Electronic Supplementary Material (ESI) for Dalton Transactions

Supporting Information

Metallothionein Zn^{2+}- and Cu^{2+}-clusters from first-principles calculations

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PART 1: MT2-alpha

Table S1. Optimized M-S bond lengths of MT2-alpha with restrained C-alpha; highest spin state (Å).

C33-63	C34-63	C34 – 65	C36 – 65	C37 - 64	C37 – 65	C41 – 64	C44 - 63	C44 – 64	C48 – 63	C50 - 62	C50 – 65	C57 – 62	C59 – 62	C60 – 62	C60 - 64	Metal (62-63-64-65)	E_{el}(a.u.)	M_S
2.38	**2.46**	**2.43**	**2.35**	**2.43**	**2.42**	**2.36**	**2.52**	**2.45**	**2.36**	**2.44**	**2.39**	**2.36**	**2.37**	**2.47**	**2.47**	**Zn-Zn-Zn-Zn**	**-12444.528428**	0
2.38	2.46	2.43	2.35	2.42	2.41	2.36	2.51	2.46	2.36	2.37	2.40	2.29	2.35	2.45	2.44	Cu-Zn-Zn-Zn	-12303.503189	½
2.32	2.39	2.42	2.35	2.43	2.42	2.37	2.51	2.44	2.31	2.42	2.38	2.36	2.37	2.45	2.49	Zn-Cu-Zn-Zn	-12303.499973	
2.38	2.44	2.43	2.35	2.39	2.41	2.30	2.48	2.39	2.36	2.44	2.39	2.36	2.38	2.44	2.48	Zn-Zn-Cu-Zn	-12303.499013	
2.37	**2.48**	**2.34**	**2.34**	**2.40**	**2.38**	**2.36**	**2.49**	**2.45**	**2.37**	**2.40**	**2.34**	**2.36**	**2.36**	**2.45**	**2.48**	**Zn-Zn-Zn-Cu**	**-12303.503219**	
2.32	2.39	2.42	2.35	2.43	2.41	2.37	2.50	2.45	2.31	2.37	2.39	2.29	2.35	2.45	2.46	Cu-Cu-Zn-Zn	-12162.474388	1
2.37	2.45	2.44	2.35	2.40	2.40	2.34	2.49	2.38	2.36	2.38	2.40	2.31	2.35	2.40	2.43	Cu-Zn-Cu-Zn	-12162.473808	
2.38	**2.48**	**2.35**	**2.34**	**2.39**	**2.39**	**2.36**	**2.49**	**2.46**	**2.36**	**2.32**	**2.33**	**2.29**	**2.35**	**2.44**	**2.45**	**Cu-Zn-Zn-Cu**	**-12162.477108**	
2.32	2.38	2.43	2.35	2.40	2.41	2.32	2.42	2.39	2.31	2.43	2.38	2.36	2.38	2.43	2.51	Zn-Cu-Cu-Zn	-12162.474158	
2.32	2.38	2.32	2.34	2.40	2.40	2.36	2.46	2.44	2.32	2.39	2.33	2.36	2.37	2.45	2.49	Zn-Cu-Zn-Cu	-12162.474388	
2.37	2.46	2.34	2.35	2.35	2.37	2.29	2.46	2.40	2.37	2.41	2.34	2.36	2.37	2.43	2.46	Zn-Zn-Cu-Cu	-12162.475186	
2.33	2.39	2.44	2.35	2.39	2.40	2.33	2.43	2.39	2.31	2.38	2.39	2.31	2.36	2.40	2.47	Cu-Cu-Cu-Zn	-12021.449494	3/2
2.32	**2.38**	**2.33**	**2.35**	**2.40**	**2.40**	**2.36**	**2.46**	**2.45**	**2.32**	**2.32**	**2.33**	**2.30**	**2.35**	**2.43**	**2.46**	**Cu-Cu-Zn-Cu**	**-12021.449627**	
2.37	2.47	2.36	2.35	2.36	2.36	2.33	2.47	2.37	2.37	2.33	2.33	2.32	2.34	2.38	2.44	Cu-Zn-Cu-Cu	-12021.449260	
2.32	2.38	2.33	2.35	2.37	2.39	2.32	2.43	2.37	2.32	2.41	2.32	2.36	2.38	2.43	2.50	Zn-Cu-Cu-Cu	-12021.448052	
2.32	**2.39**	**2.34**	**2.35**	**2.37**	**2.37**	**2.33**	**2.43**	**2.38**	**2.32**	**2.33**	**2.33**	**2.32**	**2.35**	**2.38**	**2.47**	**Cu-Cu-Cu-Cu**	**-11880.425690**	**2**

Table S2. Optimized M-S bond lengths of MT2-alpha with unrestrained C-alpha; highest spin state (Å).

C33-63	C34-63	C34 – 65	C36 – 65	C37 - 64	C37 – 65	C41 – 64	C44 - 63	C44 – 64	C48 – 63	C50 - 62	C50 – 65	C57 – 62	C59 – 62	C60 – 62	C60 - 64	Metal (62-63-64-65)	E_{el}(a.u.)	M_S
2.36	**2.43**	**2.42**	**2.35**	**2.41**	**2.41**	**2.35**	**2.46**	**2.43**	**2.36**	**2.44**	**2.41**	**2.34**	**2.36**	**2.44**	**2.38**	**Zn-Zn-Zn-Zn**	**-12444.556672**	**0**
2.37	**2.43**	**2.42**	**2.34**	**2.41**	**2.40**	**2.36**	**2.47**	**2.40**	**2.36**	**2.37**	**2.39**	**2.30**	**2.32**	**2.39**	**2.39**	**Cu-Zn-Zn-Zn**	**-12303.527613**	**½**
2.33	2.37	2.40	2.34	2.39	2.41	2.34	2.38	2.39	2.33	2.44	2.39	2.36	2.36	2.44	2.42	Zn-Cu-Zn-Zn	-12303.526246	
2.35	2.44	2.45	2.34	2.36	2.39	2.31	2.44	2.31	2.37	2.45	2.41	2.37	2.36	2.42	2.41	Zn-Zn-Cu-Zn	-12303.526554	
2.36	2.45	2.34	2.31	2.40	2.36	2.35	2.46	2.39	2.37	2.41	2.32	2.35	2.36	2.43	2.43	Zn-Zn-Zn-Cu	-12303.521606	
2.33	**2.38**	**2.41**	**2.35**	**2.41**	**2.41**	**2.37**	**2.42**	**2.41**	**2.32**	**2.36**	**2.39**	**2.30**	**2.32**	**2.40**	**2.39**	**Cu-Cu-Zn-Zn**	**-12162.503970**	**1**
2.37	2.41	2.43	2.34	2.37	2.39	2.32	2.46	2.34	2.36	2.38	2.39	2.31	2.31	2.36	2.38	Cu-Zn-Cu-Zn	-12162.497532	
2.37	2.45	2.35	2.32	2.39	2.37	2.37	2.47	2.40	2.36	2.34	2.32	2.30	2.32	2.39	2.40	Cu-Zn-Zn-Cu	-12162.500294	
2.32	2.36	2.42	2.34	2.36	2.39	2.30	2.37	2.31	2.33	2.43	2.38	2.36	2.36	2.41	2.40	Zn-Cu-Cu-Zn	-12162.498922	
2.32	2.37	2.33	2.32	2.38	2.36	2.35	2.40	2.39	2.33	2.40	2.33	2.35	2.37	2.43	2.43	Zn-Cu-Zn-Cu	-12162.499353	
2.37	2.44	2.35	2.34	2.35	2.36	2.31	2.47	2.33	2.36	2.42	2.33	2.35	2.37	2.41	2.41	Zn-Zn-Cu-Cu	-12162.496790	
2.33	2.37	2.42	2.35	2.38	2.39	2.31	2.36	2.32	2.32	2.38	2.39	2.31	2.32	2.36	2.37	Cu-Cu-Cu-Zn	-12021.474752	3/2
2.32	2.37	2.33	2.33	2.38	2.38	2.37	2.41	2.42	2.32	2.34	2.33	2.31	2.32	2.39	2.40	Cu-Cu-Zn-Cu	-12021.476046	
2.36	**2.43**	**2.37**	**2.34**	**2.35**	**2.34**	**2.30**	**2.42**	**2.32**	**2.37**	**2.37**	**2.36**	**2.31**	**2.32**	**2.36**	**2.38**	**Cu-Zn-Cu-Cu**	**-12021.479843**	
2.36	2.33	2.32	2.35	2.35	2.30	2.30	2.37	2.31	2.33	2.42	2.35	2.36	2.37	2.41	2.40	Zn-Cu-Cu-Cu	-12021.479586	
2.32	**2.37**	**2.34**	**2.34**	**2.35**	**2.36**	**2.31**	**2.36**	**2.31**	**2.32**	**2.36**	**2.35**	**2.32**	**2.32**	**2.35**	**2.37**	**Cu-Cu-Cu-Cu**	**-11880.457344**	**2**

Table S3. Optimized M-S bond lengths of MT2-alpha with restrained C-alpha; lowest spin state (Å).

C33-63	C34-63	C34 – 65	C36 – 65	C37 - 64	C37 – 65	C41 – 64	C44 - 63	C44 – 64	C48 – 63	C50 - 62	C50 – 65	C57 – 62	C59 – 62	C60 – 62	C60 - 64	Metal (62-63-64-65)	E_{el}(a.u.)	M_S
2.32	2.37	2.42	2.35	2.43	2.41	2.37	2.54	2.43	2.32	2.35	2.40	2.29	2.35	2.43	2.47	Cu-Cu-Zn-Zn	-12162.465279	0
2.38	2.46	2.45	2.36	2.43	2.38	2.32	2.45	2.43	2.37	2.35	2.45	2.28	2.35	2.39	2.44	Cu-Zn-Cu-Zn	-12162.469358	
2.38	2.45	2.36	2.32	2.39	2.41	2.37	2.50	2.47	2.37	2.27	2.33	2.30	2.32	2.44	2.47	Cu-Zn-Zn-Cu	-12162.473470	
2.31	2.43	2.43	2.35	2.38	2.41	2.30	2.44	2.43	2.30	2.44	2.39	2.36	2.38	2.42	2.55	Zn-Cu-Cu-Zn	-12162.468976	
2.31	2.38	2.29	2.33	2.40	2.40	2.37	2.47	2.44	2.32	2.39	2.32	2.35	2.37	2.45	2.50	Zn-Cu-Zn-Cu	-12162.468113	
2.37	**2.46**	**2.35**	**2.34**	**2.35**	**2.35**	**2.29**	**2.46**	**2.39**	**2.37**	**2.41**	**2.35**	**2.36**	**2.37**	**2.43**	**2.49**	**Zn-Zn-Cu-Cu**	**-12162.475499**	
2.32	2.38	2.44	2.35	2.40	2.40	2.33	2.45	2.39	2.31	2.38	2.39	2.30	2.36	2.41	2.47	Cu-Cu-Cu-Zn	-12021.447903	½
2.32	**2.39**	**2.43**	**2.35**	**2.43**	**2.41**	**2.36**	**2.50**	**2.45**	**2.31**	**2.37**	**2.39**	**2.29**	**2.35**	**2.44**	**2.46**	**Cu-Cu-Zn-Cu**	**-12021.449651**	
2.38	2.48	2.38	2.33	2.33	2.33	2.32	2.46	2.43	2.37	2.32	2.38	2.29	2.33	2.37	2.43	Cu-Zn-Cu-Cu	-12021.448841	
2.32	2.37	2.34	2.33	2.35	2.36	2.32	2.38	2.37	2.33	2.41	2.34	2.36	2.38	2.41	2.54	Zn-Cu-Cu-Cu	-12021.449477	
2.34	**2.36**	**2.33**	**2.33**	**2.32**	**2.36**	**2.32**	**2.35**	**2.40**	**2.34**	**2.32**	**2.37**	**2.30**	**2.32**	**2.36**	**2.49**	**Cu-Cu-Cu-Cu**	**-11880.424240**	**0**

Table S4. Optimized M-S bond lengths of MT2-alpha with unrestrained C-alpha; lowest spin state (Å).

