57161300	-1.11788200
S	-1.95418200	3.03565800	-0.46975200

S_{16}

S	1.66961300	4.03080200	0.00000000
S	-1.66961300	-4.03080200	0.00000000
S	-4.03080200	1.66961300	0.00000000
S	4.03080200	-1.66961300	0.00000000
S	4.03080200	1.66961300	0.00000000
S	1.66961300	-4.03080200	0.00000000
S	-4.03080200	-1.66961300	0.00000000
S	3.43249600	0.00000000	1.10517800
S	2.42714100	2.42714100	-1.10517800
S	0.00000000	3.43249600	1.10517800
S	-2.42714100	2.42714100	-1.10517800
S	-3.43249600	0.00000000	1.10517800
S	-2.42714100	-2.42714100	-1.10517800
S	0.00000000	-3.43249600	1.10517800
S	2.42714100	-2.42714100	-1.10517800
S	-1.66961300	4.03080200	0.00000000

S_{17}

S	-0.35566300	3.43360200	-2.56505800
S	-1.35503200	-2.86337400	3.04227600
S	0.35566300	-3.43360200	-2.56505800
S	1.35503200	2.86337400	3.04227600
S	1.07423500	4.68695200	0.18661300
S	0.00000000	0.00000000	4.00550500
S	-1.07423500	-4.68695200	0.18661300
S	0.00000000	3.70804600	1.68397600
S	1.41080200	3.42729400	-1.44197100
S	1.33054700	-1.65822600	-2.05714500
S	-1.41080200	-3.42729400	-1.44197100

S	0.00000000	-3.70804600	1.68397600
S	-1.49758800	-0.80273300	2.77287800
S	1.49758800	0.80273300	2.77287800
S	0.99194800	-0.31501500	-3.62432200
S	-0.99194800	0.31501500	-3.62432200
S	-1.33054700	1.65822600	-2.05714500

S_{18}

S	0.00000000	3.82471000	0.36072100
S	-0.09030200	-4.48738100	-1.63106300
S	0.83615800	1.43682600	-2.85429200
S	-3.01399200	-0.73270800	2.22337900
S	-0.83615800	-1.43682600	-2.85429200
S	-0.60012900	2.95626500	-2.87264000
S	1.52385300	-0.72273100	2.32500500
S	-1.52385300	0.72273100	2.32500500
S	3.01399200	0.73270800	2.22337900
S	1.99002600	3.72828100	0.97505800
S	0.09030200	4.48738100	-1.63106300
S	2.84095400	1.87374400	0.48378800
S	0.00000000	-3.82471000	0.36072100
S	-2.84095400	-1.87374400	0.48378800
S	0.60012900	-2.95626500	-2.87264000
S	-1.99002600	-3.72828100	0.97505800
S	0.00000000	0.00000000	-1.58092000
S	0.00000000	0.00000000	3.56100800

S_{19}

S	-0.77066000	-4.95182600	1.00777100
S	-0.76495400	3.86532200	1.92862200
S	-1.17700500	-1.26652400	2.92135700
S	1.40714300	3.78104100	-0.60370200
S	-1.45650200	1.64732600	-3.29079300
S	1.17700500	1.26652400	2.92135700
S	0.00000000	-2.87320600	3.59009700
S	1.03560600	-0.14426500	-1.90335300
S	-1.03560600	0.14426500	-1.90335300
S	1.45650200	-1.64732600	-3.29079300
S	-0.32035800	-4.39279600	-2.28252000
S	0.76495400	-3.86532200	1.92862200
S	1.54814500	-3.45481100	-2.25080300
S	-1.40714300	-3.78104100	-0.60370200
S	0.77066000	4.95182600	1.00777100
S	-1.54814500	3.45481100	-2.25080300

