Small chromium-doped silicon clusters $CrSi_n$: Structures, IR spectra, charge effect, magnetism and chirality

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Content:

- Mass spectrum of produced chromium doped silicon clusters (Figure S1).
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- Stable strucutres of cationic, neutral and anionic chromium doped silicon clusters $CrSi_n^{+/0}$ with n = 3-10 (Figures S2 to S14).
- IR spectra comparison of $CrSi_8^+$ and $CrSi_9^+$ clusters between calculation and experiment (Figure S15 and Figure S16).
- Calculated-IR and UV-Vis spectra of ground-state enantiomers $CrSi_n^{+/0/-}$ (n = 9 for cation, n = 7 for neutral and anion) (Figure S17).
- Calculated-ECD spectra of higher-energy enantiomers of cationic, neutral and anionic $CrSi_n^{+/0/-}$ clusters with n = 3-10 (Figure S18).

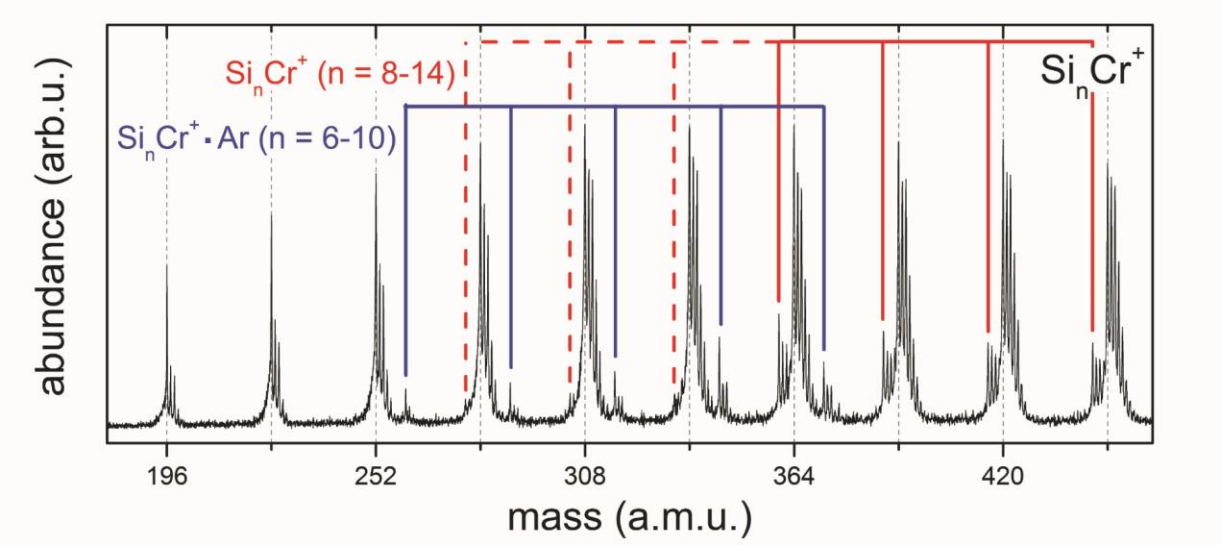


Figure S1. Mass spectrum of $CrSi_n^+$ clusters with the source at a temperature of 80 K and a 5 % argon mixture in the helium carrier gas. Argon complexes are found for $CrSi_n^+$.Ar (n = 6-10). For these sizes, no $CrSi_n^+$ clusters are observed in the mass spectrum. The exohedrally doped clusters react completely with the argon atoms. Starting from $CrSi_{11}Cr^+$, unreacted doped clusters appear in the mass spectrum. The highest peaks (on the grid lines) in the mass spectrum correspond to pure Si_n^+ clusters (note that Si and Cr have multiple isotopes, of which ^{28}Si and ^{52}Cr have the highest abundances).

Table S1. Relative energy comparison between B3P86/6-311+G(*d*) and B3P86/aug-cc-pVTZ theory levels of cationic, neutral and anionic $CrSi_n^{+/0/-}$ clusters (n = 3-10)

Isomer	Electronic state	6-311+G(*d*)	aug-cc-pVTZ	Isomer	Electronic state	6-311+G(*d*)	aug-cc-pVTZ
C3-iso1	$^4B_2$	0.00	0.00	**A9-iso1**	^6A	0.00	0.00
C3-iso2	$^4A_1$	0.33	0.33	**A9-iso2**	$^2A_1$	0.05	0.18
C3-iso3	$^4A'$	0.36	0.38	**N3-iso1**	$^5A_1$	0.00	0.00
C4-iso1	$^6A'$	0.00	0.00	**N3-iso1**	$^7A_1$	0.39	0.15
C4-iso2	$^6A_1$	0.22	0.24	**N3-iso2**	$^5A'$	0.50	0.37
C5-iso1	$^6A'$	0.00	0.00	**N4-iso1**	$^5B_2$	0.00	0.00
C5-iso2	$^4A''$	0.36	0.39	**N4-iso1**	^7A	0.31	0.32
C6-iso1	$^6A'$	0.00	0.01	**N4-iso2**	$^5A''$	0.29	0.25
C6-iso2	$^6A'$	0.03	0.00	**N4-iso3**	^7A	0.31	0.30
C7-iso1	$^6A_1$	0.00	0.00	**N5-iso1**	$^5B_1$	0.00	0.00
C7-iso2	^6A	0.16	0.15	**N5-iso2**	$^5A'$	0.09	0.08
C7-iso3	$^6A_1$	0.19	0.22	**N7-iso1**	$^5A'$	0.00	0.05
C8-iso1	$^4A''$	0.00	0.00	**N7-iso2**	^5A	0.02	0.01
C8-iso2	$^6A'$	0.06	0.04	**N7-iso3**	$^5A'$	0.06	0.00
C10-iso1	^6A	0.00	0.00	**N7-iso4**	$^5A'$	0.07	0.06
C10-iso2	$^6A'$	0.04	0.03	**N8-iso1**	$^5A''$	0.00	0.00
A6-iso1	$^4A_1$	0.00	0.00	**N8-iso2**	^5A	0.05	0.00
A6-iso2	^6A	0.32	0.33	**N8-iso3**	$^3A'$	0.06	0.01
A7-iso1	$^6A'$	0.00	0.04	**N8-iso4**	$^5A''$	0.16	0.18
A7-iso1	$^4A'$	0.01	0.04	**N9-iso1**	^5A	0.00	0.00
A7-iso2	^6A	0.05	0.00	**N9-iso2**	^5A	0.07	0.12
A8-iso1	$^6A_1$	0.00	0.00	**N10-iso1**	$^5A'$	0.00	0.00
A8-iso2	$^6A'$	0.18	0.23	**N10-iso2**	$^7A_1$	0.01	0.02
A8-iso3	$^6A'$	0.22	0.16	**N10-iso3**	$^5A'$	0.02	0.01

Table S2. Relative energy comparison between B3P86/6-311+G(*d*) and B3LYP/6-311+G(*d*) theory levels of anionic $CrSi_n^-$ cluster (n = 6-10)

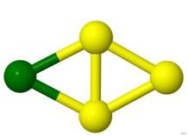
Isomer	B3P86/6-311+G(*d*)	B3LYP/6-311+G(*d*)	Isomer	B3P86/6-311+G(*d*)	B3LYP/6-311+G(*d*)
A6-iso1	0.00	0.00	**A8-iso1**	0.00	0.00
A6-iso2	0.32	0.46	**A8-iso2**	0.18	0.02
A7-iso1 ($^6A'$)	0.00	0.00	**A8-iso3**	0.22	0.13
A7-iso1 ($^4A'$)	0.01	0.03	**A9-iso1**	0.00	0.00
A7-iso2-L/R	0.05	0.10	**A9-iso2**	0.05	0.17

Table S3. Average binding energy, fragmentation energies and natural population analysis for the ground-states of the cationic, neutral, anionic $CrSi_n^{+/0-}$ clusters.