C33-63	C34-63	C34 – 65	C36 – 65	C37 - 64	C37 – 65	C41 – 64	C44 - 63	C44 – 64	C48 – 63	C50 - 62	C50 – 65	C57 – 62	C59 – 62	C60 – 62	C60 - 64	Metal (62-63-64-65)	E_{el}(a.u.)	M_S
2.32	2.34	2.40	2.34	2.42	2.41	2.37	2.41	2.41	2.33	2.35	2.39	2.31	2.32	2.40	2.41	Cu-Cu-Zn-Zn	-12162.490447	0
2.36	2.44	2.40	2.34	2.55	2.37	2.35	2.45	2.41	2.36	2.36	2.47	2.29	2.33	2.37	2.39	Cu-Zn-Cu-Zn	-12162.501228	
2.36	2.43	2.36	2.28	2.39	2.35	2.36	2.44	2.39	2.37	2.29	2.35	2.30	2.31	2.37	2.40	Cu-Zn-Zn-Cu	-12162.502162	
2.31	2.35	2.44	2.35	2.35	2.39	2.29	2.38	2.35	2.33	2.44	2.38	2.36	2.37	2.40	2.42	Zn-Cu-Cu-Zn	-12162.509202	
2.32	**2.34**	**2.33**	**2.31**	**2.37**	**2.35**	**2.34**	**2.37**	**2.43**	**2.30**	**2.40**	**2.38**	**2.35**	**2.36**	**2.46**	**2.40**	**Zn-Cu-Zn-Cu**	**-12162.510495**	
2.37	2.43	2.36	2.32	2.35	2.34	2.31	2.46	2.33	2.36	2.42	2.33	2.35	2.37	2.40	2.43	Zn-Zn-Cu-Cu	-12162.497912	
2.31	2.36	2.44	2.36	2.38	2.39	2.30	2.37	2.35	2.32	2.38	2.39	2.31	2.33	2.36	2.37	Cu-Cu-Cu-Zn	-12021.482051	½
2.33	**2.35**	**2.33**	**2.29**	**2.38**	**2.38**	**2.37**	**2.39**	**2.41**	**2.32**	**2.30**	**2.32**	**2.31**	**2.31**	**2.36**	**2.41**	**Cu-Cu-Zn-Cu**	**-12021.482491**	
2.36	2.43	2.37	2.33	2.36	2.33	2.30	2.42	2.32	2.37	2.34	2.36	2.31	2.32	2.36	2.38	Cu-Zn-Cu-Cu	-12021.479968	
2.32	2.36	2.32	2.31	2.34	2.34	2.30	2.37	2.32	2.31	2.42	2.34	2.36	2.37	2.40	2.43	Zn-Cu-Cu-Cu	-12021.481537	
2.35	**2.32**	**2.35**	**2.31**	**2.32**	**2.33**	**2.32**	**2.34**	**2.37**	**2.33**	**2.32**	**2.37**	**2.28**	**2.29**	**2.34**	**2.43**	**Cu-Cu-Cu-Cu**	**-11880.455722**	**0**

Table S5. Optimized M-S bond lengths of MT2-alpha with restrained C-alpha; medium spin state (Å).

C33-63	C34-63	C34 – 65	C36 – 65	C37 - 64	C37 – 65	C41 – 64	C44 - 63	C44 – 64	C48 – 63	C50 - 62	C50 – 65	C57 – 62	C59 – 62	C60 – 62	C60 - 64	Metal (62-63-64-65)	E_{el}(a.u.)	M_S
2.32	**2.38**	**2.33**	**2.35**	**2.36**	**2.38**	**2.33**	**2.44**	**2.38**	**2.33**	**2.33**	**2.33**	**2.31**	**2.35**	**2.39**	**2.47**	**Cu-Cu-Cu-Cu**	**-11880.424591**	**1**

Table S6. Optimized M-S bond lengths of MT2-alpha with unrestrained C-alpha; medium spin state (Å).

C33-63	C34-63	C34 – 65	C36 – 65	C37 - 64	C37 – 65	C41 – 64	C44 - 63	C44 – 64	C48 – 63	C50 - 62	C50 – 65	C57 – 62	C59 – 62	C60 – 62	C60 - 64	Metal (62-63-64-65)	E_{el}(a.u.)	M_S
2.33	**2.37**	**2.35**	**2.29**	**2.37**	**2.33**	**2.31**	**2.35**	**2.32**	**2.33**	**2.33**	**2.36**	**2.31**	**2.31**	**2.34**	**2.38**	**Cu-Cu-Cu-Cu**	**-11880.458628**	**1**

Table S7. Optimized angles of MT2-alpha with restrained C-alpha; highest spin state (degrees).

C33-63-C34	C33-63-C44	C33-63-C48	C34-63-C44	C34-63-C48	C44-63-C48	C34-65-C36	C34-65-C37	C34-65-C50	C36-65-C37	C36-65-C50	C37-65-C50	C37-64-C41	C37-64-C44	C37-64-C60	C41-64-C44	C41-64-C60	C44-64-C60	C50-62-C57	C50-62-C59	C50-62-C60	C57-62-C59	C57-62-C60	C59-62-C60	Metal 62-63-64-65
109.1	**110.1**	**109.6**	**109.2**	**105.1**	**113.5**	**110.5**	**100.9**	**113.0**	**120.1**	**108.8**	**103.5**	**115.3**	**113.7**	**97.8**	**109.0**	**105.5**	**114.9**	**122.6**	**102.9**	**98.1**	**115.9**	**105.6**	**110.1**	**Zn-Zn-Zn-Zn**
109.1	110.2	109.6	109.1	104.7	114.0	109.6	100.9	113.4	118.4	109.7	104.7	115.5	112.8	95.8	108.8	107.9	115.8	136.3	103.6	97.0	98.6	104.7	118.6	Cu-Zn-Zn-Zn
121.4	110.9	99.6	105.6	99.9	120.0	110.8	101.3	113.1	119.3	109.8	102.4	115.2	111.1	98.0	109.3	102.8	120.1	121.7	99.6	100.0	116.3	107.5	110.4	Zn-Cu-Zn-Zn
113.5	107.5	108.3	106.8	105.0	116.0	109.5	99.1	115.9	122.2	108.6	101.7	123.9	109.6	93.9	107.2	102.2	120.7	121.7	98.3	101.1	116.1	107.9	110.6	Zn-Zn-Cu-Zn
106.2	**110.6**	**109.8**	**113.1**	**105.8**	**111.1**	**104.2**	**99.2**	**124.7**	**125.6**	**104.7**	**100.7**	**116.6**	**112.1**	**99.4**	**109.2**	**105.2**	**114.0**	**117.3**	**101.6**	**102.7**	**117.0**	**107.5**	**109.7**	**Zn-Zn-Zn-Cu**
120.4	111.3	99.5	105.5	100.1	120.5	109.7	102.7	111.5	117.4	110.4	104.9	115.4	110.1	95.1	108.8	106.5	120.8	136.6	103.0	96.5	98.8	104.8	119.3	Cu-Cu-Zn-Zn
113.3	107.8	109.1	106.0	103.2	117.5	109.2	100.1	114.8	119.5	108.8	104.4	123.8	109.0	91.4	103.9	106.5	123.8	136.0	100.0	98.3	99.7	104.9	120.2	Cu-Zn-Cu-Zn
106.1	**109.9**	**109.9**	**112.6**	**106.3**	**111.7**	**104.9**	**99.4**	**122.5**	**122.4**	**104.9**	**104.3**	**118.1**	**110.8**	**97.1**	**108.4**	**107.9**	**114.4**	**130.2**	**101.7**	**100.2**	**101.4**	**106.2**	**118.7**	**Cu-Zn-Zn-Cu**
121.7	107.8	101.2	105.5	98.3	123.6	109.5	99.2	114.2	121.9	109.9	101.9	125.2	106.9	95.3	101.3	102.8	128.3	121.7	97.5	100.6	116.0	108.8	111.0	Zn-Cu-Cu-Zn
116.8	110.1	99.8	110.5	101.2	118.2	105.4	100.2	122.9	122.5	105.7	101.6	117.9	108.9	99.3	108.9	103.5	118.5	118.6	100.4	102.0	116.9	107.9	109.8	Zn-Cu-Zn-Cu
110.1	107.6	108.4	110.7	105.8	114.3	104.4	96.4	125.9	127.2	105.3	100.2	125.6	104.8	95.1	109.7	103.0	119.0	118.2	99.2	102.9	117.5	107.8	110.1	Zn-Zn-Cu-Cu
121.9	107.7	100.7	104.7	98.4	124.9	109.1	100.1	114.0	119.7	109.6	104.1	124.8	106.3	91.4	101.6	105.7	129.9	135.5	98.2	97.9	99.7	106.4	121.7	Cu-Cu-Cu-Zn
117.2	**110.3**	**100.2**	**109.8**	**100.9**	**118.5**	**105.2**	**100.9**	**121.0**	**120.8**	**106.5**	**103.5**	**118.1**	**108.2**	**97.0**	**108.6**	**106.0**	**119.2**	**130.4**	**99.6**	**100.1**	**102.0**	**106.9**	**119.4**	**Cu-Cu-Zn-Cu**
110.4	107.5	109.7	109.6	104.3	115.3	105.6	96.9	123.3	125.9	104.8	102.2	126.9	105.6	92.6	105.3	106.2	121.7	127.9	98.9	102.4	103.2	105.5	120.7	Cu-Zn-Cu-Cu
119.8	107.0	102.0	107.3	100.2	121.5	106.1	97.3	124.4	124.7	105.3	100.9	128.0	103.3	95.9	102.6	103.2	127.0	118.6	98.2	102.6	116.9	108.7	110.7	Zn-Cu-Cu-Cu
119.9	**106.9**	**101.6**	**106.8**	**99.3**	**123.4**	**106.4**	**98.0**	**121.7**	**124.1**	**105.8**	**102.2**	**128.0**	**103.3**	**92.5**	**102.8**	**105.3**	**128.0**	**128.7**	**97.6**	**101.8**	**103.0**	**106.5**	**120.9**	**Cu-Cu-Cu-Cu**

Table S8. Optimized angles of MT2-alpha with unrestrained C-alpha; highest spin state (degrees).

C33-63-C34	C33-63-C44	C33-63-C48	C34-63-C44	C34-63-C48	C44-63-C48	C34-65-C36	C34-65-C37	C34-65-C50	C36-65-C37	C36-65-C50	C37-65-C50	C37-64-C41	C37-64-C44	C37-64-C60	C41-64-C44	C41-64-C60	C44-64-C60	C50-62-C57	C50-62-C59	C50-62-C60	C57-62-C59	C57-62-C60	C59-62-C60	Metal 62-63-64-65
112.8	**103.8**	**109.1**	**110.5**	**106.9**	**113.7**	**110.3**	**102.5**	**108.4**	**113.6**	**105.6**	**116.3**	**106.4**	**107.0**	**119.3**	**111.4**	**113.2**	**99.3**	**114.9**	**113.4**	**97.1**	**109.3**	**112.8**	**108.7**	**Zn-Zn-Zn-Zn**
113.7	**102.0**	**109.3**	**112.2**	**104.4**	**115.6**	**109.0**	**102.6**	**110.1**	**116.6**	**109.6**	**108.7**	**106.8**	**111.3**	**108.2**	**110.6**	**111.9**	**108.1**	**139.9**	**97.8**	**90.0**	**101.2**	**101.2**	**133.9**	**Cu-Zn-Zn-Zn**
138.6	97.9	96.9	94.8	98.0	140.2	109.1	101.5	113.7	112.1	114.2	105.4	111.5	117.7	99.1	111.4	112.3	103.9	112.3	101.6	109.1	114.6	111.0	107.7	Zn-Cu-Zn-Zn
112.6	122.1	110.0	94.7	111.5	105.0	104.0	111.6	108.5	115.0	110.6	107.0	135.4	99.0	95.2	105.6	92.1	136.2	108.4	104.0	109.1	114.6	113.8	106.4	Zn-Zn-Cu-Zn
115.0	102.2	108.8	112.7	102.5	116.1	103.2	99.2	132.3	134.5	100.0	93.1	113.8	111.9	101.0	112.7	103.0	113.5	116.8	103.4	99.5	114.0	111.6	110.4	Zn-Zn-Zn-Cu
131.3	**97.9**	**96.4**	**103.9**	**98.8**	**133.8**	**111.0**	**103.4**	**107.7**	**113.8**	**109.0**	**111.7**	**108.5**	**106.5**	**109.2**	**110.2**	**109.5**	**112.8**	**140.4**	**97.4**	**88.9**	**101.3**	**101.6**	**134.8**	**Cu-Cu-Zn-Zn**
115.8	102.0	108.8	109.4	104.5	116.9	106.9	102.7	113.7	120.9	108.6	104.3	125.4	103.2	95.5	113.1	92.9	126.9	140.7	96.7	91.1	101.9	100.7	132.9	Cu-Zn-Cu-Zn
113.4	102.3	109.1	115.2	103.4	113.6	102.5	101.5	122.2	127.4	102.3	103.1	108.9	108.5	109.0	111.1	110.3	109.0	134.6	98.5	90.8	102.4	103.0	133.2	Cu-Zn-Zn-Cu
138.1	97.0	97.9	100.0	99.0	131.8	103.5	103.1	115.9	119.0	112.9	102.6	129.7	101.9	98.0	106.9	94.4	129.5	119.4	101.0	103.1	112.3	110.7	109.5	Zn-Cu-Cu-Zn
132.3	97.4	96.3	105.0	98.4	132.7	104.1	98.1	129.8	129.1	103.3	96.1	115.9	109.5	100.5	110.5	104.5	115.7	115.9	103.2	102.6	113.9	112.0	108.3	Zn-Cu-Zn-Cu
114.6	100.8	109.2	112.2	104.3	116.1	102.0	95.0	139.5	137.1	100.8	90.5	130.1	95.4	96.1	112.5	91.5	135.8	116.8	102.1	100.9	113.9	112.6	109.4	Zn-Zn-Cu-Cu
133.7	98.0	98.4	102.3	98.6	131.1	108.5	101.5	113.3	119.3	107.8	106.6	126.7	101.1	98.2	108.7	95.4	129.8	140.2	96.6	91.2	101.6	101.4	133.0	Cu-Cu-Cu-Zn
126.7	97.7	96.6	110.2	99.0	129.2	103.5	101.8	119.2	123.0	103.8	106.7	110.4	104.5	110.9	109.7	108.5	112.8	134.7	98.0	90.1	102.4	102.9	134.7	Cu-Cu-Zn-Cu
116.4	**105.9**	**109.3**	**104.5**	**107.4**	**113.5**	**100.3**	**98.5**	**135.8**	**135.5**	**100.9**	**92.8**	**131.9**	**98.1**	**92.3**	**111.2**	**95.7**	**131.1**	**131.6**	**98.9**	**98.1**	**100.1**	**102.7**	**130.1**	**Cu-Zn-Cu-Cu**
133.4	96.8	100.1	100.9	98.1	133.5	102.3	96.5	135.1	134.6	102.7	91.7	131.4	96.0	96.3	108.9	95.4	133.8	116.0	102.2	104.5	113.0	112.4	107.8	Zn-Cu-Cu-Cu
131.8	**96.8**	**101.0**	**101.1**	**98.7**	**132.8**	**102.5**	**97.6**	**132.6**	**131.8**	**101.9**	**95.5**	**135.4**	**97.2**	**95.3**	**104.4**	**96.6**	**134.5**	**133.6**	**99.1**	**94.6**	**100.6**	**102.4**	**132.1**	**Cu-Cu-Cu-Cu**