S	0.00000000	2.87320600	3.59009700
S	0.32035800	4.39279600	-2.28252000
S	0.00000000	0.00000000	1.76664800

S_{20}

S	-1.28690800	0.45940100	-1.83785300
S	0.92548200	3.74969800	-0.10602200
S	-3.39912400	-1.66549800	1.88430500
S	2.85438300	-0.46857200	2.18989200
S	-1.39180500	-2.61033400	-0.62078600
S	3.56838000	0.46294100	-1.00244300
S	-4.89000300	-0.77445500	0.71671100
S	1.66994400	0.85908500	3.26478900
S	-3.37579600	0.49437300	-2.04661800
S	0.06545200	3.38783100	1.75955300
S	-0.52519600	2.17322400	-2.74633100
S	-0.65189400	3.82864400	-1.48712100
S	1.87065700	-3.40309900	-0.29769800
S	2.57024100	-2.58887400	-2.09888300
S	-4.14303000	0.96902000	-0.15737100
S	-0.05981900	1.35079300	2.17805300
S	0.41023500	-2.04801300	0.31818100
S	4.23745400	-1.39681100	-1.72741600
S	-2.67324400	-3.33309000	0.84666500
S	4.22459200	0.55373800	0.97039200

Table S16. Cartesian coordinates of ground state S_n^- (n = 3-20) clusters reported in Fig. 2.

S_3^-

S	0.00000000	0.00000000	0.71485100
S	0.00000000	1.71854200	-0.35742600
S	0.00000000	-1.71854200	-0.35742600

S_4^-

S	1.02697000	2.35554300	0.00000000
S	1.02697000	0.34652300	0.00000000
S	-1.02697000	-0.34652300	0.00000000
S	-1.02697000	-2.35554300	0.00000000

S_5^-

S	0.00000000	0.00000000	1.09353600
S	0.00000000	1.66438300	-0.25038300
S	0.00000000	-1.66438300	-0.25038300
S	1.77265300	2.60678500	-0.29638500
S	-1.77265300	-2.60678500	-0.29638500

S_6^-

S	0.60443100	2.20220200	-0.67091500
S	-0.60443100	3.69225100	-0.07819400
S	0.60443100	-3.69225100	-0.07819400
S	-0.60443100	-2.20220200	-0.67091500
S	-0.84324100	-0.70019800	0.74910900
S	0.84324100	0.70019800	0.74910900

S_7^-

S	-1.40122700	1.54057400	1.50678900
S	0.33034800	-1.39823200	1.72285400
S	0.33034800	-1.39823200	-1.72285400
S	-1.40122700	1.54057400	-1.50678900
S	0.33034800	0.71174100	2.12207400
S	1.48106300	-1.70816500	0.00000000
S	0.33034800	0.71174100	-2.12207400

S_8^-

S	-0.42518200	0.96034100	-2.19926200
S	-0.67275600	-2.29255600	-0.99185600
S	-0.20773100	2.47912500	0.92099800
S	0.42518200	-0.96034100	-2.19926200
S	-0.67275600	-1.24489700	2.27012000
S	0.67275600	1.24489700	2.27012000

S	0.20773100	-2.47912500	0.92099800
S	0.67275600	2.29255600	-0.99185600

S_9^-

S	1.22353100	-2.17073900	-0.92918400
S	1.13749300	1.24665400	1.37358600
S	0.00000000	2.85727100	0.69968300
S	-0.09951100	1.49161100	-2.45618200
S	0.09951100	-1.49161100	-2.45618200
S	-1.13749300	-1.24665400	1.37358600
S	0.00000000	-2.85727100	0.69968300
S	0.00000000	0.00000000	2.62419600
S	-1.22353100	2.17073900	-0.92918400

S_{10}^-

S	1.33884000	-1.52034300	1.21104800
S	-0.85333700	-1.63626000	-1.35201300
S	-2.69551600	-1.38769300	-0.37318900
S	-2.51477000	2.01353800	0.40419000
S	1.89399700	2.55434200	0.21949500
S	2.60979000	0.98482900	-0.82958400
S	-0.85077500	2.47068600	-0.67100100
S	3.07922500	-0.69804000	0.40546000
S	0.40057700	-2.80969200	-0.16425600
S	-2.40803200	0.02863300	1.14984900