	Isomer	State	$D_{C/N}(Cr^+)$	$D_{N/A}(Cr)$	$D_{C/N/A}(Si)$	$E_{b\text{-}C/N/A}$	Atomic charge on Cr	Electron population on Cr's 3d shell	Electron population on Cr's 4s shell	Spin density of Cr	Total Wiberg bond of Cr	Total Wiberg bond of cleaved Si
cation	**C3-iso1**	$^4B_2$	4.77		5.02	2.40	0.83	4.86	0.30	4.54	1.36	2.32
	C4-iso1	$^6A'$	2.10		5.70	2.74	0.84	4.92	0.23	4.82	0.66	2.23
	C5-iso1	$^6A'$	2.58		5.98	2.95	0.77	4.90	0.29	4.62	1.04	2.79
	C6-iso1	$^6A'$	2.21		6.42	3.12	0.86	4.87	0.25	4.77	0.79	2.47
	C6-iso2	$^6A'$	2.19		6.55	3.11	0.82	4.94	0.21	4.73	0.74	2.41
	C7-iso1	$^6A_1$	2.32		6.20	3.27	0.83	4.96	0.20	4.87	0.55	2.83
	C7-iso2	^6A	2.20		6.01	3.25	0.82	4.96	0.20	4.74	0.72	2.72
	C7-iso3	$^6A_1$	2.13		6.08	3.25	0.82	4.92	0.26	5.01	0.44	2.76
	C8-iso1	$^4A''$	5.20		5.72	3.23	0.78	4.97	0.22	4.34	1.45	2.48
	C8-iso2	$^6A'$	2.95		5.26	3.22	0.72	4.94	0.28	4.69	0.94	2.82
	C9-iso1-L/R	^6A	2.42		6.33	3.33	0.75	5.03	0.18	4.54	0.91	2.76
	C10-iso1	^6A	2.52		7.07	3.42	0.82	4.96	0.20	4.71	0.74	2.98
	C10-iso2	$^6A'$	2.48		6.94	3.42	0.76	4.98	0.23	4.66	0.87	3.39
neutral	**N3-iso1**	$^5A_1$	6.78	3.69	4.96	2.59	0.66	4.94	0.38	4.67	1.22	2.74
	N3-iso1	$^7A_1$	6.63	2.02	5.52	2.54	0.60	5.00	0.38	4.98	0.69	3.02
	N4-iso1	$^5B_2$	6.76	2.87	5.62	2.89	0.61	4.98	0.33	4.56	1.32	2.65
	N5-iso1	$^5B_1$	7.27	4.22	6.07	3.12	0.58	4.99	0.36	4.47	1.50	2.85
	N5-iso2	$^5A'$	6.77	4.19	6.74	3.10	0.63	4.99	0.35	4.73	1.07	2.85
	N6-iso1	$^5B_2$	6.77	3.25	5.92	3.25	0.64	5.05	0.25	4.44	1.34	2.79
	N7-iso1	$^5A'$	7.44	4.14	5.67	3.26	0.44	5.04	0.40	4.32	1.76	2.63
	N7-iso2-L/R	^5A	6.81	3.65	5.63	3.25	0.61	4.98	0.35	4.48	1.43	2.82
	N7-iso3	$^5A'$	6.12	2.41	5.41	3.25	0.67	5.07	0.24	4.62	1.01	2.82
	N8-iso1	$^5A''$	6.40	3.85	5.67	3.30	0.58	5.14	0.22	4.31	1.38	2.73
	N8-iso2	$^5A''$	7.00	4.13	6.38	3.29	0.61	5.03	0.29	4.44	1.38	3.52
	N8-iso3	$^3A'$	9.74	7.10	5.89	3.25	0.27	5.23	0.35	3.90	2.45	2.65
	N9-iso1	^5A	6.82	4.15	6.40	3.36	0.56	5.10	0.23	4.18	1.60	3.02
	N10-iso1	$^5A'$	6.14	2.80	6.09	3.43	0.67	5.04	0.27	4.59	1.02	2.68
	N10-iso2	$^7A_1$	6.15	1.62	6.65	3.43	0.57	4.96	0.45	5.04	0.77	2.71
	N10-iso3	$^5A'$	6.13	2.89	6.30	3.43	0.60	5.09	0.27	4.53	1.16	3.07
anion	**A3-iso1**	$^6A_1$		3.06	5.69	2.80	0.30	4.99	0.66	5.08	1.04	2.96
	A4-iso1	$^4A_2$		5.34	6.15	3.00	0.42	5.09	0.40	4.38	1.81	2.78
	A5-iso1	$^6A'$		3.37	7.07	3.29	0.46	5.05	0.44	4.87	1.03	2.84
	A6-iso1	$^4A_1$		7.42	7.29	3.37	0.24	5.04	0.57	4.15	2.41	3.07
	A6-iso2	^6A		2.91	5.67	3.33	0.50	5.16	0.27	4.65	1.00	2.85
	A7-iso1	$^6A'$		4.40	6.39	3.38	0.28	5.14	0.41	4.51	1.48	3.06
	A7-iso1	$^4A'$		6.96	6.20	3.38	0.03	5.28	0.47	3.89	2.50	3.06
	A7-iso2-L/R	^6A		3.09	5.71	3.37	0.45	5.07	0.43	4.75	1.14	2.95
	A8-iso1	$^6A_1$		3.24	6.33	3.45	0.52	5.21	0.23	4.54	1.03	2.98
	A8-iso2	$^6A'$		2.92	6.68	3.43	0.47	4.97	0.55	5.03	0.94	3.04
	A8-iso3	$^6A'$		3.80	6.66	3.42	0.52	5.09	0.33	4.62	1.12	3.60
	A9-iso1	^6A		3.75	6.19	3.50	0.43	5.23	0.23	4.43	1.21	3.06
	A9-iso2	$^2A_1$		10.02	6.60	3.49	-1.73	6.90	0.47	0.96	4.26	3.13
	A10-iso1	^2A		10.73	6.33	3.55	-2.64	7.77	0.45	0.81	4.22	2.91

Table S4. The adiabatic ionization energies (IE_a) and the adiabatic electron affinities (AE_a) of neutral isomers having similar shape to the ground-state cationic and anionic structures.

Structure	IE_a (eV)	AE_a (eV)
C5-iso1	7.51	
A5-iso1		3.10
C6-iso1	7.03	
C6-iso2	7.07	
C7-iso1	6.30	
C7-iso2	6.50	
C7-iso3	6.15	
N7-iso2	7.09	2.90
N7-iso3	6.50	2.60
C8-iso2	6.65	
N8-iso2	7.34	3.11
N8-iso3	7.63	3.25
A8-iso2		3.57
C9-iso1	6.67	
N9-iso1	7.87	3.31
A9-iso2		3.52
N10-iso2	6.96	2.95
N10-iso3	6.63	2.98
A10-iso1		3.57

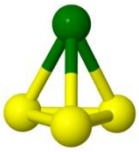


C3-iso1

[0.00; C_{2v}; $^4B_2$]

[0.79; C_{2v}; $^6A_1$]

[0.87; C_{2v}; $^8B_2$]

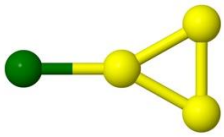


C3-iso2

[0.33; C_{3v}; $^4A_1$]

[0.57; C_{3v}; $^8A_1$]

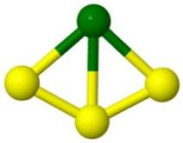
[0.96; C_1; ^2A]



C3-iso3

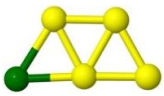
[0.36; C_s; $^4A'$]

[0.51; C_s; $^8A'$]