Table S9. Optimized angles of MT2-alpha with restrained C-alpha; lowest spin state (degrees).

C33-63-C34	C33-63-C44	C33-63-C48	C34-63-C44	C34-63-C48	C44-63-C48	C34-65-C36	C34-65-C37	C34-65-C50	C36-65-C37	C36-65-C50	C37-65-C50	C37-64-C41	C37-64-C44	C37-64-C60	C41-64-C44	C41-64-C60	C44-64-C60	C50-62-C57	C50-62-C59	C50-62-C60	C57-62-C59	C57-62-C60	C59-62-C60	Metal 62-63-64-65
122.9	110.6	101.8	104.3	100.1	117.5	110.3	102.7	112.0	117.6	109.6	104.5	115.2	110.2	94.3	109.5	105.9	121.4	136.8	102.5	96.8	98.1	105.1	120.1	Cu-Cu-Zn-Zn
111.1	110.3	108.8	108.6	102.7	115.0	108.4	102.1	113.5	119.6	107.3	106.2	119.0	111.5	89.6	104.7	111.8	120.7	141.8	103.7	94.0	93.5	103.1	125.9	Cu-Zn-Cu-Zn
108.4	110.6	109.1	111.4	106.2	111.1	109.0	101.3	127.8	121.4	97.2	102.1	118.1	111.5	96.1	107.6	108.5	114.9	126.4	104.8	102.0	100.2	105.4	119.6	Cu-Zn-Zn-Cu
118.8	101.2	101.0	100.1	98.4	139.4	110.0	98.8	114.7	121.9	109.4	101.9	128.1	103.1	94.5	94.9	103.4	138.3	121.9	97.5	100.2	115.5	108.9	111.6	Zn-Cu-Cu-Zn
120.1	113.0	107.6	107.9	93.0	113.9	108.2	101.5	120.6	121.9	105.2	100.5	117.9	108.9	99.1	109.1	103.0	118.9	118.3	99.8	102.5	117.3	108.1	109.7	Zn-Cu-Zn-Cu
110.1	**107.6**	**108.6**	**111.1**	**105.8**	**113.6**	**104.1**	**97.3**	**125.4**	**127.8**	**104.4**	**100.6**	**124.5**	**106.3**	**95.2**	**109.7**	**102.6**	**118.7**	**117.8**	**99.3**	**103.1**	**117.2**	**108.2**	**110.2**	**Zn-Zn-Cu-Cu**
123.0	108.1	100.6	104.5	98.2	123.9	109.0	100.1	113.9	119.7	109.8	104.2	124.8	106.4	91.3	101.7	105.6	129.8	135.8	98.3	97.8	99.4	106.3	121.9	Cu-Cu-Cu-Zn
121.0	**111.0**	**99.6**	**105.4**	**100.0**	**120.5**	**109.9**	**102.2**	**112.3**	**117.6**	**110.1**	**104.6**	**115.5**	**110.0**	**95.2**	**108.8**	**106.0**	**121.0**	**135.9**	**101.5**	**97.2**	**99.2**	**105.7**	**119.6**	**Cu-Cu-Zn-Cu**
108.0	109.5	109.1	111.4	103.8	114.8	108.1	97.8	116.0	124.6	106.0	104.9	116.7	110.7	93.9	109.3	108.6	117.1	137.4	101.8	97.1	95.4	104.4	125.0	Cu-Zn-Cu-Cu
116.4	107.1	101.6	110.5	100.2	121.4	104.6	97.8	122.4	128.0	105.2	100.8	127.1	105.1	96.6	102.7	103.1	124.7	117.8	98.6	102.8	116.5	109.0	111.0	Zn-Cu-Cu-Cu
105.1	**108.9**	**116.9**	**117.0**	**98.3**	**110.4**	**107.8**	**98.0**	**117.5**	**124.8**	**106.8**	**102.6**	**123.8**	**108.7**	**94.1**	**103.9**	**108.4**	**119.0**	**135.7**	**99.8**	**97.5**	**96.9**	**105.9**	**124.9**	**Cu-Cu-Cu-Cu**

Table S10. Optimized angles of MT2-alpha with unrestrained C-alpha; lowest spin state (degrees).

C33-63-C34	C33-63-C44	C33-63-C48	C34-63-C44	C34-63-C48	C44-63-C48	C34-65-C36	C34-65-C37	C34-65-C50	C36-65-C37	C36-65-C50	C37-65-C50	C37-64-C41	C37-64-C44	C37-64-C60	C41-64-C44	C41-64-C60	C44-64-C60	C50-62-C57	C50-62-C59	C50-62-C60	C57-62-C59	C57-62-C60	C59-62-C60	Metal 62-63-64-65
132.8	98.6	98.5	101.1	98.9	132.4	111.3	102.8	105.0	116.2	109.8	111.0	115.1	106.5	106.3	110.9	101.3	116.8	139.7	96.8	89.4	101.4	102.0	135.0	Cu-Cu-Zn-Zn
113.3	105.3	108.9	109.1	103.8	116.8	112.0	105.2	110.2	116.1	103.6	109.8	108.8	104.6	92.6	109.0	89.0	148.9	165.3	93.4	74.3	101.2	91.1	163.5	Cu-Zn-Cu-Zn
116.4	103.7	108.6	109.8	103.4	115.4	102.2	100.9	136.7	137.4	93.0	94.2	114.0	113.8	103.7	113.3	103.2	107.6	137.5	97.4	95.1	103.6	100.9	126.8	Cu-Zn-Zn-Cu
147.6	90.5	96.1	87.5	94.8	163.7	106.3	101.8	113.5	114.8	113.7	106.3	131.7	106.6	98.8	95.4	96.0	133.2	117.8	100.4	103.2	112.8	111.8	110.0	Zn-Cu-Cu-Zn
151.3	**97.1**	**97.5**	**89.2**	**93.2**	**144.5**	**103.1**	**96.6**	**124.3**	**132.7**	**104.0**	**99.3**	**114.2**	**107.0**	**112.0**	**113.8**	**105.8**	**103.6**	**117.8**	**111.1**	**95.9**	**111.1**	**111.1**	**108.6**	**Zn-Cu-Zn-Cu**
116.1	102.0	108.2	111.5	103.5	116.0	101.5	96.2	139.2	137.9	99.7	90.8	127.9	97.5	96.9	112.2	91.1	135.0	116.6	101.4	99.4	114.1	114.1	109.6	Zn-Zn-Cu-Cu
142.1	89.6	97.0	92.3	95.5	157.4	109.2	101.7	109.8	116.9	108.1	110.9	130.2	101.7	100.0	95.6	96.2	139.1	140.5	97.2	88.7	100.9	101.3	136.4	Cu-Cu-Cu-Zn
134.5	**98.7**	**97.8**	**100.2**	**97.4**	**134.9**	**102.8**	**102.6**	**132.1**	**130.9**	**92.3**	**100.8**	**117.1**	**109.1**	**107.3**	**110.7**	**99.2**	**113.1**	**135.0**	**97.2**	**93.9**	**103.7**	**101.6**	**130.3**	**Cu-Cu-Zn-Cu**
116.7	105.4	108.9	105.1	106.6	114.3	100.6	98.8	139.1	136.9	99.3	90.3	130.7	97.8	93.9	110.9	95.4	131.8	129.6	99.4	99.6	99.3	104.2	128.6	Cu-Zn-Cu-Cu
137.7	97.3	100.2	98.3	96.2	134.6	101.8	97.5	134.5	133.9	101.8	93.3	131.4	97.1	96.4	107.8	94.4	134.8	116.3	101.4	102.4	113.2	114.4	107.7	Zn-Cu-Cu-Cu
112.4	**101.7**	**114.8**	**119.9**	**100.6**	**108.0**	**103.3**	**103.0**	**111.8**	**131.6**	**106.0**	**100.9**	**127.5**	**100.7**	**99.0**	**106.8**	**98.0**	**128.0**	**149.8**	**98.2**	**85.6**	**94.7**	**99.4**	**144.7**	**Cu-Cu-Cu-Cu**

Table S11. Optimized angles of MT2-alpha with restrained C-alpha; medium spin state (degrees).

C33-63-C34	C33-63-C44	C33-63-C48	C34-63-C44	C34-63-C48	C44-63-C48	C34-65-C36	C34-65-C37	C34-65-C50	C36-65-C37	C36-65-C50	C37-65-C50	C37-64-C41	C37-64-C44	C37-64-C60	C41-64-C44	C41-64-C60	C44-64-C60	C50-62-C57	C50-62-C59	C50-62-C60	C57-62-C59	C57-62-C60	C59-62-C60	Metal 62-63-64-65
119.5	**106.9**	**102.6**	**106.9**	**99.6**	**122.4**	**106.6**	**98.0**	**123.3**	**123.5**	**105.3**	**101.7**	**128.0**	**103.5**	**92.8**	**102.6**	**105.2**	**127.8**	**129.2**	**97.5**	**101.9**	**101.6**	**107.2**	**121.1**	**Cu-Cu-Cu-Cu**

Table S12. Optimized angles of MT2-alpha with unrestrained C-alpha; medium spin state (degrees).

C33-63-C34	C33-63-C44	C33-63-C48	C34-63-C44	C34-63-C48	C44-63-C48	C34-65-C36	C34-65-C37	C34-65-C50	C36-65-C37	C36-65-C50	C37-65-C50	C37-64-C41	C37-64-C44	C37-64-C60	C41-64-C44	C41-64-C60	C44-64-C60	C50-62-C57	C50-62-C59	C50-62-C60	C57-62-C59	C57-62-C60	C59-62-C60	Metal 62-63-64-65
136.8	**96.8**	**99.1**	**97.7**	**98.3**	**135.8**	**100.9**	**97.6**	**145.1**	**144.8**	**92.0**	**89.7**	**127.4**	**102.1**	**99.4**	**106.2**	**96.8**	**128.0**	**137.5**	**96.9**	**92.5**	**103.4**	**101.0**	**131.4**	**Cu-Cu-Cu-Cu**

Table S13. Mulliken Charges for High-Spin Models (a.u.).