S_{11}^-

S	-1.83977400	2.57084700	0.76581600
S	2.57731500	-0.82304700	-1.20257200
S	-3.28143700	-0.36553400	-0.57549400
S	-0.05784800	2.02425300	1.54138200
S	-1.11318600	-2.75539100	0.67426000
S	-1.92138700	-1.88114400	-1.05517700
S	1.93075900	2.47405000	-0.54339000
S	-2.36409300	1.48986700	-0.98555200
S	2.25138500	-2.13018900	0.40758900
S	0.48373000	-1.62808100	1.40702600
S	3.33453600	1.02436900	-0.43388800

S_{12}^-

S	2.64577400	0.55522300	-1.43633300
S	0.76918300	-2.61561000	1.36473100
S	-2.55300700	-1.20866200	-1.33414700
S	-3.57870000	0.68029000	-1.15638400

S	3.26216400	-1.29464100	-0.64104500
S	1.10742200	2.34171500	1.20489500
S	-2.46018000	-1.98410300	0.58627900
S	-2.61762400	1.80474400	0.23493000
S	2.83872900	2.05430800	0.02118500
S	1.63306300	-2.59519000	-0.54882100
S	-0.31510600	3.42346300	0.24892300
S	-0.73171700	-1.16153700	1.45578800

S_{13}^-

S	0.56968100	2.34280700	-1.35151500
S	1.99524100	-2.00822500	-1.43954200
S	0.07317800	-1.17658100	-1.57425000
S	-3.42043000	0.23095800	1.27208000
S	-3.22914600	1.21414600	-0.61900600
S	-1.62734400	-2.69291200	0.95410100
S	2.35539600	1.47793100	1.47621400
S	-2.12611200	2.90072800	-0.48717600
S	2.64962100	-1.80349600	0.54289000
S	-1.61045700	-0.74610400	1.66285800
S	-1.34341200	-2.62555500	-1.17267800
S	3.69034900	-0.01055600	0.79439400
S	2.02343500	2.89685900	-0.05837200

S_{14}^-

S	1.16421800	-1.13119700	-1.93943700
S	-2.65829900	-0.76233400	1.41767700
S	-3.69347800	-0.19775600	-0.29993600
S	3.33346900	0.33374700	1.66350100
S	0.56498800	1.05068000	2.18486600
S	0.21790700	2.88861800	1.42474200
S	0.14965100	-2.68755000	0.97279200
S	3.21980600	-0.80006500	-1.60095200
S	-0.03092600	2.83357200	-0.68652000
S	0.65101500	-2.94540900	-1.04539500
S	-2.42292300	0.63907300	-1.73669700
S	-1.92758700	-2.71499200	1.19367900
S	3.47340400	0.84396600	-0.28475700
S	-2.04124400	2.64964800	-1.26356400

S_{15}^-

S	2.55999100	-2.73204800	-0.33926200
S	-3.76526100	2.09481500	-0.28074500
S	3.29313600	2.49636800	0.31042600

S	-1.84853800	-3.09354600	0.27189200
S	-4.66146700	-1.19066600	0.15876100
S	-0.61651800	3.47941900	-0.04814600
S	-0.43593700	-1.62521900	0.78671500
S	0.94418900	-1.46249000	-0.75779400
S	3.78047900	-1.66272800	0.98100800
S	3.96895700	1.13818900	-1.02345700
S	0.58852000	2.44465200	1.20623800
S	-1.86862800	2.20870100	-1.19762300
S	-3.81922800	0.49835100	1.06901100
S	-3.20082900	-2.14310600	-1.00535700
S	5.08113200	-0.45069100	-0.13166600

S_{16}^-

S	-1.17023200	1.89444600	-1.54038200
S	1.76736600	-2.42248500	1.09369100
S	3.92295400	1.50746500	-0.21889800
S	3.65973800	0.52561700	1.53306000
S	0.61618600	-1.74625600	-2.09545900
S	-1.75390400	2.18082800	1.78205600
S	-2.72406900	-1.30580600	-1.29603900
S	-1.92941200	-1.21238700	2.02208400
S	0.35801600	3.29790600	-1.94987200
S	3.69045500	-1.58068900	1.34431100
S	-2.48019900	2.70784000	-0.11383000
S	1.60926800	-3.12228400	-0.86889400
S	-1.33651100	-2.46296600	-2.34411800
S	-2.86692200	0.59016200	2.55247400
S	-3.12293800	-2.13744000	0.58112300
S	1.76020300	3.28605000	-0.48130800