C3-iso4

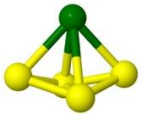
[0.51; C_s; $^6A'$]



C4-iso1

[0.00; C_s; $^6A'$]

[0.63; C_s; $^4A''$]

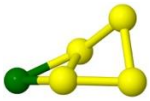


C4-iso2

[0.22; C_{2v}; $^6A_1$]

[0.46; C_s; $^4A'$]

[1.23; C_s; $^2A'$]



C4-iso3

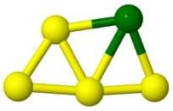
[0.61; C_1; ^4A]

C4-iso4

[0.78; C_s; $^6A''$]

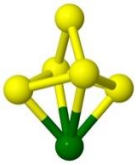
[0.99; C_1; ^8A]

[1.21; C_s; $^4A''$]



C4-iso5

[0.96; C_1; ^4A]



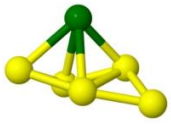
C5-iso1

[0.00; C_s; $^6A'$]

C5-iso2

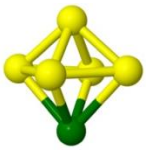
[0.36; C_s; $^4A''$]

[1.24; C_s; $^2A''$]



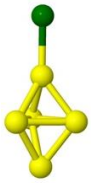
C5-iso3

[0.76; C_1; ^4A]



C5-iso4

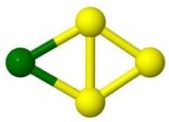
[0.79; C_{2v}; $^4B_1$]



C5-iso5

[0.94; C_1; ^4A]

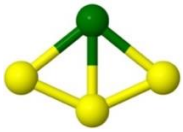
Figure S2. Stable isomers of the cationic $CrSi_n^+$ cluster (n = 3-5)



N3-iso1

[0.00; C_{2v}; $^5A_1$]

[0.15; C_{2v}; $^7A_1$]

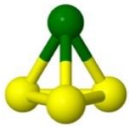


N3-iso2

[0.50; C_s; $^5A'$]

[0.89; C_s; $^7A'$]

[1.18; C_s; $^3A'$]

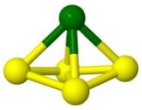


N3-iso3

[0.47; C_1; ^5A]

[0.63; C_1; ^7A]

[1.19; C_{3v}; $^1A_1$]

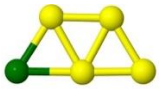


N4-iso1

[0.00; C_{2v}; $^5B_2$]

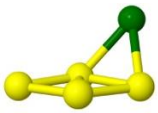
[0.31; C_1; ^7A]

[0.59; C_{2v}; $^3A_2$]



N4-iso2

[0.29; C_s; $^5A''$]



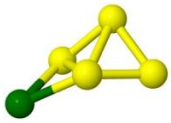
N4-iso3

[0.31; C_1; ^7A]

N4-iso4

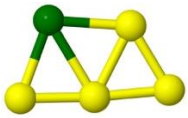
[0.38; C_s; $^7A'$]

[0.42; C_s; $^5A''$]



N4-iso6

[0.50; C_1; ^5A]

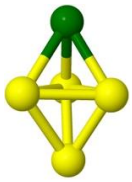


N4-iso5

[0.55; C_s; $^5A''$]

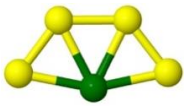
[0.74; C_s; $^3A''$]

[0.77; C_s; $^7A''$]



N4-iso7

[0.77; C_s; $^3A''$]



N4-iso8

[1.16; C_1; ^5A]

N5-iso1

[0.00; C_{2v}; $^5B_1$]

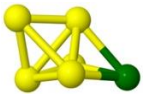
N5-iso2

[0.09; C_s; $^5A'$]

[1.43; C_s; $^3A'$]

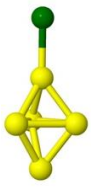
N5-iso3

[0.61; C_s; $^3A'$]



N5-iso4

[0.65; C_1; ^7A]

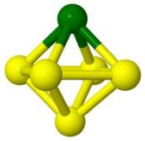


N5-iso5

[0.85; C_1; ^5A]

N5-iso6

[0.94; C_1; ^3A]



N5-iso7

[1.10; C_{4v}; $^3B_2$]

[1.38; C_{4v}; $^1A_1$]

N5-iso8

[1.28; C_s; $^5A''$]

[1.32; C_s; $^7A''$]

Figure S3. . Stable isomers of the neutral $CrSi_n$ cluster (n = 3-5)

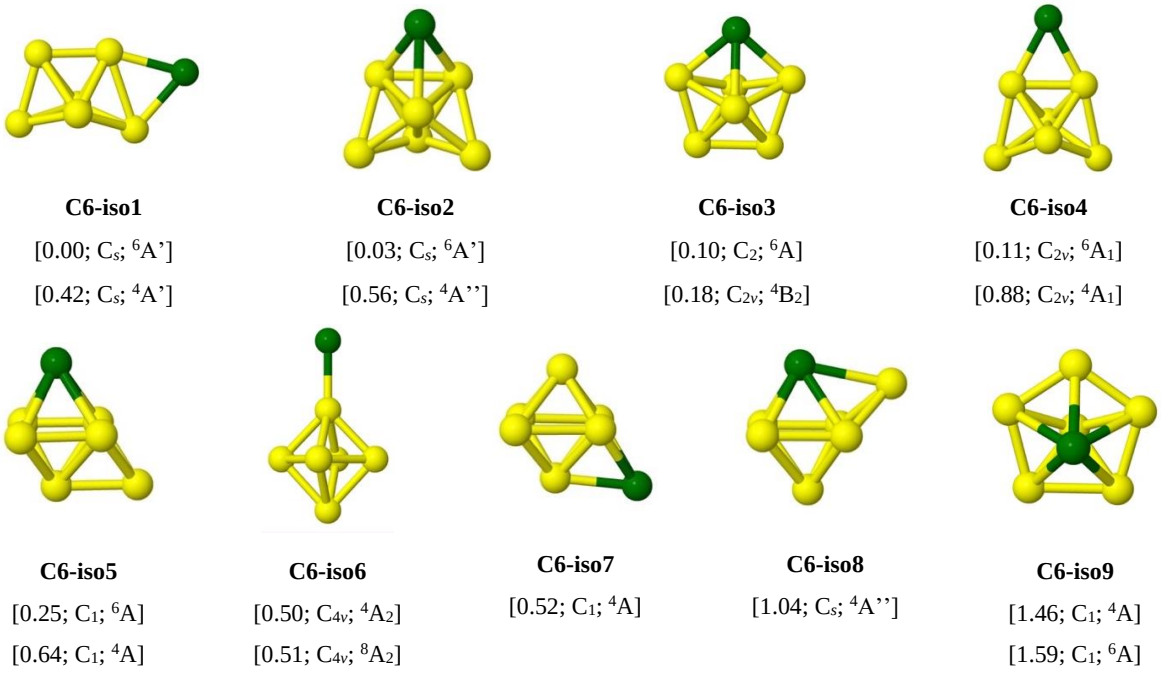


Figure S4. Stable isomers of the cationic $CrSi_6^+$ cluster.