	62		63		64		65	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Zn-Zn-Zn-Zn	0.4543	0.4316	0.4772	0.4518	0.4529	0.4660	0.5007	0.5102
Cu-Zn-Zn-Zn	0.1010	0.0814	0.4769	0.5065	0.4450	0.4572	0.5046	0.4810
Zn-Cu-Zn-Zn	0.4580	0.4480	0.1479	0.1279	0.4506	0.4400	0.4979	0.5078
Zn-Zn-Cu-Zn	0.4581	0.4686	0.4574	0.4516	0.1521	0.0952	0.4951	0.4698
Zn-Zn-Zn-Cu	0.4605	0.4550	0.4708	0.5237	0.4492	0.4607	0.1734	0.1199
Cu-Cu-Zn-Zn	0.1009	0.0807	0.1477	0.1404	0.4413	0.4290	0.5119	0.4972
Cu-Zn-Cu-Zn	0.0999	0.0854	0.4598	0.4988	0.1424	0.0938	0.5093	0.4891
Cu-Zn-Zn-Cu	0.1031	0.0852	0.4766	0.5212	0.4400	0.4583	0.1834	0.1378
Zn-Cu-Cu-Zn	0.4589	0.4614	0.1274	0.1089	0.1505	0.0979	0.4975	0.5154
Zn-Cu-Zn-Cu	0.4585	0.4493	0.1411	0.1549	0.4461	0.4414	0.1886	0.1491
Zn-Zn-Cu-Cu	0.4591	0.4594	0.4584	0.5158	0.1356	0.0684	0.1694	0.1175
Cu-Cu-Cu-Zn	0.0993	0.0814	0.1281	0.1211	0.1451	0.0963	0.5085	0.5052
Cu-Cu-Zn-Cu	0.1019	0.0810	0.1413	0.1409	0.4387	0.4268	0.1996	0.1662
Cu-Zn-Cu-Cu	0.1019	0.0821	0.4655	0.4733	0.1303	0.0858	0.1821	0.1404
Zn-Cu-Cu-Cu	0.4601	0.4658	0.1277	0.0964	0.1334	0.0824	0.1898	0.1367
Cu-Cu-Cu-Cu	0.1042	0.0755	0.1287	0.0918	0.1337	0.0792	0.1932	0.1674

Table S14. Mulliken Charges for Low-Spin Models (a.u.).

	62		63		64		65	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Cu-Cu-Zn-Zn	0.0888	0.0740	0.1426	0.1424	0.4408	0.4262	0.5096	0.4900
Cu-Zn-Cu-Zn	0.0540	0.0394	0.4610	0.5222	0.1344	0.0588	0.5114	0.4956
Cu-Zn-Zn-Cu	0.0816	0.0980	0.4660	0.5398	0.4438	0.4742	0.1742	0.0950
Zn-Cu-Cu-Zn	0.4610	0.4780	0.1182	0.0764	0.1434	0.0782	0.4960	0.5036
Zn-Cu-Zn-Cu	0.4578	0.4306	0.1304	0.0684	0.4462	0.4406	0.1726	0.1002
Zn-Zn-Cu-Cu	0.4607	0.4603	0.4576	0.5085	0.1376	0.0737	0.1652	0.1086
Cu-Cu-Cu-Zn	0.0982	0.0741	0.1280	0.1084	0.1445	0.0689	0.5083	0.4999
Cu-Cu-Zn-Cu	0.1017	0.0843	0.1402	0.1392	0.4387	0.4275	0.2002	0.1125
Cu-Zn-Cu-Cu	0.0530	0.0813	0.4704	0.4742	0.1272	0.0828	0.1866	0.1361
Zn-Cu-Cu-Cu	0.4603	0.4631	0.1212	0.0903	0.1352	0.0843	0.1810	0.1255
Cu-Cu-Cu-Cu	0.0530	0.0228	0.1136	0.1136	0.1232	0.0630	0.1874	0.1380

Table S15. Mulliken Charges for Medium-Spin Models (a.u.).

	62		63		64		65	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Cu-Cu-Cu-Cu	0.0983	0.0813	0.1289	0.1286	0.1328	0.1047	0.1932	0.0922

Table S16. Molecular areas and volumes of MT models.

	Molecular area ($Bohr^2$)						Molecular volume ($Bohr^3$)					
	RHS	UHS	RLS	ULS	RMS	UMS	RHS	UHS	RLS	ULS	RMS	UMS
Zn-Zn-Zn-Zn	2225	2179					6552	6593				
Cu-Zn-Zn-Zn	2222	2203					6527	6564				
Zn-Cu-Zn-Zn	2232	2161					6543	6566				
Zn-Zn-Cu-Zn	2237	2219					6577	6537				
Zn-Zn-Zn-Cu	2246	2157					6555	6597				
Cu-Cu-Zn-Zn	2236	2218	2235	2190			6541	6580	6535	6526		
Cu-Zn-Cu-Zn	2227	2191	2231	2196			6518	6486	6518	6530		
Cu-Zn-Zn-Cu	2227	2178	2242	2134			6521	6555	6549	6478		
Zn-Cu-Cu-Zn	2235	2203	2251	2203			6527	6546	6556	6546		
Zn-Cu-Zn-Cu	2239	2120	2226	2205			6546	6503	6523	6574		
Zn-Zn-Cu-Cu	2261	2189	2260	2198			6582	6520	6582	6513		
Cu-Cu-Cu-Zn	2239	2190	2235	2198			6521	6542	6507	6509		
Cu-Cu-Zn-Cu	2227	2173	2225	2209			6515	6538	6510	6467		
Cu-Zn-Cu-Cu	2231	2198	2234	2214			6497	6498	6516	6518		
Zn-Cu-Cu-Cu	2240	2164	2241	2164			6521	6473	6527	6459		
Cu-Cu-Cu-Cu	2246	2186	2234	2201	2246	2146	6508	6471	6506	6513	6508	6435

RHS: Restrained High-Spin. UHS: Unrestrained High-Spin. RLS: Restrained Low-Spin. ULS: Unrestrained Low-Spin.
RMS: Restrained Medium-Spin. UMS: Unrestrained Medium-Spin.

PART 2: MT2-beta

Table S17. Optimized M-S bond lengths of MT2-beta with restrained C-alpha; highest spin state (Å).

Cys5-66	Cys7-66	Cys7-68	Cys13-68	Cys15-67	Cys15-68	Cys19-67	Cys21-66	Cys24-66	Cys24-67	Cys26-68	Cys29-67	Metal 66-67-68	Energy	M_S
2.37	**2.47**	**2.51**	**2.38**	**2.45**	**2.42**	**2.32**	**2.42**	**2.42**	**2.41**	**2.34**	**2.37**	**Zn-Zn-Zn**	**-9692.245065**	**0**
2.32	**2.40**	**2.47**	**2.38**	**2.46**	**2.43**	**2.33**	**2.33**	**2.37**	**2.39**	**2.34**	**2.37**	**Cu-Zn-Zn**	**-9551.219565**	½
2.37	2.46	2.48	2.39	2.41	2.39	2.30	2.41	2.46	2.35	2.35	2.38	Zn-Cu-Zn	-9551.214300	
2.38	2.49	2.43	2.35	2.43	2.40	2.33	2.42	2.41	2.42	2.31	2.37	Zn-Zn-Cu	-9551.214472	
2.31	**2.41**	**2.48**	**2.38**	**2.43**	**2.40**	**2.31**	**2.34**	**2.35**	**2.33**	**2.35**	**2.37**	**Cu-Cu-Zn**	**-9410.194011**	**1**
2.33	2.42	2.40	2.35	2.43	2.41	2.33	2.34	2.35	2.41	2.31	2.37	Cu-Zn-Cu	-9410.190716	
2.39	2.47	2.44	2.35	2.40	2.40	2.30	2.40	2.42	2.36	2.32	2.37	Zn-Cu-Cu	-9410.186926	
2.32	**2.42**	**2.41**	**2.35**	**2.40**	**2.37**	**2.30**	**2.35**	**2.35**	**2.35**	**2.32**	**2.37**	**Cu-Cu-Cu**	**-9269.165177**	**3/2**

Table S18. Optimized M-S bond lengths of MT2-beta with unrestrained C-alpha; highest spin state (Å).

Cys5-66	Cys7-66	Cys7-68	Cys13-68	Cys15-67	Cys15-68	Cys19-67	Cys21-66	Cys24-66	Cys24-67	Cys26-68	Cys29-67	Metal 66-67-68	Energy	M_S
2.38	**2.44**	**2.44**	**2.36**	**2.44**	**2.43**	**2.33**	**2.37**	**2.42**	**2.42**	**2.36**	**2.36**	**Zn-Zn-Zn**	**-9692.264201**	**0**
2.33	**2.40**	**2.42**	**2.38**	**2.43**	**2.44**	**2.34**	**2.32**	**2.35**	**2.41**	**2.34**	**2.37**	**Cu-Zn-Zn**	**-9551.238469**	½
2.38	2.42	2.44	2.37	2.38	2.41	2.32	2.37	2.42	2.36	2.36	2.34	Zn-Cu-Zn	-9551.235877	
2.37	2.45	2.36	2.34	2.42	2.42	2.33	2.37	2.39	2.41	2.30	2.37	Zn-Zn-Cu	-9551.236815	
2.32	2.37	2.42	2.37	2.39	2.42	2.30	2.33	2.37	2.36	2.35	2.36	Cu-Cu-Zn	-9410.214985	1
2.33	2.38	2.35	2.33	2.42	2.42	2.34	2.31	2.36	2.41	2.30	2.37	Cu-Zn-Cu	-9410.215537	
2.35	**2.41**	**2.35**	**2.33**	**2.36**	**2.40**	**2.31**	**2.37**	**2.43**	**2.38**	**2.32**	**2.36**	**Zn-Cu-Cu**	**-9410.216689**	
2.33	**2.38**	**2.36**	**2.33**	**2.36**	**2.39**	**2.33**	**2.32**	**2.35**	**2.35**	**2.31**	**2.34**	**Cu-Cu-Cu**	**-9269.190707**	**3/2**

Table S19. Optimized M-S bond lengths of MT2-beta with restrained C-alpha; lowest spin state (Å).

Cys5-66	Cys7-66	Cys7-68	Cys13-68	Cys15-67	Cys15-68	Cys19-67	Cys21 66	Cys24 66	Cys24 67	Cys26-68	Cys29-67	Metal 66-67-68	Energy	M_S
2.31	2.41	2.47	2.38	2.43	2.40	2.31	2.34	2.36	2.33	2.34	2.37	Cu-Cu-Zn	-9410.192553	0
2.31	2.40	2.33	2.33	2.42	2.44	2.34	2.33	2.37	2.41	2.31	2.38	Cu-Zn-Cu	-9410.190177	
2.38	**2.43**	**2.36**	**2.33**	**2.38**	**2.40**	**2.30**	**2.37**	**2.41**	**2.37**	**2.26**	**2.35**	**Zn-Cu-Cu**	**-9410.206034**	
2.31	2.42	2.37	2.34	2.39	2.38	2.32	2.34	2.35	2.34	2.32	2.37	Cu-Cu-Cu	-9269.166253	½

Table S20. Optimized M-S bond lengths of MT2-beta with unrestrained C-alpha; lowest spin state (Å).

Cys5-66	Cys7-66	Cys7-68	Cys13-68	Cys15-67	Cys15-68	Cys19-67	Cys21 66	Cys24 66	Cys24 67	Cys26-68	Cys29-67	Metal 66-67-68	Energy	Ms
2.32	2.35	2.44	2.37	2.39	2.41	2.30	2.30	2.36	2.29	2.35	2.33	Cu-Cu-Zn	-9410.216853	0
2.30	2.40	2.33	2.32	2.41	2.40	2.34	2.32	2.34	2.42	2.30	2.37	Cu-Zn-Cu	-9410.219816	
2.36	**2.43**	**2.36**	**2.32**	**2.34**	**2.37**	**2.28**	**2.36**	**2.42**	**2.35**	**2.29**	**2.34**	**Zn-Cu-Cu**	**-9410.221564**	
2.31	**2.40**	**2.36**	**2.31**	**2.35**	**2.36**	**2.33**	**2.33**	**2.37**	**2.34**	**2.31**	**2.34**	**Cu-Cu-Cu**	**-9269.194671**	½

Table S21. Optimized angles of MT2-beta with restrained C-alpha; highest spin state (degrees).