S_{17}^-

S	-3.75058100	-2.22520100	1.04573100
S	3.67886400	2.65649700	0.87525700
S	3.09284300	-2.83461000	-1.01516100
S	-3.05055500	3.07999000	-0.91860200
S	-5.28246800	0.47696500	-0.28231300
S	0.57935900	3.91221800	0.20000900
S	5.08012200	-0.37946900	0.13775000
S	-4.02200300	2.00395400	0.49435600
S	-4.18637900	-1.25162700	-0.75573900
S	1.23168000	-2.03073900	-1.54288000
S	3.69132400	-1.83193300	0.72230100
S	4.12018100	1.29741500	-0.66033700

S	1.65149600	2.53537700	1.40938300
S	-0.26979800	3.00005800	-1.39170400
S	-0.20740700	-3.42402700	-0.96097500
S	-0.56519200	-3.30712500	1.09625300
S	-1.79148400	-1.67774300	1.54667100

S_{18}^-

S	1.56243000	0.37917900	-1.51303700
S	5.70745000	1.63200800	1.44240900
S	-4.51414000	1.33437000	1.53056500
S	-0.13360100	2.53335300	0.55544100
S	-2.81718900	1.91386600	-1.40971200
S	-2.14371900	-2.05356100	-1.41147200
S	-0.44098300	-3.13518100	-0.79986300
S	-4.59984100	2.21264400	-0.36751300
S	-1.49836900	3.41082500	-0.77155600
S	-3.98510600	-2.06057500	1.50037700
S	6.06139800	-1.39594100	-0.09321600
S	-3.79031300	-2.87757400	-0.41863300
S	4.81882100	-0.31666000	-1.21061100
S	2.44328400	-1.07984200	-0.35143000
S	0.53825300	-2.08022200	0.66193500
S	-5.42697700	-0.54717700	1.48593300
S	1.63016900	2.22436000	-0.51732800
S	6.58843200	-0.09387000	1.68771100

S_{19}^-

S	-0.78795300	-0.88886700	1.24169400
S	-5.17414600	1.10879500	-0.45753100
S	2.68995500	1.93705400	1.02506900
S	3.80180300	0.51542600	-1.87751500
S	0.74155300	3.95441200	-0.98938600
S	-1.98111300	1.67448600	0.65036600
S	2.65154900	-2.66310700	-1.20437900
S	-2.73760900	-2.95339600	-0.64880000
S	2.32986400	-2.01684000	2.14806400
S	-4.27465300	-1.97747600	-1.67928900
S	2.26410400	3.92208500	0.51413600
S	4.28543100	-1.50995700	-1.84047700
S	-3.93739400	1.68892900	1.16121500
S	0.30717700	-2.35553200	2.45251800
S	-1.00759500	-1.77326400	-0.63326900
S	-1.06122000	4.26087500	-0.15037200
S	3.04818500	-3.37144500	0.71995800

S	-5.55063000	-0.95365800	-0.37164500
S	4.39269200	1.40148000	-0.06035600

S_{20}^-

S	-3.56802200	0.17169100	-2.25531200
S	3.52258700	2.85714800	-0.20430800
S	-2.06631100	3.45987200	-0.61243000
S	3.95969000	-0.35926500	0.80816000
S	1.65614700	-2.57397400	-2.51442200
S	0.50906800	2.00716900	1.06030000
S	-3.67494200	2.46285000	0.25359700
S	0.78858900	-2.27936500	1.91968800
S	-0.08334400	-2.46780200	-1.33163100
S	-0.92151300	-2.00094100	3.09349100
S	-3.76878700	-1.49797500	1.30310900
S	-4.75384100	1.51671500	-1.34589500
S	-2.13246300	-0.51815000	2.21267700
S	-3.30358200	-2.19855800	-0.53615400
S	4.79235600	1.56325800	0.83893100
S	2.82551400	-0.85248000	-2.34905600
S	1.98941600	3.50070500	1.05920200
S	4.42038700	-1.22931900	-1.03021700
S	-0.52062100	2.04897000	-0.74696800
S	0.32967100	-3.61054600	0.37723900

Table S17. Cartesian coordinates of ground state S_n^+ (n = 3-20) clusters reported in Fig. 3.