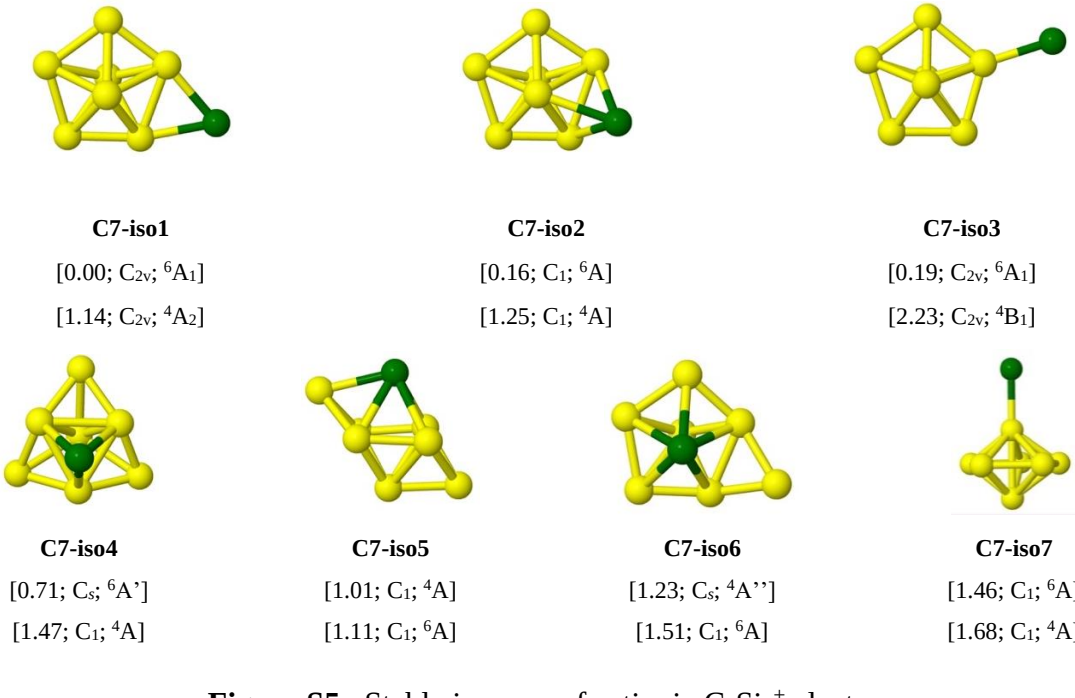
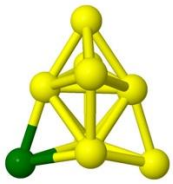


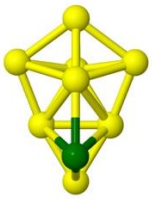
Figure S5. Stable isomers of cationic $CrSi_7^+$ cluster.



C8-iso1

[0.00; C_s, ^{4}A’’]

[0.43, C_1, ^{6}A/ 86*i* cm^{-1}]



C8-iso2

[0.06; C_s; ^{6}A’]

[0.34; C_s; ^{4}A’]

C8-iso3

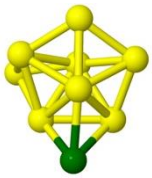
[0.18; C_s; ^{4}A’]

[0.53; C_s; ^{6}A’]

C8-iso4-L

[0.19; C_1; ^{4}A]

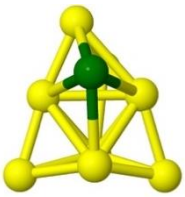
[0.39; C_1; ^{6}A]



C8-iso4-R

[0.19; C_1; ^{4}A]

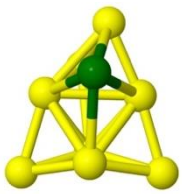
[0.39; C_1; ^{6}A]



C8-iso5-L

[0.23; C_1; ^{6}A]

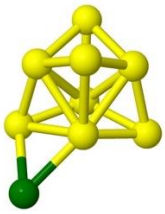
[0.31; C_s; ^{4}A’’]



C8-iso5-R

[0.23; C_1; ^{6}A]

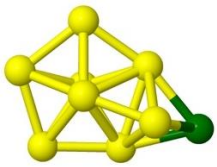
[0.31; C_s; ^{4}A’’]



C8-iso6

[0.24; C_1; ^{6}A]

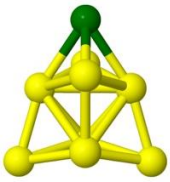
[0.49; C_1; ^{4}A]



C8-iso7

[0.25; C_s; ^{4}A’’]

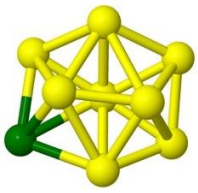
[0.53; C_1; ^{6}A]



C8-iso8

[0.26; C_s; ^{4}A’’]

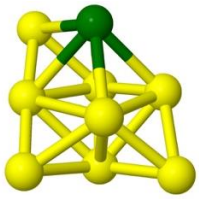
[0.34; C_1; ^{6}A]



C8-iso9

[0.28; C_1; ^{4}A]

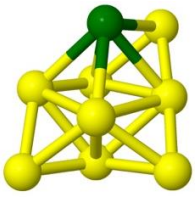
[0.46; C_1; ^{6}A]



C8-iso10-L

[0.29; C_1; ^{4}A]

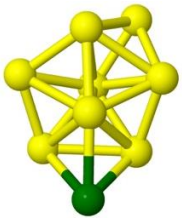
[0.29; C_1; ^{6}A]



C8-iso10-R

[0.29; C_1; ^{4}A]

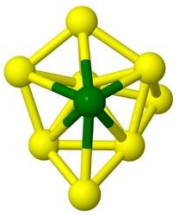
[0.29; C_1; ^{6}A]



C8-iso11

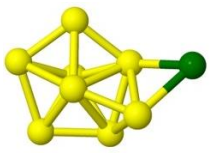
[0.32; C_1; ^{4}A]

[0.55; C_1; ^{6}A]



C8-iso12

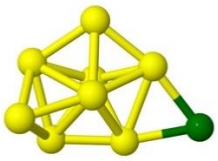
[0.33; C_1; ^{4}A]



C8-iso13

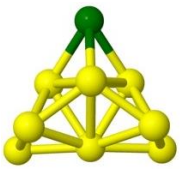
[0.37; C_1; ^{4}A]

[0.60; C_1; ^{6}A]



C8-iso14

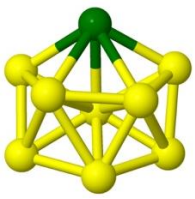
[0.52; C_1; ^{4}A]



C8-iso15

[0.54; C_s; ^{4}A’’]

[0.73; C_s; ^{6}A’’]



C8-iso16

[0.56; C_1; ^{4}A]

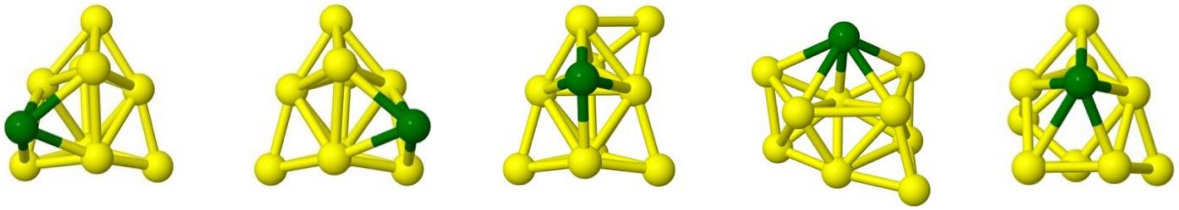
[0.61; C_1; ^{6}A]

C8-iso17

[0.67; C_s; ^{4}A’’]

[0.75; C_s; ^{6}A’’]

Figure S6. Stable isomers of cationic $CrSi_8^+$ cluster.