C5-66-C7	C5-66-C21	C5-66-C24	C7-66-C21	C7-66-C24	C21-66-C24	C15-67-C19	C15-67-C24	C15-67-C29	C19-67-C24	C19-67-C29	C24-67-C29	C7-68-C13	C7-68-C15	C7-68-C26	C13-68-C15	C13-68-C26	C15-68-C26	Metal 66-67-68
99.7	**119.9**	**110.1**	**115.1**	**99.5**	**112.5**	**111.5**	**105.4**	**112.1**	**124.7**	**98.5**	**104.2**	**100.9**	**107.6**	**115.0**	**114.6**	**110.0**	**108.8**	**Zn-Zn-Zn**
93.8	**109.4**	**117.3**	**125.4**	**98.7**	**111.6**	**111.1**	**106.0**	**112.3**	**124.0**	**97.4**	**105.7**	**101.4**	**106.3**	**114.9**	**115.3**	**110.7**	**108.4**	**Cu-Zn-Zn**
102.0	117.5	108.6	107.7	105.7	114.1	106.4	101.8	115.8	137.3	95.8	100.2	103.8	108.2	106.1	115.1	109.7	113.0	Zn-Cu-Zn
98.5	116.0	110.5	116.0	101.0	113.7	113.5	103.4	113.2	124.5	98.3	103.6	94.1	108.9	130.9	125.8	99.6	100.4	Zn-Zn-Cu
93.4	**109.0**	**118.3**	**121.7**	**102.6**	**111.4**	**107.1**	**103.6**	**115.6**	**132.9**	**95.8**	**102.1**	**102.0**	**103.1**	**114.0**	**117.5**	**109.6**	**110.5**	**Cu-Cu-Zn**
94.1	107.8	118.4	124.7	100.6	111.3	113.5	104.2	113.7	123.1	97.6	104.4	94.4	107.1	129.9	125.9	101.8	100.8	Cu-Zn-Cu
103.8	118.2	103.3	110.9	101.6	117.1	106.0	99.9	120.4	139.2	94.3	99.2	94.8	101.9	135.8	135.7	96.3	99.4	Zn-Cu-Cu
92.7	**106.8**	**119.4**	**121.6**	**103.4**	**112.3**	**107.0**	**100.6**	**118.4**	**134.4**	**95.9**	**102.0**	**94.4**	**102.6**	**131.2**	**130.0**	**101.3**	**101.7**	**Cu-Cu-Cu**

Table S22. Optimized angles of MT2-beta with unrestrained C-alpha; highest spin state (degrees).

C5-66-C7	C5-66-C21	C5-66-C24	C7-66-C21	C7-66-C24	C21-66-C24	C15-67-C19	C15-67-C24	C15-67-C29	C19-67-C24	C19-67-C29	C24-67-C29	C7-68-C13	C7-68-C15	C7-68-C26	C13-68-C15	C13-68-C26	C15-68-C26	Metal 66-67-68
116.6	**110.0**	**107.8**	**105.9**	**98.9**	**117.6**	**109.5**	**95.9**	**110.8**	**122.6**	**110.4**	**106.6**	**103.9**	**114.0**	**110.0**	**110.8**	**110.6**	**107.5**	**Zn-Zn-Zn**
96.7	**96.4**	**135.5**	**141.1**	**88.9**	**106.5**	**112.7**	**103.4**	**109.7**	**116.2**	**106.4**	**108.3**	**103.0**	**99.4**	**121.7**	**110.4**	**109.2**	**112.4**	**Cu-Zn-Zn**
115.0	110.9	105.0	107.6	104.8	113.5	102.3	89.1	132.2	144.3	96.2	100.9	102.3	110.3	111.2	112.7	109.0	111.1	Zn-Cu-Zn
110.2	108.3	109.2	111.1	97.9	119.7	115.5	95.4	111.9	124.5	105.0	104.0	95.7	92.6	148.3	130.3	98.3	99.7	Zn-Zn-Cu
103.1	100.8	131.3	132.9	93.0	100.8	104.2	89.2	127.2	146.4	95.2	100.9	106.0	103.6	110.7	112.1	109.6	114.4	Cu-Cu-Zn
97.3	97.8	131.2	135.5	91.1	109.0	116.6	100.3	112.4	116.5	102.6	108.5	98.2	92.2	142.2	132.5	98.2	101.2	Cu-Zn-Cu
115.3	**111.4**	**111.4**	**107.1**	**103.1**	**108.0**	**103.0**	**87.1**	**136.2**	**152.9**	**91.8**	**98.1**	**93.0**	**94.8**	**146.8**	**136.3**	**100.5**	**96.0**	**Zn-Cu-Cu**
97.7	**96.7**	**131.2**	**134.9**	**93.4**	**108.3**	**105.8**	**91.2**	**131.8**	**140.5**	**91.6**	**103.4**	**96.0**	**87.0**	**150.8**	**137.1**	**97.4**	**100.5**	**Cu-Cu-Cu**

Table S20. Optimized angles of MT2-beta with restrained C-alpha; lowest spin state (degrees).

C5-66-C7	C5-66-C21	C5-66-C24	C7-66-C21	C7-66-C24	C21-66-C24	C15-67-C19	C15-67-C24	C15-67-C29	C19-67-C24	C19-67-C29	C24-67-C29	C7-68-C13	C7-68-C15	C7-68-C26	C13-68-C15	C13-68-C26	C15-68-C26	Metal 66-67-68
93.1	108.6	119.8	121.7	102.5	110.8	106.7	103.1	115.9	133.7	95.6	102.3	102.1	102.9	114.1	117.5	109.3	110.6	Cu-Cu-Zn
90.0	108.0	119.3	132.2	93.5	113.3	113.6	104.9	113.7	123.5	96.1	104.6	95.8	104.7	129.3	124.9	104.4	100.7	Cu-Zn-Cu
109.1	**107.7**	**109.0**	**114.1**	**104.8**	**112.0**	**98.6**	**85.3**	**141.9**	**150.9**	**94.5**	**100.1**	**93.6**	**83.1**	**155.1**	**138.5**	**101.5**	**98.0**	**Zn-Cu-Cu**
91.4	107.2	119.3	124.3	100.8	113.1	108.0	103.3	117.6	132.4	94.9	101.4	94.9	103.2	129.7	129.2	101.7	102.1	Cu-Cu-Cu

Table S21. Optimized angles of MT2-beta with unrestrained C-alpha; lowest spin state (degrees).

C5-66-C7	C5-66-C21	C5-66-C24	C7-66-C21	C7-66-C24	C21-66-C24	C15-67-C19	C15-67-C24	C15-67-C29	C19-67-C24	C19-67-C29	C24-67-C29	C7-68-C13	C7-68-C15	C7-68-C26	C13-68-C15	C13-68-C26	C15-68-C26	Metal 66-67-68
104.0	101.9	138.5	134.8	91.5	92.9	107.8	93.8	118.7	139.9	97.8	100.7	104.4	103.4	110.6	112.8	109.3	115.6	Cu-Cu-Zn
90.3	97.4	135.8	148.3	86.1	108.4	114.3	101.6	112.2	116.5	104.5	107.9	98.5	94.9	133.4	124.4	104.2	104.5	Cu-Zn-Cu
114.0	**109.8**	**109.2**	**107.3**	**107.0**	**109.5**	**93.1**	**89.8**	**145.4**	**154.4**	**91.7**	**100.2**	**96.9**	**87.9**	**144.8**	**143.8**	**104.3**	**91.0**	**Zn-Cu-Cu**
88.4	**98.5**	**140.1**	**154.0**	**83.4**	**105.9**	**103.9**	**93.8**	**131.3**	**138.9**	**91.1**	**104.5**	**94.1**	**89.3**	**146.4**	**140.3**	**97.5**	**101.1**	**Cu-Cu-Cu**

Table S22. Mulliken charges for High-Spin Models (a.u.).

	66		67		68	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Zn-Zn-Zn	0.44	0.45	0.43	0.42	0.47	0.49
Cu-Zn-Zn	0.12	0.11	0.43	0.41	0.46	0.46
Zn-Cu-Zn	0.45	0.44	0.11	0.08	0.45	0.48
Zn-Zn-Cu	0.44	0.51	0.42	0.40	0.15	0.11
Cu-Cu-Zn	0.12	0.13	0.11	0.07	0.45	0.45
Cu-Zn-Cu	0.12	0.10	0.42	0.42	0.14	0.11
Zn-Cu-Cu	0.45	0.49	0.11	0.06	0.14	0.08
Cu-Cu-Cu	0.12	0.09	0.10	0.05	0.13	0.09

Table S23. Mulliken charges for Low-Spin Models (a.u.).

	66		67		68	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Cu-Cu-Zn	0.12	0.07	0.11	0.05	0.45	0.43
Cu-Zn-Cu	0.09	0.07	0.43	0.41	0.12	0.09
Zn-Cu-Cu	0.51	0.51	0.04	0.02	0.06	0.05
Cu-Cu-Cu	0.11	0.08	0.10	0.05	0.12	0.07

Table S27. Molecular areas and volumes of MT models.

	Molecular area ($Bohr^2$)				Molecular volume ($Bohr^3$)			
	RHS	UHS	RLS	ULS	RHS	UHS	RLS	ULS
Zn-Zn-Zn	1903	1898			5316	5384		
Cu-Zn-Zn	1909	1874			5291	5266		
Zn-Cu-Zn	1870	1868			5247	5320		
Zn-Zn-Cu	1899	1890			5316	5351		
Cu-Cu-Zn	1906	1885	1899	1913	5289	5335	5274	5374
Cu-Zn-Cu	1907	1875	1914	1938	5294	5261	5295	5312
Zn-Cu-Cu	1895	1834	1876	1883	5327	5249	5311	5296
Cu-Cu-Cu	1904	1908	1912	1851	5292	5303	5302	5247

RHS: Restrained High-Spin. UHS: Unrestrained High-Spin. RLS: Restrained Low-Spin. ULS: Unrestrained Low-Spin.
RMS: Restrained Medium-Spin. UMS: Unrestrained Medium-Spin.

PART 3: MT3-alpha

Table S28. Optimized M-S bond lengths of MT3-alpha with restrained C-alpha; highest spin state (Å).

C34-69	C35-69	C35 – 70	C37-70	C38-70	C38-71	C42-71	C45-69	C45-71	C49-69	C51-70	C51-72	C64-72	C66-72	C67-71	C67-72	Metal 69-70-71-72	Energy	M_S
2.39	**2.45**	**2.53**	**2.35**	**2.49**	**2.43**	**2.40**	**2.57**	**2.49**	**2.35**	**2.48**	**2.50**	**2.34**	**2.39**	**2.70**	**2.47**	**Zn-Zn-Zn-Zn**	**-12444.498127**	**0**
2.31	2.37	2.52	2.35	2.48	2.42	2.39	2.61	2.44	2.30	2.46	2.51	2.35	2.40	2.87	2.45	Cu-Zn-Zn-Zn	-12303.469155	½
2.40	2.49	2.37	2.33	2.52	2.38	2.40	2.51	2.50	2.36	2.42	2.47	2.35	2.40	2.71	2.47	Zn-Cu-Zn-Zn	-12303.468105	
2.37	**2.42**	**2.53**	**2.35**	**2.45**	**2.32**	**2.24**	**2.49**	**2.31**	**2.35**	**2.45**	**2.52**	**2.37**	**2.40**	**3.77**	**2.38**	**Zn-Zn-Cu-Zn**	**-12303.480380**	
2.39	2.47	2.54	2.36	2.48	2.44	2.41	2.57	2.51	2.35	2.48	2.43	2.28	2.36	2.61	2.46	Zn-Zn-Zn-Cu	-12303.470915	
2.34	2.39	2.36	2.34	2.51	2.39	2.39	2.45	2.49	2.32	2.42	2.46	2.35	2.40	2.76	2.47	Cu-Cu-Zn-Zn	-12162.440541	1
2.31	**2.34**	**2.49**	**2.34**	**2.44**	**2.30**	**2.24**	**2.39**	**2.29**	**2.29**	**2.41**	**2.52**	**2.36**	**2.43**	**4.52**	**2.36**	**Cu-Zn-Cu-Zn**	**-12162.465120**	
2.32	2.42	2.52	2.36	2.47	2.44	2.41	2.59	2.48	2.30	2.47	2.42	2.28	2.36	2.62	2.46	Cu-Zn-Zn-Cu	-12162.441163	
2.37	2.41	2.40	2.32	2.47	2.27	2.26	2.46	2.30	2.35	2.35	2.51	2.36	2.41	4.42	2.35	Zn-Cu-Cu-Zn	-12162.461426	
2.39	2.50	2.41	2.34	2.51	2.39	2.41	2.53	2.51	2.36	2.41	2.38	2.28	2.37	2.62	2.46	Zn-Cu-Zn-Cu	-12162.444621	
2.38	2.44	2.53	2.36	2.44	2.33	2.25	2.49	2.33	2.35	2.47	2.45	2.29	2.35	3.56	2.35	Zn-Zn-Cu-Cu	-12162.450476	
2.31	**2.34**	**2.40**	**2.31**	**2.46**	**2.27**	**2.26**	**2.39**	**2.29**	**2.30**	**2.36**	**2.50**	**2.36**	**2.42**	**4.50**	**2.35**	**Cu-Cu-Cu-Zn**	**-12021.441620**	**3/2**
2.34	2.40	2.38	2.35	2.51	2.40	2.40	2.47	2.52	2.32	2.42	2.37	2.28	2.37	2.65	2.46	Cu-Cu-Zn-Cu	-12021.416308	
2.31	2.34	2.49	2.34	2.44	2.31	2.24	2.39	2.29	2.30	2.40	2.50	2.31	2.34	4.64	2.31	Cu-Zn-Cu-Cu	-12021.440281	
2.38	2.45	2.42	2.34	2.44	2.30	2.25	2.49	2.32	2.35	2.42	2.39	2.30	2.35	3.61	2.35	Zn-Cu-Cu-Cu	-12021.424571	
2.31	**2.34**	**2.40**	**2.32**	**2.47**	**2.27**	**2.26**	**2.40**	**2.29**	**2.31**	**2.34**	**2.50**	**2.31**	**2.33**	**4.64**	**2.31**	**Cu-Cu-Cu-Cu**	**-11880.417804**	**2**

Table S29. Optimized M-S bond lengths of MT3-alpha with unrestrained C-alpha; highest spin state (Å).