S_3^+			
S	0.00000000	0.00000000	1.13977600
S	0.00000000	1.09301500	-0.56988800
S	0.00000000	-1.09301500	-0.56988800
S_4^+			
S	0.00000000	1.19594900	0.91832400
S	0.00000000	-1.19594900	0.91832400
S	0.00000000	-1.58598200	-0.91832400
S	0.00000000	1.58598200	-0.91832400
S_5^+			
S	-0.98674800	-1.36181800	0.00000000
S	0.24668700	-0.68834100	1.51934400
S	0.24668700	1.36925000	1.02767100
S	0.24668700	1.36925000	-1.02767100
S	0.24668700	-0.68834100	-1.51934400
S_6^+			
S	0.00000000	1.94986700	0.36454300
S	-1.68863500	0.97493400	-0.36454300
S	-1.68863500	-0.97493400	0.36454300
S	0.00000000	-1.94986700	-0.36454300
S	1.68863500	-0.97493400	0.36454300
S	1.68863500	0.97493400	-0.36454300
S_7^+			
S	0.00000000	1.72827600	1.20111800
S	-0.02243100	1.03100500	-2.00101600
S	1.29989400	-1.51534700	-0.38922700
S	0.00000000	0.00000000	2.37824900
S	-1.29989400	1.51534700	-0.38922700
S	0.02243100	-1.03100500	-2.00101600
S	0.00000000	-1.72827600	1.20111800
S_8^+			
S	1.31962100	1.33595000	-0.72663900
S	0.26573800	1.08460000	2.43663200
S	-0.23187400	-1.05804000	-2.40884700
S	-0.23187400	1.91067500	0.69885500
S	0.23187400	-1.91067500	0.69885500

S	-1.31962100	-1.33595000	-0.72663900
S	-0.26573800	-1.08460000	2.43663200
S	0.23187400	1.05804000	-2.40884700

S_9^+

S	1.20264100	-1.75950000	-0.89778700
S	1.18209700	1.20485300	1.34923900
S	0.00000000	2.70545500	0.59336100
S	0.07320000	1.06972000	-2.33932000
S	-0.07320000	-1.06972000	-2.33932000
S	-1.18209700	-1.20485300	1.34923900
S	0.00000000	-2.70545500	0.59336100
S	0.00000000	0.00000000	2.58901600
S	-1.20264100	1.75950000	-0.89778700

S_{10}^+

S	2.88270800	0.10885800	1.04619200
S	-2.88270800	0.10885800	-1.04619200
S	2.88270800	-0.10885800	-1.04619200
S	-2.88270800	-0.10885800	1.04619200
S	1.18575900	1.20637500	1.50319000
S	-1.18575900	1.20637500	-1.50319000
S	1.18575900	-1.20637500	-1.50319000
S	0.00000000	0.00000000	-2.73410200
S	0.00000000	0.00000000	2.73410200
S	-1.18575900	-1.20637500	1.50319000

S_{11}^+

S	1.68851800	-2.13460500	1.10826800
S	-2.39463100	0.66980200	-1.21347900
S	3.13593400	0.44079400	-0.51789400
S	-0.37661900	-1.89556700	1.27245700
S	0.94045600	2.77431900	0.51626600
S	1.64120700	1.76374800	-1.15499800
S	-1.18796800	-2.39107900	-0.57203700
S	2.38502800	-1.48424700	-0.75690800
S	-2.35697200	1.90212500	0.48768700
S	-0.52776000	1.58563700	1.42984300
S	-2.94719300	-1.23092600	-0.59920600