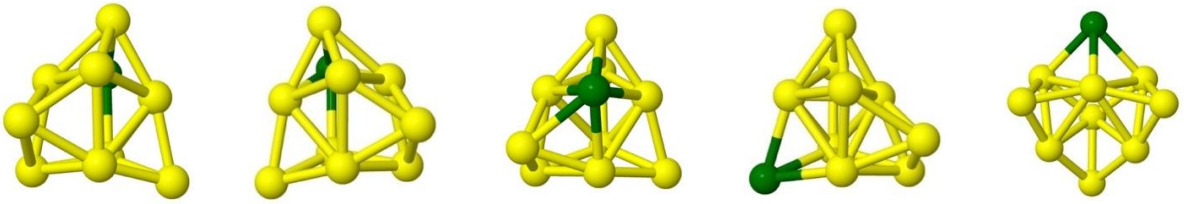
C9-iso1-L
[0.00; C_1; ^6A]
[0.77; C_1; ^4A]

C9-iso1-R
[0.00; C_1; ^6A]
[0.77; C_1; ^4A]

C9-iso2
[0.11; C_1; ^6A]
[0.69; C_1; ^4A]

C9-iso3
[0.12; C_1; ^6A]
[0.91; C_1; ^4A]

C9-iso4
[0.29; C_1; ^6A]



C9-iso5-L
[0.37; C_1; ^6A]
[0.59; C_1; ^4A]

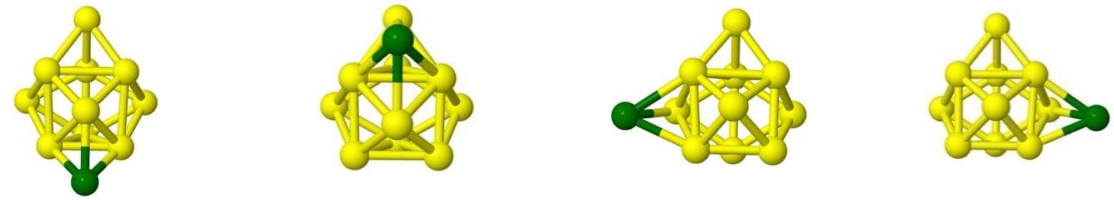
C9-iso5-R
[0.37; C_1; ^6A]
[0.59; C_1; ^4A]

C9-iso6
[0.52; C_1; ^6A]
[0.73; C_1; ^4A]

C9-iso7
[0.55; C_1; ^6A]

C9-iso8
[0.65; C_1; ^6A]
[0.72; C_1; ^4A]

Figure S7. Stable isomers of cationic $CrSi_9^+$ cluster.

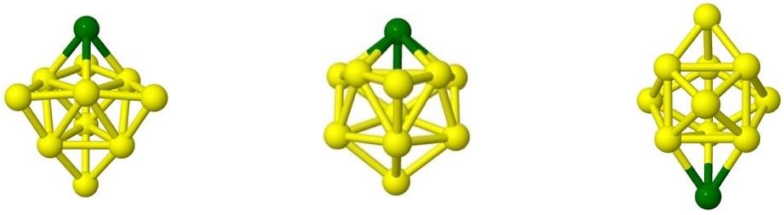


C10-iso1
[0.00; C_1; ^6A]
[1.08; C_{3v}; $^4A_1$]

C10-iso2
[0.04; C_s; $^6A'$]
[1.13; C_s; $^4A'$]

C10-iso3-L
[0.10; C_1; ^6A]
[0.96; C_1; ^4A]

C10-iso3-R
[0.10; C_1; ^6A]
[0.96; C_1; ^4A]

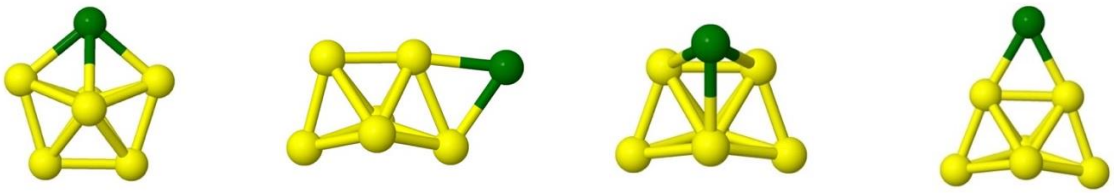


C10-iso4
[0.30; C_s; $^6A'$]
[1.41; C_1; ^4A]

C10-iso5
[0.33; C_s; $^6A'$]
[1.20; C_s; $^4A'$]

C10-iso6
[0.34; C_{3v}; $^6A_1$]

Figure S8. Stable isomers of cationic $CrSi_{10}^+$ cluster.

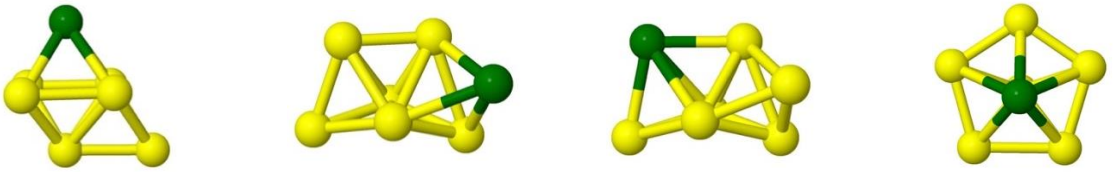


N6-iso1
[0.00; C_{2v}; $^5B_2$]
[0.44; C_{2v}; $^7B_2$]

N6-iso2
[0.36; C_s; $^5A'$]
[0.48; C_s; $^7A'$]

N6-iso3
[0.36; C_s; $^5A''$]
[0.50; C_1; ^7A]

N6-iso4
[0.41; C_{2v}; $^5B_2$]
[0.84; C_1; ^7A]



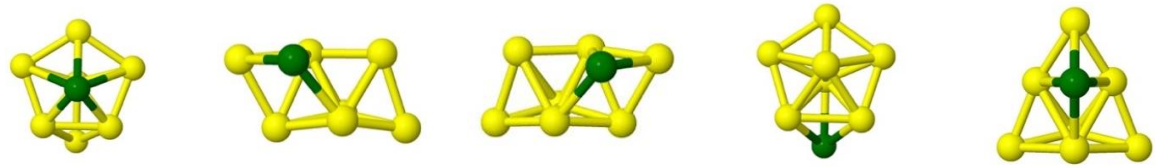
N6-iso5
[0.42; C_s; $^5A'$]
[0.91; C_1; ^3A]

N6-iso6
[0.43; C_1; ^5A]
[1.62; C_1; ^3A]

N6-iso7
[0.64; C_1; ^5A]

N6-iso8
[0.89; C_1; ^3A]
[1.09; C1; ^5A]

Figure S9. Stable isomers of neutral $CrSi_6$ cluster



N7-iso1
[0.00; C_s; $^5A'$]
[0.30; C_s; $^3A'$]

N7-iso2-L
[0.02; C_1; ^5A]

N7-iso2-R
[0.02; C_1; ^5A]

N7-iso3
[0.06; C_s; $^5A'$]
[0.21; C_s; $^7A'$]

N7-iso4
[0.07; C_s; $^5A'$]
[0.45; C_s; $^7A'$]

N7-iso5
[0.10; C_{2v}; $^5B_1$]
[0.13; C_{2v}; $^7B_1$]

N7-iso6
[0.15; C_s; $^5A''$]
[0.58; C_s; $^3A''$]

N7-iso7
[0.22; C_1; ^5A]
[0.83; C_{3v}; $^3A_1$]

N7-iso8
[0.44; C_{2v}; $^5B_1$]
[0.47; C_{2v}; $^7B_1$]

N7-iso9
[0.45; C_s; $^7A'$]
[0.47; C_s; $^5A'$]