C34-69	C35-69	C35 – 70	C37-70	C38-70	C38-71	C42-71	C45-69	C45-71	C49-69	C51-70	C51-72	C64-72	C66-72	C67-71	C67-72	Metal 69-70-71-72	Energy	M_S
2.35	**2.43**	**2.41**	**2.34**	**2.39**	**2.40**	**2.34**	**2.43**	**2.39**	**2.37**	**2.40**	**2.44**	**2.36**	**2.36**	**2.42**	**2.43**	**Zn-Zn-Zn-Zn**	**-12444.551488**	**0**
2.32	2.37	2.41	2.34	2.40	2.41	2.34	2.37	2.39	2.33	2.40	2.45	2.37	2.36	2.42	2.43	Cu-Zn-Zn-Zn	-12303.526705	½
2.37	2.45	2.35	2.31	2.35	2.40	2.34	2.44	2.39	2.37	2.33	2.41	2.35	2.36	2.42	2.43	Zn-Cu-Zn-Zn	-12303.521581	
2.37	2.41	2.43	2.34	2.39	2.37	2.31	2.48	2.33	2.36	2.39	2.43	2.35	2.36	2.40	2.43	Zn-Zn-Cu-Zn	-12303.520746	
2.36	**2.42**	**2.43**	**2.34**	**2.40**	**2.41**	**2.36**	**2.46**	**2.41**	**2.36**	**2.39**	**2.39**	**2.30**	**2.32**	**2.39**	**2.39**	**Zn-Zn-Zn-Cu**	**-12303.529559**	
2.32	2.36	2.35	2.32	2.36	2.38	2.34	2.37	2.39	2.33	2.36	2.41	2.35	2.36	2.41	2.45	Cu-Cu-Zn-Zn	-12162.505976	1
2.31	**2.40**	**2.41**	**2.34**	**2.38**	**2.36**	**2.30**	**2.36**	**2.32**	**2.32**	**2.40**	**2.45**	**2.36**	**2.36**	**2.40**	**2.42**	**Cu-Zn-Cu-Zn**	**-12162.506764**	
2.33	2.37	2.41	2.35	2.41	2.42	2.37	2.42	2.41	2.32	2.39	2.36	2.30	2.32	2.39	2.40	Cu-Zn-Zn-Cu	-12162.504072	
2.36	2.40	2.40	2.28	2.33	2.30	2.22	2.46	2.29	2.36	2.33	2.44	2.37	2.38	6.66	2.38	Zn-Cu-Cu-Zn	-12162.489562	
2.37	2.44	2.35	2.33	2.37	2.38	2.37	2.46	2.40	2.36	2.32	2.34	2.31	2.32	2.40	2.39	Zn-Cu-Zn-Cu	-12162.499952	
2.37	2.41	2.44	2.34	2.38	2.37	2.32	2.46	2.34	2.36	2.38	2.38	2.31	2.32	2.38	2.36	Zn-Zn-Cu-Cu	-12162.498861	
2.32	2.33	2.39	2.29	2.36	2.29	2.22	2.38	2.38	2.32	2.33	2.44	2.37	2.39	5.91	2.38	Cu-Cu-Cu-Zn	-12021.472281	3/2
2.31	**2.37**	**2.34**	**2.33**	**2.37**	**2.38**	**2.35**	**2.37**	**2.40**	**2.33**	**2.34**	**2.34**	**2.31**	**2.33**	**2.41**	**2.40**	**Cu-Cu-Zn-Cu**	**-12021.474956**	
2.33	2.36	2.41	2.34	2.38	2.35	2.30	2.36	2.34	2.33	2.41	2.37	2.32	2.33	2.39	2.40	Cu-Zn-Cu-Cu	-12021.472057	
2.37	2.43	2.35	2.34	2.36	2.34	2.31	2.47	2.34	2.36	2.33	2.34	2.30	2.33	2.39	2.35	Zn-Cu-Cu-Cu	-12021.474059	
2.32	**2.37**	**2.33**	**2.34**	**2.35**	**2.35**	**2.30**	**2.36**	**2.32**	**2.32**	**2.35**	**2.36**	**2.31**	**2.32**	**2.38**	**2.36**	**Cu-Cu-Cu-Cu**	**-11880.459435**	**2**

Table S30. Optimized M-S bond lengths of MT3-alpha with restrained C-alpha; lowest spin state (Å).

C34-69	C35-69	C35 – 70	C37-70	C38-70	C38-71	C42-71	C45-69	C45-71	C49-69	C51-70	C51-72	C64-72	C66-72	C67-71	C67-72	Metal 69-70-71-72	E_{el}(a.u.)	M_S
2.38	2.33	2.31	2.39	2.55	2.39	2.42	2.53	2.44	2.37	2.40	2.45	2.36	2.41	2.70	2.47	Cu-Cu-Zn-Zn	-12162.442358	0
2.32	**2.34**	**2.48**	**2.34**	**2.43**	**2.30**	**2.24**	**2.35**	**2.30**	**2.28**	**2.41**	**2.52**	**2.36**	**2.42**	**4.54**	**2.36**	**Cu-Zn-Cu-Zn**	**-12162.458810**	
2.32	2.39	2.51	2.36	2.47	2.44	2.41	2.66	2.45	2.31	2.49	2.41	2.28	2.36	2.66	2.44	Cu-Zn-Zn-Cu	-12162.431994	
2.36	2.43	2.34	2.34	2.43	2.26	2.25	2.42	2.30	2.34	2.38	2.49	2.36	2.42	4.44	2.36	Zn-Cu-Cu-Zn	-12162.455939	
2.40	2.47	2.43	2.34	2.49	2.39	2.41	2.55	2.50	2.36	2.46	2.31	2.27	2.35	2.67	2.46	Zn-Cu-Zn-Cu	-12162.439449	
2.38	2.45	2.53	2.36	2.41	2.35	2.26	2.46	2.31	2.36	2.53	2.40	2.26	2.36	3.67	2.34	Zn-Zn-Cu-Cu	-12162.447844	
2.31	2.35	2.36	2.33	2.47	2.27	2.25	2.37	2.29	2.30	2.36	2.50	2.36	2.42	4.52	2.35	Cu-Cu-Cu-Zn	-12021.440563	½
2.34	2.38	2.33	2.37	2.54	2.40	2.42	2.53	2.46	2.35	2.43	2.38	2.28	2.37	2.62	2.46	Cu-Cu-Zn-Cu	-12021.415465	
2.32	2.35	2.47	2.34	2.43	2.32	2.25	2.40	2.29	2.31	2.43	2.47	2.30	2.32	4.69	2.31	Cu-Zn-Cu-Cu	-12021.440359	
2.37	**2.40**	**2.41**	**2.33**	**2.45**	**2.27**	**2.27**	**2.46**	**2.30**	**2.36**	**2.37**	**2.44**	**2.29**	**2.31**	**4.70**	**2.31**	**Zn-Cu-Cu-Cu**	**-12021.440853**	
2.33	**2.36**	**2.34**	**2.33**	**2.33**	**2.35**	**2.32**	**2.35**	**2.32**	**2.32**	**2.39**	**2.35**	**2.30**	**2.31**	**2.41**	**2.34**	**Cu-Cu-Cu-Cu**	**-11880.451356**	**0**

Table S31. Optimized M-S bond lengths of MT3-alpha with unrestrained C-alpha; lowest spin state (Å).

C34-69	C35-69	C35 – 70	C37-70	C38-70	C38-71	C42-71	C45-69	C45-71	C49-69	C51-70	C51-72	C64-72	C66-72	C67-71	C67-72	Metal 69-70-71-72	E_{el}(a.u.)	M_S
2.32	2.39	2.29	2.35	2.46	2.39	2.37	2.42	2.40	2.35	2.30	2.42	2.35	2.36	2.43	2.43	Cu-Cu-Zn-Zn	-12162.492060	0
2.31	2.35	2.44	2.35	2.40	2.30	2.23	2.33	2.30	2.30	2.36	2.44	2.38	2.39	4.61	2.37	Cu-Zn-Cu-Zn	-12162.491072	
2.32	2.35	2.42	2.34	2.41	2.42	2.36	2.40	2.40	2.33	2.38	2.35	2.31	2.33	2.40	2.40	Cu-Zn-Zn-Cu	-12162.490547	
2.36	2.45	2.34	2.28	2.34	2.26	2.24	2.41	2.30	2.35	230	2.49	2.35	2.37	7.06	2.35	Zn-Cu-Cu-Zn	-12162.497409	
2.36	**2.43**	**2.36**	**2.29**	**2.36**	**2.39**	**2.35**	**2.44**	**2.40**	**2.37**	**2.35**	**2.31**	**2.30**	**2.31**	**2.40**	**2.36**	**Zn-Cu-Zn-Cu**	**-12162.504571**	
2.36	2.44	2.40	2.34	2.37	2.58	2.35	2.44	2.41	2.37	2.46	2.36	2.29	2.33	2.40	2.38	Zn-Zn-Cu-Cu	-12162.500418	
2.30	2.34	2.35	2.35	2.36	2.30	2.25	2.33	2.30	2.28	2.33	2.44	2.37	2.38	6.17	2.37	Cu-Cu-Cu-Zn	-12021.479967	½
2.32	**2.36**	**2.34**	**2.29**	**2.35**	**2.39**	**2.35**	**2.37**	**2.40**	**2.32**	**2.35**	**2.34**	**2.30**	**2.32**	**2.41**	**2.36**	**Cu-Cu-Zn-Cu**	**-12021.483555**	
2.31	2.37	2.45	2.35	2.37	2.37	2.31	2.33	2.33	2.32	2.40	2.37	2.32	2.33	2.40	2.40	Cu-Zn-Cu-Cu	-12021.479006	
2.38	2.41	2.36	2.30	2.41	2.25	2.27	2.44	2.30	2.36	2.25	2.38	2.27	2.30	3.12	2.28	Zn-Cu-Cu-Cu	-12021.481723	
2.33	**2.34**	**2.40**	**2.28**	**2.34**	**2.27**	**2.28**	**2.35**	**2.30**	**2.30**	**2.35**	**2.35**	**2.29**	**2.30**	**5.95**	**2.31**	**Cu-Cu-Cu-Cu**	**-11880.459202**	**0**

Table S32. Optimized M-S bond lengths of MT3-alpha with restrained C-alpha; medium spin state (Å).

C34-69	C35-69	C35 – 70	C37-70	C38-70	C38-71	C42-71	C45-69	C45-71	C49-69	C51-70	C51-72	C64-72	C66-72	C67-71	C67-72	Metal 69-70-71-72	E_{el}(a.u.)	M_S
2.32	**2.37**	**2.33**	**2.33**	**2.34**	**2.35**	**2.30**	**2.36**	**2.32**	**2.31**	**2.35**	**2.35**	**2.31**	**2.32**	**2.38**	**2.37**	**Cu-Cu-Cu-Cu**	**-11880.458825**	**1**

Table S33. Optimized M-S bond lengths of MT3-alpha with unrestrained C-alpha; lowest spin state (Å).