S_{12}^+

S	0.00000000	0.00000000	3.59771500
S	0.00000000	2.73061700	1.73325100
S	0.00000000	2.73061700	-1.73325100

S	0.00000000	0.00000000	-3.59771500
S	0.00000000	-2.73061700	-1.73325100
S	0.00000000	-2.73061700	1.73325100
S	1.20739600	-1.13222700	-2.33010100
S	1.05380500	2.27015000	0.00000000
S	-1.05380500	-2.27015000	0.00000000
S	-1.20739600	1.13222700	-2.33010100
S	1.20739600	-1.13222700	2.33010100
S	-1.20739600	1.13222700	2.33010100

S_{13}^{+}

S	0.81274500	-2.16225800	-1.45806600
S	-2.59517100	0.91895700	-1.50994800
S	-0.50524900	1.17466300	-1.61764100
S	2.69347800	0.70910500	1.47894600
S	3.04994000	0.20681000	-0.50560800
S	0.45848500	3.11161800	1.00329300
S	-1.01805400	-2.05221400	1.48255600
S	2.78429500	-1.83871000	-0.82429500
S	-3.03246400	0.48719100	0.47713100
S	0.63794800	1.15725600	1.63603400
S	-0.18595000	3.12101100	-1.01132300
S	-2.94820500	-1.56786400	0.82387400
S	-0.15179700	-3.26556400	0.02504700

S_{14}^{+}

S	0.95087200	-0.89145400	-1.69193200
S	-2.49696600	-0.45603100	1.54921400
S	-3.60353700	0.49346500	0.05932000
S	3.14277300	0.16471600	1.65556700
S	1.10514200	0.54621800	1.72700600
S	0.75763700	2.56902300	1.36896600
S	-0.44221900	-2.97302200	0.60305200
S	2.92527200	-1.47694700	-1.16971100
S	0.44145900	2.82723100	-0.69885000
S	-0.15345400	-2.59996000	-1.48192900
S	-2.32525900	1.04896600	-1.49142200
S	-2.39704400	-2.48450800	1.02164700
S	3.68522800	0.26555700	-0.38184100
S	-1.58990500	2.96674700	-1.06908500

S_{15}^{+}

S	-0.65942800	2.44374600	-3.34941200
S	1.98370600	-2.27393800	2.72291500

S	-1.98370600	2.27393800	2.72291500
S	0.65942800	-2.44374600	-3.34941200
S	2.20887400	-3.28919500	-0.47493900
S	0.00000000	0.00000000	4.21166900
S	0.97310800	-0.41023700	-3.48568200
S	-0.97310800	0.41023700	-3.48568200
S	-0.34673000	2.96392400	-1.32751900
S	-2.60332900	1.69186600	0.81117400
S	0.00000000	1.69982800	2.99762800
S	0.00000000	-1.69982800	2.99762800
S	2.60332900	-1.69186600	0.81117400
S	0.34673000	-2.96392400	-1.32751900
S	-2.20887400	3.28919500	-0.47493900

S_{16}^{+}

S	1.68537300	4.06885000	0.00000000
S	-1.68537300	-4.06885000	0.00000000
S	-4.06885000	1.68537300	0.00000000
S	4.06885000	-1.68537300	0.00000000
S	4.06885000	1.68537300	0.00000000
S	1.68537300	-4.06885000	0.00000000
S	-4.06885000	-1.68537300	0.00000000
S	3.42657400	0.00000000	1.05079100
S	2.42295400	2.42295400	-1.05079100
S	0.00000000	3.42657400	1.05079100
S	-2.42295400	2.42295400	-1.05079100
S	-3.42657400	0.00000000	1.05079100
S	-2.42295400	-2.42295400	-1.05079100
S	0.00000000	-3.42657400	1.05079100
S	2.42295400	-2.42295400	-1.05079100
S	-1.68537300	4.06885000	0.00000000