Figure S10. Stable isomers of neutral $CrSi_7$ cluster

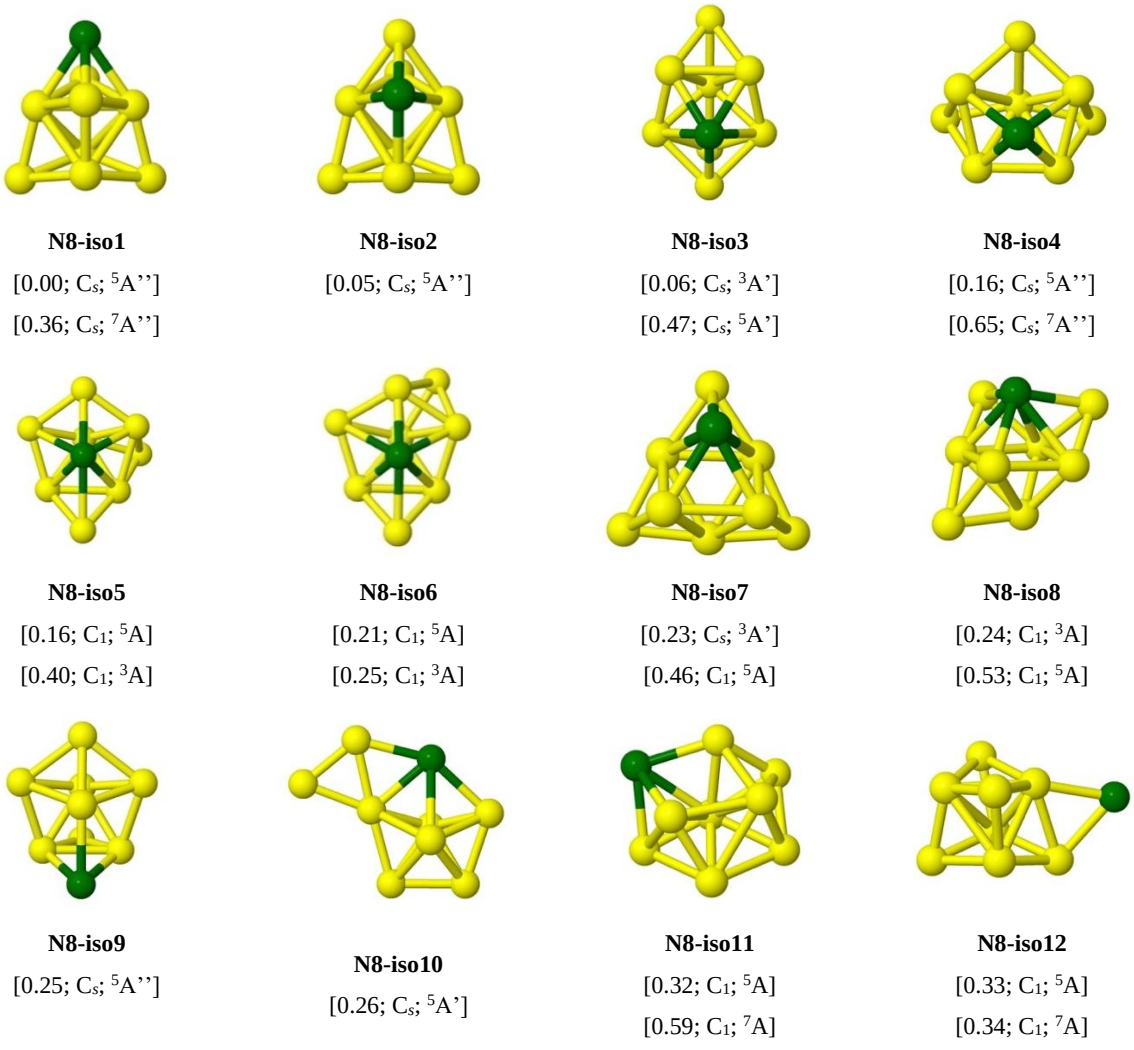
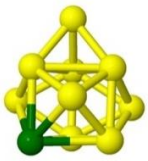


Figure S11. Stable isomers of neutral $CrSi_8$ cluster



N9-iso1

[0.00; C_1; ^{5}A]

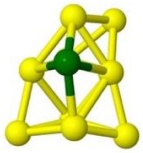
[0.44; C_1; ^{7}A]

N9-iso2-L

[0.07; C_1; ^{5}A]

N9-iso2-R

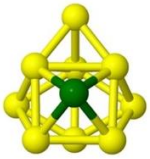
[0.07; C_1; ^{5}A]



N9-iso3

[0.15; C_1; ^{5}A]

[0.39; C_1; ^{7}A]



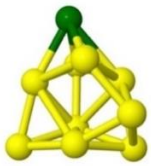
N9-iso4

[0.25; C_s; ^{5}A’’]

[0.51; C_s; ^{7}A’]

N9-iso5-L

[0.26; C_1; ^{5}A]

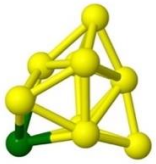


N9-iso5-R

[0.26; C_1; ^{5}A]

N9-iso6-L

[0.38; C_1; ^{7}A]



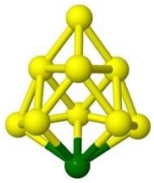
N9-iso6-R

[0.38; C_1; ^{7}A]

N9-iso7

[0.39; C_{3v}; ^{1}A$_1$]

[0.85; C_1; ^{3}A]



N9-iso8

[0.40; C_s; ^{5}A’]

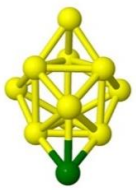
[0.54; C_s; ^{7}A’]

N9-iso9

[0.42; C_s; ^{3}A’’]

[0.71; C_s; ^{5}A’’]

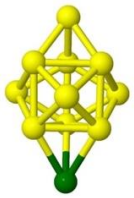
Figure S12. Stable isomers of neutral $CrSi_9$ cluster



N10-iso1

[0.00; C_s; $^5A'$]

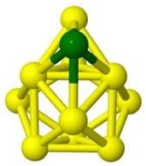
[0.15; C_s; $^7A'$]



N10-iso2

[0.01; C_{3v}; $^7A_1$]

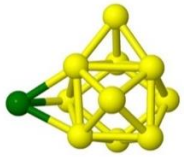
[0.23; C_{3v}; $^5A_1$]



N10-iso3

[0.02; C_s; $^5A'$]

[0.20; C_s; $^7A'$]



N10-iso4

[0.11; C_1; ^5A]

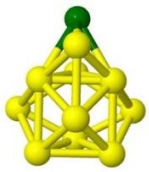
[0.13; C_1; ^7A]

N10-iso5

[0.14; C_{2v}; $^1A_1$]

N10-iso6

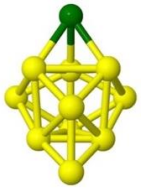
[0.16; C_{2v}; $^1A_1$]



N10-iso7

[0.21; C_s; $^5A'$]

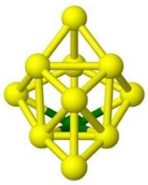
[0.37. C_s; $^7A'$]



N10-iso8

[0.25; C_s; $^5A'$]

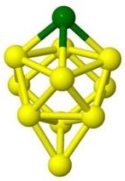
[0.38; C_s; $^7A'$]



N10-iso9

[0.28; C_s; $^5A'$]

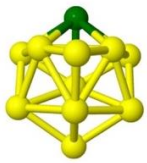
[0.49; C_s; $^7A'$]



N10-iso10

[0.29; C_s; $^5A'$]

[0.48; C_s; $^7A'$]



N10-iso11

[0.31; C_s; $^5A''$]

[0.64; C_s; $^7A''$]

Figure S13. Stable isomers of neutral $CrSi_{10}$ cluster

A3-iso1
[0.00; C_{2v}; $^6A_1$]

A4-iso1
[0.00; C_1; ^4A]

A5-iso1
[0.00; C_s; $^6A'$]

A6-iso1
[0.00; C_{5v}; $^4A_1$]

A6-iso2
[0.32; C_1; ^6A]

A7-iso1
[0.00; C_s; $^6A'$]
[0.01; C_s; $4A'$]

A7-iso2-L
[0.05; C_1; ^6A]

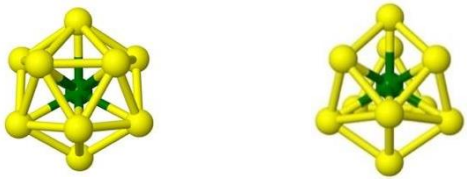
A7-iso2-R
[0.05; C_1; ^6A]

A8-iso1
[0.00; C_{2v}; $^6A_1$]

A8-iso2
[0.18; C_s; $6A'$]

A8-iso3
[0.22; C_s; $6A'$]

A9-iso1
[0.00; C_1; ^6A]



A9-iso2
[0.05; C_{3v}; $^2A_1$]

A10-iso1
[0.00; C_1; ^2A]

Figure S14. Stable structures of anionic $CrSi_n^-$ cluster (n = 6-10).