C34-69	C35-69	C35 – 70	C37-70	C38-70	C38-71	C42-71	C45-69	C45-71	C49-69	C51-70	C51-72	C64-72	C66-72	C67-71	C67-72	Metal 69-70-71-72	E_{el}(a.u.)	M_S
2.33	**2.34**	**2.36**	**2.29**	**2.31**	**2.39**	**2.33**	**2.35**	**2.34**	**2.34**	**2.40**	**2.32**	**2.30**	**2.30**	**2.36**	**2.35**	**Cu-Cu-Cu-Cu**	**-11880.456787**	**1**

Table S34. Optimized angles of MT3-alpha with restrained C-alpha; high-spin state (degrees).

C34-69-C35	C34-69-C45	C34-69-C49	C35-69-C45	C35-69-C49	C45-69-C49	C35-70-C37	C35-70-C38	C35-70-C51	C37-70-C38	C37-70-C38	C38-70-C51	C38-71-C42	C38-71-C45	C38-71-C67	C42-71-C45	C42-71-C67	C45-71-C67	C51-72-C64	C51-72-C66	C51-72-C67	C64-72-C66	C64-72-C67	C66-72-C67	Metal 69-70-71-72
109.4	**107.9**	**108.8**	**110.7**	**108.0**	**112.0**	**104.4**	**98.2**	**118.1**	**115.5**	**117.1**	**102.3**	**123.4**	**105.8**	**96.9**	**110.0**	**105.7**	**114.6**	**115.5**	**104.7**	**105.1**	**114.3**	**103.2**	**113.9**	**Zn-Zn-Zn-Zn**
124.4	106.9	99.5	104.5	105.5	117.1	103.6	99.9	116.2	114.0	119.0	102.8	125.1	105.3	93.6	111.6	102.2	118.6	113.9	104.7	105.3	113.9	104.5	114.5	Cu-Zn-Zn-Zn
102.9	109.3	108.7	115.9	107.8	111.7	93.6	98.0	129.5	112.6	119.2	102.3	125.4	104.2	98.4	109.3	106.2	113.1	112.6	106.5	106.2	113.6	104.7	113.1	Zn-Cu-Zn-Zn
117.3	**104.3**	**108.9**	**106.7**	**106.7**	**113.2**	**102.8**	**94.1**	**119.2**	**116.2**	**119.0**	**103.5**	**129.7**	**111.1**	**82.5**	**118.6**	**93.1**	**104.1**	**110.3**	**108.7**	**105.6**	**112.0**	**106.9**	**113.1**	**Zn-Zn-Cu-Zn**
108.9	108.6	108.6	111.3	107.1	112.2	106.0	98.5	119.2	112.7	115.7	103.5	122.8	104.3	97.5	109.0	108.0	115.2	131.9	106.9	105.9	97.4	96.8	119.1	Zn-Zn-Zn-Cu
111.7	108.5	99.0	114.2	104.6	117.9	97.0	100.8	126.5	112.0	117.1	102.5	127.7	101.9	97.4	109.4	104.5	116.3	114.3	104.6	107.0	113.9	103.5	113.7	Cu-Cu-Zn-Zn
129.2	**102.9**	**101.8**	**102.6**	**101.2**	**121.5**	**97.0**	**94.4**	**121.0**	**109.3**	**125.8**	**104.9**	**130.6**	**113.9**	**63.3**	**115.1**	**93.8**	**110.6**	**101.0**	**119.5**	**99.3**	**103.4**	**127.8**	**107.1**	**Cu-Zn-Cu-Zn**
119.1	110.9	99.2	107.1	102.7	118.0	106.3	100.7	117.6	111.2	116.3	103.5	123.2	102.5	97.1	108.8	106.7	119.0	131.9	106.2	106.2	97.6	96.6	119.8	Cu-Zn-Zn-Cu
115.1	104.2	109.3	105.2	105.5	118.0	89.7	92.1	130.5	104.8	128.3	104.4	132.5	113.6	65.4	113.2	96.4	103.1	98.8	121.8	98.8	104.0	127.8	107.3	Zn-Cu-Cu-Zn
104.0	108.8	109.2	115.6	108.3	110.6	99.9	97.8	128.9	113.0	113.7	102.7	124.8	103.6	98.9	108.3	107.8	113.3	128.7	106.5	108.3	98.5	96.8	119.0	Zn-Cu-Zn-Cu
116.1	104.3	108.2	106.8	106.7	115.1	105.2	95.3	120.9	115.2	115.3	103.7	129.9	110.4	82.6	118.3	93.8	106.7	126.0	106.4	106.7	102.0	96.2	120.8	Zn-Zn-Cu-Cu
125.2	**102.7**	**100.5**	**104.1**	**102.8**	**123.7**	**89.8**	**94.7**	**126.4**	**103.1**	**131.3**	**104.6**	**131.3**	**114.6**	**64.4**	**113.1**	**94.4**	**108.9**	**99.5**	**121.1**	**99.4**	**103.5**	**127.8**	**107.2**	**Cu-Cu-Cu-Zn**
111.8	109.3	99.5	114.0	103.5	117.7	100.2	100.6	126.4	111.3	114.5	102.7	126.8	100.7	98.2	107.8	107.0	116.8	130.3	105.9	107.7	98.2	96.4	119.6	Cu-Cu-Zn-Cu
129.0	103.2	101.4	102.6	101.3	121.6	96.1	94.1	123.5	109.5	124.2	104.0	129.5	114.1	63.8	116.0	94.7	107.8	101.1	126.6	96.0	96.9	138.8	102.4	Cu-Zn-Cu-Cu
114.2	103.3	108.6	110.1	107.5	113.2	101.3	93.2	128.3	116.6	113.3	103.5	133.2	108.0	84.5	117.7	93.6	104.2	122.9	106.6	109.8	102.0	96.5	119.8	Zn-Cu-Cu-Cu
126.5	**102.4**	**101.2**	**103.6**	**102.0**	**123.4**	**90.7**	**94.2**	**128.3**	**103.8**	**129.4**	**103.7**	**131.5**	**114.9**	**64.5**	**113.1**	**95.2**	**107.0**	**99.6**	**125.2**	**96.3**	**97.3**	**140.8**	**102.3**	**Cu-Cu-Cu-Cu**

Table S35. Optimized angles of MT3-alpha with unrestrained C-alpha; high-spin state (degrees).

C34-69-C35	C34-69-C45	C34-69-C49	C35-69-C45	C35-69-C49	C45-69-C49	C35-70-C37	C35-70-C38	C35-70-C51	C37-70-C38	C37-70-C38	C38-70-C51	C38-71-C42	C38-71-C45	C38-71-C67	C42-71-C45	C42-71-C67	C45-71-C67	C51-72-C64	C51-72-C66	C51-72-C67	C64-72-C66	C64-72-C67	C66-72-C67	Metal 69-70-71-72
115.1	**106.5**	**108.7**	**106.6**	**104.8**	**115.6**	**105.9**	**105.4**	**111.3**	**115.6**	**112.2**	**106.3**	**115.8**	**112.7**	**100.4**	**114.6**	**103.9**	**107.5**	**116.8**	**100.9**	**107.0**	**113.4**	**107.7**	**110.9**	**Zn-Zn-Zn-Zn**
138.6	99.3	97.6	94.5	99.0	135.9	106.4	101.8	114.8	115.5	112.9	105.2	114.7	116.0	98.1	110.8	110.8	105.2	105.9	104.8	110.8	114.8	112.3	108.0	Cu-Zn-Zn-Zn
114.0	103.9	107.7	111.3	104.1	116.1	103.2	98.4	130.4	137.5	102.0	90.8	112.1	114.2	100.5	112.8	107.8	108.5	117.0	102.8	101.6	113.6	110.5	110.5	Zn-Cu-Zn-Zn
118.6	100.5	108.6	107.0	104.6	118.3	103.9	102.3	113.5	123.6	112.4	101.1	128.7	101.3	96.0	113.0	90.9	129.1	120.8	105.1	99.1	114.4	107.7	108.3	Zn-Zn-Cu-Zn
113.1	**103.6**	**109.3**	**111.2**	**106.1**	**113.8**	**108.7**	**102.4**	**110.1**	**116.7**	**108.5**	**110.2**	**106.9**	**111.0**	**109.2**	**109.9**	**112.3**	**107.6**	**138.3**	**98.7**	**92.1**	**99.8**	**100.6**	**134.6**	**Zn-Zn-Zn-Cu**
133.4	98.7	98.1	101.3	99.1	131.6	100.6	96.5	130.5	133.4	103.5	97.3	116.6	108.8	103.4	112.2	105.7	109.5	105.8	114.0	104.1	109.7	115.7	107.6	Cu-Cu-Zn-Zn
143.2	**96.4**	**100.6**	**95.1**	**92.6**	**139.9**	**107.0**	**108.4**	**110.7**	**112.0**	**113.1**	**105.7**	**132.9**	**98.9**	**96.3**	**109.2**	**95.9**	**126.8**	**110.3**	**103.7**	**110.5**	**111.0**	**110.5**	**110.7**	**Cu-Zn-Cu-Zn**
132.6	97.4	96.3	103.3	98.7	134.3	110.8	103.5	107.5	113.6	109.2	112.0	107.7	106.8	109.6	110.3	110.4	111.9	139.2	97.4	89.8	101.8	101.5	134.3	Cu-Zn-Zn-Cu
116.1	102.8	108.3	105.7	106.2	118.2	97.7	112.3	112.6	105.7	134.4	93.8	123.5	107.0	26.8	129.1	139.1	85.6	107.0	110.8	104.3	107.2	120.0	107.5	Zn-Cu-Cu-Zn
113.1	102.2	109.3	115.0	103.4	114.0	102.0	101.0	122.8	126.9	102.8	103.5	108.8	108.6	109.4	111.1	110.1	108.8	133.7	98.6	91.2	102.4	102.7	133.8	Zn-Cu-Zn-Cu
116.7	102.1	108.4	108.2	104.8	117.2	105.8	102.4	113.5	117.6	112.4	105.1	128.8	103.0	95.0	112.7	92.8	125.7	140.9	96.7	91.2	101.5	100.2	133.6	Zn-Zn-Cu-Cu
138.8	98.8	99.7	91.9	98.9	137.3	90.3	125.7	106.9	108.6	142.3	88.6	123.8	102.5	14.1	132.2	126.2	101.6	107.0	109.8	106.4	107.3	118.9	107.4	Cu-Cu-Cu-Zn
131.9	**98.5**	**98.4**	**104.6**	**99.0**	**128.5**	**104.2**	**98.3**	**125.9**	**129.1**	**102.8**	**99.6**	**116.8**	**109.1**	**103.1**	**111.2**	**104.6**	**111.6**	**132.2**	**98.8**	**94.2**	**101.2**	**102.6**	**133.1**	**Cu-Cu-Zn-Cu**
135.9	98.4	99.2	95.3	99.1	136.3	106.9	104.3	113.0	114.9	115.0	102.4	127.4	106.5	93.7	108.9	103.5	116.5	95.8	142.4	93.5	94.8	146.2	97.3	Cu-Zn-Cu-Cu
115.3	101.1	108.7	112.1	103.2	116.9	102.0	97.5	131.6	132.9	101.1	96.9	130.3	97.3	93.3	115.1	92.4	130.9	133.1	99.4	93.1	102.9	105.2	126.6	Zn-Cu-Cu-Cu
131.0	**97.1**	**101.4**	**101.1**	**98.8**	**132.6**	**102.2**	**96.6**	**134.6**	**130.2**	**102.9**	**95.3**	**133.0**	**96.0**	**93.3**	**111.4**	**95.7**	**131.3**	**132.8**	**98.8**	**97.3**	**100.0**	**102.4**	**130.6**	**Cu-Cu-Cu-Cu**

Table S36. Optimized angles of MT3-alpha with restrained C-alpha; low-spin state (degrees).