S_{17}^{+}

S	-3.64602300	2.20447500	-0.84922100
S	3.75467200	-1.87293100	-1.26118200
S	1.77252700	-2.61706300	1.30490300
S	0.48763400	-1.10335500	0.52469600
S	-2.33064000	-2.91803400	0.22006800
S	-3.45462200	-1.39826000	1.10668500
S	2.68593400	1.32663000	-1.66091300
S	-1.16255400	1.39532400	1.24733000
S	-1.58198700	2.13437700	-0.69487300
S	4.35612600	0.06908300	-1.72715100
S	3.00694200	2.63030300	-0.06209000

S	2.19144900	1.81127400	1.68720100
S	0.31736900	2.66259500	2.00765000
S	-0.81943000	-1.92483900	-0.84705200
S	-5.01184100	-0.82528000	-0.15234100
S	-4.23078300	0.36942300	-1.67537500
S	3.66522600	-1.94372100	0.83166600

S_{18}^{+}

S	0.00000000	3.86237700	0.81111500
S	0.97056000	-4.29367500	-0.99275000
S	-0.58663800	1.50006000	-2.87447800
S	-2.98746400	-0.74530000	1.68143800
S	0.58663800	-1.50006000	-2.87447800
S	-1.91650400	2.56661100	-1.65786500
S	1.52015000	-0.71362900	1.91611400
S	-1.52015000	0.71362900	1.91611400
S	2.98746400	0.74530000	1.68143800
S	2.03750700	3.75758000	0.38792300
S	-0.97056000	4.29367500	-0.99275000
S	2.60534400	1.80675000	-0.08641500
S	0.00000000	-3.86237700	0.81111500
S	-2.60534400	-1.80675000	-0.08641500
S	1.91650400	-2.56661100	-1.65786500
S	-2.03750700	-3.75758000	0.38792300
S	0.00000000	0.00000000	-1.53520800
S	0.00000000	0.00000000	3.16504300

S_{19}^{+}

S	-4.66811800	1.59054500	-0.79660600
S	4.19594700	1.54973000	-0.83257200
S	-0.68110900	2.59519000	-1.14175400
S	3.69207700	-0.98165500	1.34630100
S	1.10640000	-3.29289100	-1.47002200
S	1.78842400	2.74198700	1.21716000
S	-2.12326700	3.66342700	-0.07471800
S	-0.54973800	-1.80708200	1.07289600
S	-0.29126500	-1.81726800	-1.01006300
S	-2.20040300	-3.00513600	1.50463100
S	-4.83422100	-1.74106400	-0.21090900
S	-3.45731300	2.29952200	0.75470600
S	-3.86904700	-1.75913100	1.63930900
S	-3.94252700	-0.27236700	-1.39360400
S	5.04166700	0.50790900	0.76322200
S	2.98501300	-2.39972700	-1.64528000

S	3.32584200	3.28990400	-0.08561400
S	4.01999900	-2.65641300	0.15239100
S	0.46163900	1.49451800	0.21052500

S_{20}^{+}

S	4.15844800	1.05790400	1.81827400
S	0.18947200	1.91299300	0.53136200
S	1.92088300	-1.76223300	0.04844800
S	-3.48778700	0.98082700	-1.26285200
S	-0.15791700	-3.05722400	-2.24533100
S	-3.41995900	0.27432300	2.03462600
S	3.86287500	-2.34798300	-0.41419100
S	-3.65108500	3.08845300	-1.31079300
S	5.72005700	0.17390600	0.74895000
S	-0.84980800	3.71387700	0.43439500
S	3.47251500	2.72578100	0.78490200
S	1.96606800	2.10657300	-0.53795800
S	-2.14243200	-3.92550000	0.32939200
S	-1.55687900	-2.60599600	1.83622500
S	4.90216100	-0.63102600	-0.99363900
S	-1.72066900	3.80956800	-1.47406400
S	-2.09315600	-2.89904200	-1.48874700
S	-3.31889600	-1.71718700	2.55532000
S	0.88674800	-1.32725900	-1.72198600
S	-4.68064000	0.42924500	0.32766700

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