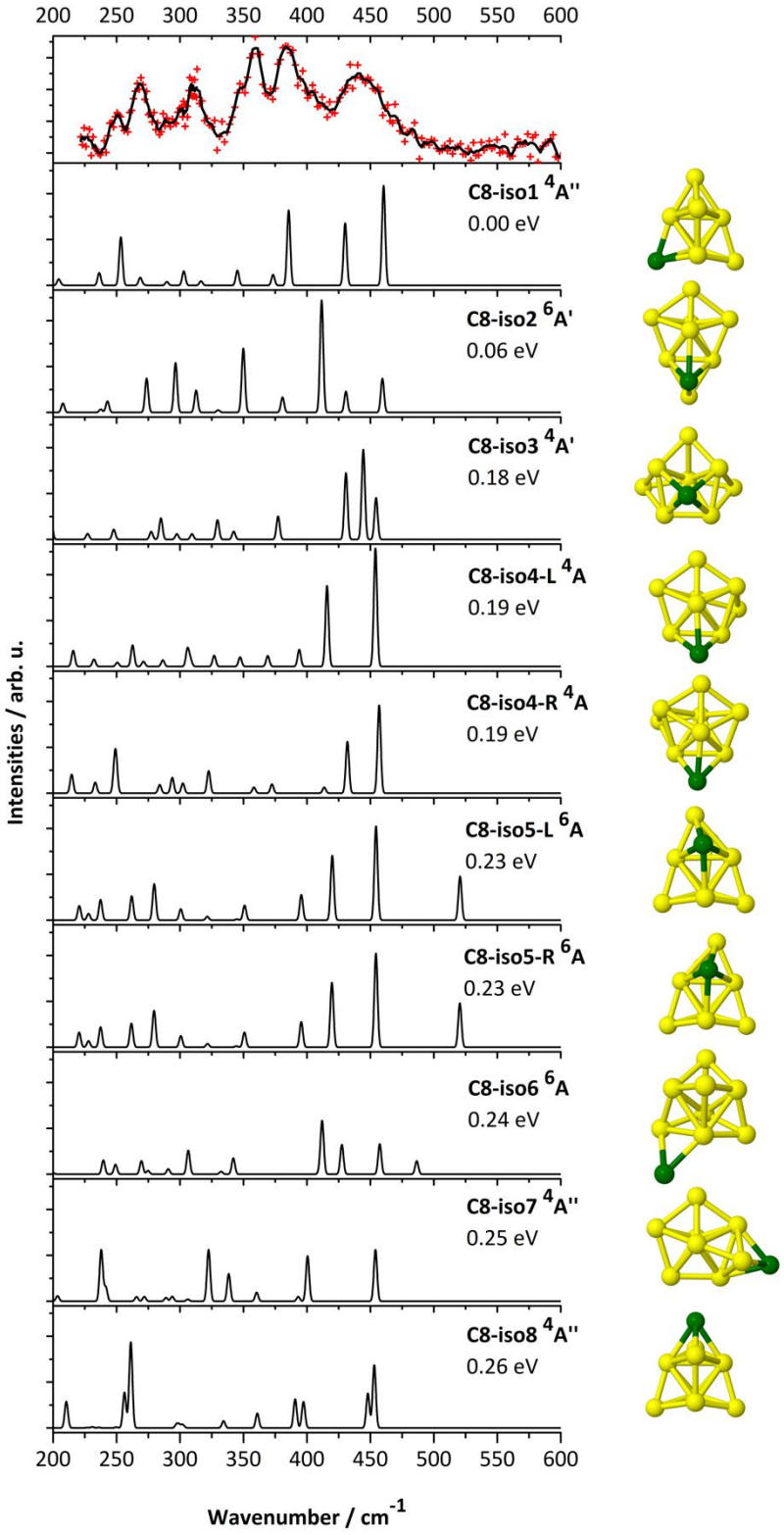


Figure S15. IR spectra comparison of $CrSi_8^+$ clusters between calculation and experiment.

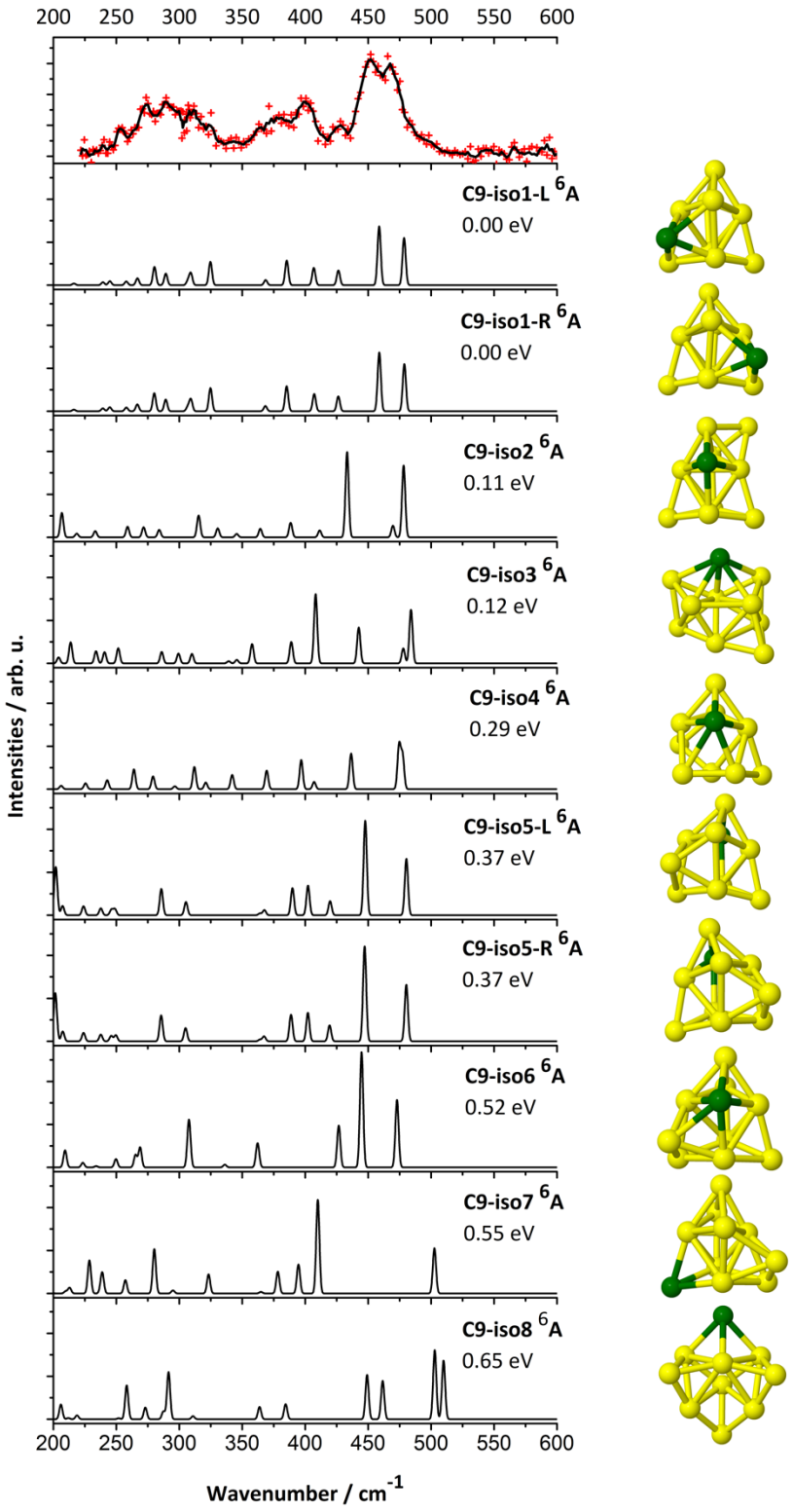


Figure S16. IR spectra comparison of $CrSi_9^+$ clusters between calculation and experiment.

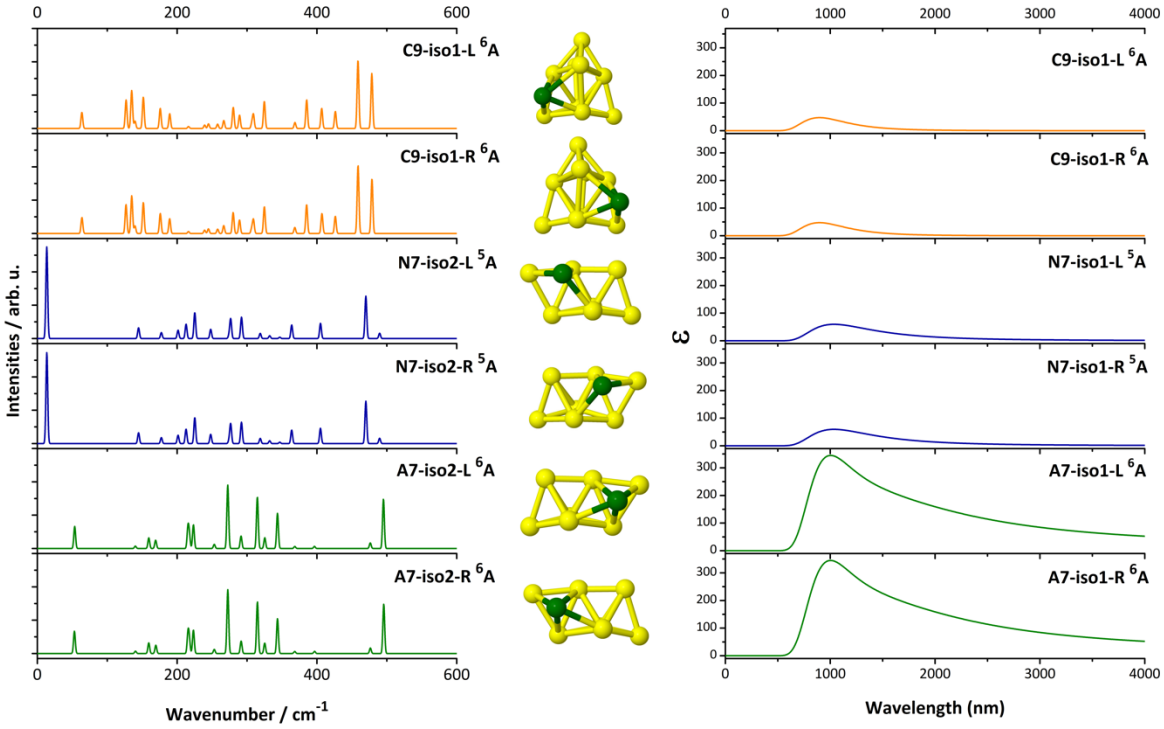


Figure S17. Calculated-IR and UV-Vis spectra of ground-state enantiomeric clusters $CrSi_n^{+/0/-}$ (n = 9 for cation, n = 7 for neutral and anion).

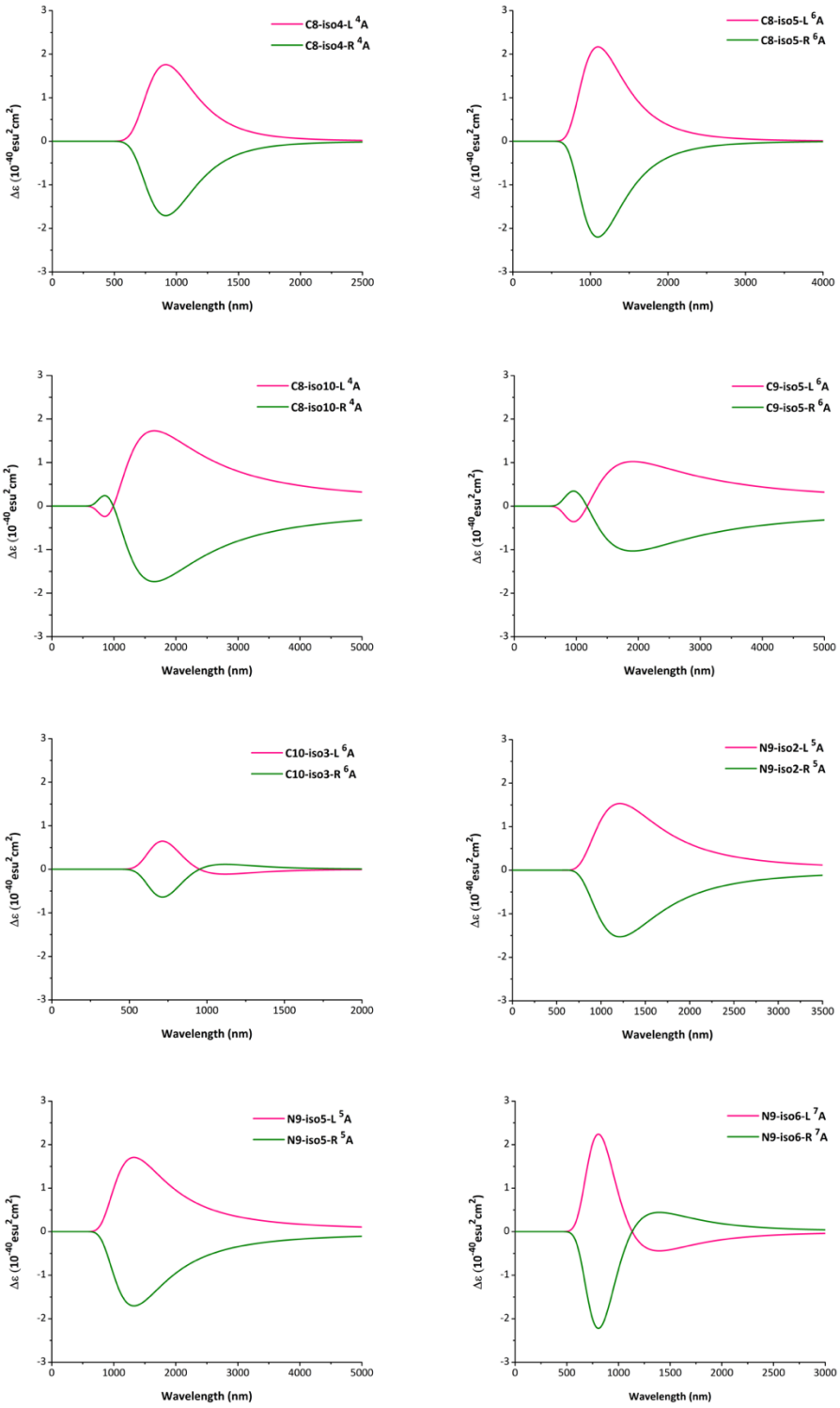


Figure S18. Calculated Electronic Circular Dichroism spectra of higher-energy enantiomeric clusters $CrSi_n^{+/0/-}$ (n = 3-10).