C34-69-C35	C34-69-C45	C34-69-C49	C35-69-C45	C35-69-C49	C45-69-C49	C35-70-C37	C35-70-C38	C35-70-C51	C37-70-C38	C37-70-C38	C38-70-C51	C38-71-C42	C38-71-C45	C38-71-C67	C42-71-C45	C42-71-C67	C45-71-C67	C51-72-C64	C51-72-C66	C51-72-C67	C64-72-C66	C64-72-C67	C66-72-C67	Metal 69-70-71-72
79.0	111.3	106.6	126.4	115.5	111.3	79.7	99.8	139.4	104.8	123.9	103.9	122.6	105.7	97.8	111.9	107.1	110.6	112.4	106.8	105.5	112.8	106.9	112.3	Cu-Cu-Zn-Zn
129.2	**109.0**	**100.3**	**104.9**	**96.9**	**116.6**	**96.7**	**94.2**	**119.6**	**109.2**	**126.8**	**105.6**	**129.0**	**113.0**	**64.0**	**117.4**	**93.9**	**105.9**	**101.0**	**119.5**	**99.1**	**103.6**	**127.7**	**107.4**	**Cu-Zn-Cu-Zn**
121.7	109.9	101.8	105.9	103.1	114.7	106.7	100.9	118.0	111.3	115.7	103.2	122.9	103.3	95.8	110.0	106.2	118.9	133.5	106.0	106.1	95.8	96.6	120.6	Cu-Zn-Zn-Cu
113.5	104.2	109.8	106.9	105.4	117.3	92.4	90.5	131.3	110.2	123.0	104.5	133.9	110.8	66.6	114.7	96.2	102.4	97.4	122.9	99.9	103.7	127.9	106.9	Zn-Cu-Cu-Zn
106.7	108.4	108.4	114.5	109.2	109.5	104.6	99.1	131.9	113.7	106.1	101.5	124.8	104.7	96.6	108.7	108.2	113.5	130.9	108.6	108.2	93.5	95.9	120.6	Zn-Cu-Zn-Cu
115.9	104.5	108.0	107.4	106.5	114.9	106.4	96.5	119.9	115.8	113.5	103.9	124.9	113.6	77.3	121.0	93.8	105.7	140.1	110.2	102.5	88.1	94.1	125.0	Zn-Zn-Cu-Cu
125.4	102.6	100.7	104.5	102.3	123.4	90.9	93.9	127.9	104.5	128.5	104.7	133.1	112.8	65.0	113.6	94.3	108.8	98.3	121.7	99.9	103.6	127.9	107.1	Cu-Cu-Cu-Zn
93.4	107.8	100.7	123.0	115.7	111.3	94.3	99.2	136.5	108.0	113.7	102.5	123.0	104.0	98.3	110.6	107.9	112.5	132.1	106.6	106.6	97.0	96.7	119.2	Cu-Cu-Zn-Cu
128.8	104.0	100.4	102.3	101.7	121.9	98.1	95.9	123.1	110.5	122.7	103.4	127.3	114.3	65.5	118.0	94.3	105.3	100.3	129.9	95.5	95.5	142.7	99.8	Cu-Zn-Cu-Cu
117.8	**104.0**	**108.2**	**104.7**	**105.0**	**117.6**	**91.9**	**91.5**	**132.4**	**107.8**	**124.9**	**102.5**	**132.1**	**113.8**	**67.2**	**113.3**	**97.4**	**98.7**	**97.5**	**140.7**	**93.9**	**95.6**	**144.1**	**96.8**	**Zn-Cu-Cu-Cu**
133.6	**101.1**	**101.5**	**99.1**	**97.3**	**128.9**	**102.7**	**97.3**	**128.6**	**127.5**	**105.6**	**98.1**	**127.1**	**100.8**	**92.2**	**113.2**	**95.4**	**129.8**	**139.7**	**98.3**	**94.3**	**96.7**	**101.7**	**133.2**	**Cu-Cu-Cu-Cu**

Table S37. Optimized angles of MT3-alpha with unrestrained C-alpha; low-spin state (degrees).

C34-69-C35	C34-69-C45	C34-69-C49	C35-69-C45	C35-69-C49	C45-69-C49	C35-70-C37	C35-70-C38	C35-70-C51	C37-70-C38	C37-70-C38	C38-70-C51	C38-71-C42	C38-71-C45	C38-71-C67	C42-71-C45	C42-71-C67	C45-71-C67	C51-72-C64	C51-72-C66	C51-72-C67	C64-72-C66	C64-72-C67	C66-72-C67	Metal 69-70-71-72
113.4	95.8	97.9	121.5	103.8	121.7	101.1	94.1	146.2	125.2	97.7	97.6	120.1	104.0	102.0	114.5	96.0	120.6	119.6	108.1	94.3	112.4	110.1	110.9	Cu-Cu-Zn-Zn
144.9	98.1	93.8	93.2	97.3	142.3	105.3	97.5	110.9	109.5	115.2	116.5	131.4	104.9	68.9	123.4	128.2	62.3	108.9	106.0	110.6	108.6	113.7	108.8	Cu-Zn-Cu-Zn
135.8	97.9	98.0	99.2	99.0	133.6	110.2	102.9	106.3	114.6	112.0	110.2	116.0	108.2	103.5	110.4	101.6	117.2	139.1	97.4	90.8	100.7	101.9	134.1	Cu-Zn-Zn-Cu
115.0	105.9	109.4	101.6	105.3	119.9	99.4	89.6	154.6	160.0	91.9	87.2	126.4	108.1	18.0	125.0	134.5	96.0	111.5	95.7	107.2	110.0	115.8	114.8	Zn-Cu-Cu-Zn
116.3	**103.5**	**109.0**	**109.6**	**103.3**	**115.7**	**102.3**	**100.8**	**139.2**	**137.2**	**91.0**	**94.6**	**115.8**	**113.5**	**104.3**	**112.8**	**102.4**	**106.5**	**136.6**	**97.0**	**94.7**	**103.3**	**100.7**	**129.8**	**Zn-Cu-Zn-Cu**
113.9	105.5	108.8	108.9	103.4	116.6	111.2	104.7	109.3	115.6	106.4	109.5	107.1	106.0	92.7	108.4	89.3	148.4	164.9	93.3	74.1	101.6	91.9	161.8	Zn-Zn-Cu-Cu
149.8	96.7	97.9.	85.0	94.3	151.6	82.4	139.9	107.0	103.3	146.6	90.4	122.7	111.3	22.8	124.3	128.5	105.5	108.1	108.8	106.8	108.5	115.8	108.6	Cu-Cu-Cu-Zn
138.0	**98.9**	**98.3**	**96.5**	**97.6**	**135.4**	**102.3**	**98.1**	**142.2**	**141.4**	**92.1**	**91.4**	**114.0**	**113.2**	**104.9**	**110.4**	**105.2**	**108.6**	**139.1**	**95.6**	**93.6**	**102.7**	**100.7**	**131.3**	**Cu-Cu-Zn-Cu**
146.1	91.0	96.4	84.3	94.9	167.6	107.1	107.5	104.6	110.1	116.5	110.6	116.7	124.3	96.2	91.2	120.0	110.7	96.3	145.8	89.1	95.8	146.5	98.0	Cu-Zn-Cu-Cu
116.5	104.5	106.4	104.1	107.6	118.3	102.4	104.0	118.9	119.0	119.3	93.1	126.9	117.9	77.5	114.6	110.2	92.7	92.7	154.4	79.2	91.5	163.0	102.3	Zn-Cu-Cu-Cu
151.2	**98.2**	**94.1**	**90.5**	**94.3**	**144.7**	**101.5**	**91.2**	**110.3**	**117.4**	**124.5**	**106.4**	**120.4**	**113.8**	**35.0**	**124.7**	**128.4**	**91.0**	**91.6**	**166.0**	**82.5**	**91.3**	**171.8**	**95.8**	**Cu-Cu-Cu-Cu**

Table S38. Optimized angles of MT3-alpha with restrained C-alpha; medium-spin state (degrees).

C34-69-C35	C34-69-C45	C34-69-C49	C35-69-C45	C35-69-C49	C45-69-C49	C35-70-C37	C35-70-C38	C35-70-C51	C37-70-C38	C37-70-C38	C38-70-C51	C38-71-C42	C38-71-C45	C38-71-C67	C42-71-C45	C42-71-C67	C45-71-C67	C51-72-C64	C51-72-C66	C51-72-C67	C64-72-C66	C64-72-C67	C66-72-C67	Metal 69-70-71-72
133.4	**98.5**	**101.1**	**99.3**	**97.5**	**132.7**	**102.3**	**97.9**	**134.6**	**130.4**	**101.4**	**95.4**	**132.6**	**96.5**	**93.6**	**110.6**	**95.6**	**132.2**	**132.2**	**98.6**	**97.2**	**100.4**	**102.9**	**130.3**	**Cu-Cu-Cu-Cu**

Table S39. Optimized angles of MT3-alpha with unrestrained C-alpha; medium-spin state (degrees).

C34-69-C35	C34-69-C45	C34-69-C49	C35-69-C45	C35-69-C49	C45-69-C49	C35-70-C37	C35-70-C38	C35-70-C51	C37-70-C38	C37-70-C38	C38-70-C51	C38-71-C42	C38-71-C45	C38-71-C67	C42-71-C45	C42-71-C67	C45-71-C67	C51-72-C64	C51-72-C66	C51-72-C67	C64-72-C66	C64-72-C67	C66-72-C67	Metal 69-70-71-72
130.2	**99.2**	**98.8**	**103.7**	**99.4**	**129.8**	**102.7**	**101.5**	**116.5**	**134.4**	**106.6**	**96.0**	**123.1**	**101.9**	**96.5**	**113.6**	**95.3**	**127.6**	**91.6**	**164.6**	**83.8**	**94.8**	**162.9**	**93.9**	**Cu-Cu-Cu-Cu**

Table S40. Mulliken charges for High-Spin Models (a.u.).

	69		70		71		72	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Zn-Zn-Zn-Zn	0.45	0.47	0.51	0.46	0.47	0.49	0.45	0.45
Cu-Zn-Zn-Zn	0.16	0.12	0.52	0.47	0.47	0.47	0.45	0.46
Zn-Cu-Zn-Zn	0.48	0.51	0.19	0.11	0.46	0.48	0.45	0.46
Zn-Zn-Cu-Zn	0.45	0.47	0.50	0.48	0.14	0.09	0.44	0.45
Zn-Zn-Zn-Cu	0.48	0.46	0.51	0.49	0.47	0.46	0.11	0.08
Cu-Cu-Zn-Zn	0.15	0.11	0.21	0.14	0.46	0.47	0.45	0.47
Cu-Zn-Cu-Zn	0.09	0.07	0.49	0.45	0.10	0.13	0.43	0.43
Cu-Zn-Zn-Cu	0.17	0.13	0.52	0.50	0.47	0.43	0.11	0.08
Zn-Cu-Cu-Zn	0.44	0.49	0.16	0.11	0.09	0.07	0.43	0.46
Zn-Cu-Zn-Cu	0.48	0.52	0.21	0.14	0.46	0.46	0.11	0.08
Zn-Zn-Cu-Cu	0.45	0.49	0.50	0.50	0.15	0.09	0.12	0.08
Cu-Cu-Cu-Zn	0.10	0.11	0.17	0.09	0.09	0.05	0.43	0.47
Cu-Cu-Zn-Cu	0.15	0.13	0.22	0.15	0.46	0.46	0.12	0.08
Cu-Zn-Cu-Cu	0.10	0.09	0.49	0.51	0.10	0.11	0.11	0.08
Zn-Cu-Cu-Cu	0.46	0.52	0.21	0.15	0.13	0.07	0.11	0.08
Cu-Cu-Cu-Cu	0.10	0.09	0.18	0.15	0.09	0.08	0.11	0.08

Table S41. Mulliken charges for Low-Spin Models (a.u.).

	69		70		71		72	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Cu-Cu-Zn-Zn	0.10	0.10	0.14	0.11	0.45	0.41	0.45	0.45
Cu-Zn-Cu-Zn	0.07	0.07	0.48	0.48	0.10	0.07	0.43	0.45
Cu-Zn-Zn-Cu	0.16	0.14	0.52	0.51	0.46	0.43	0.10	0.07
Zn-Cu-Cu-Zn	0.44	0.48	0.15	0.05	0.08	0.02	0.42	0.45
Zn-Cu-Zn-Cu	0.48	0.53	0.21	0.10	0.46	0.47	0.09	0.08
Zn-Zn-Cu-Cu	0.45	0.52	0.51	0.49	0.13	0.07	0.06	0.04
Cu-Cu-Cu-Zn	0.10	0.03	0.17	0.06	0.09	0.02	0.43	0.47
Cu-Cu-Zn-Cu	0.13	0.14	0.18	0.09	0.45	0.47	0.11	0.08
Cu-Zn-Cu-Cu	0.10	0.08	0.49	0.52	0.10	0.08	0.09	0.09
Zn-Cu-Cu-Cu	0.44	0.50	0.17	0.08	0.08	0.09	0.08	0.04
Cu-Cu-Cu-Cu	0.08	0.10	0.15	0.09	0.09	0.03	0.05	0.01

Table S42. Mulliken charges for Medium-Spin Models (a.u.).

	69		70		71		72	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Cu-Cu-Cu-Cu	0.0886	0.1052	0.1447	0.1057	0.0827	0.1076	0.0821	0.0294

Table S43. Molecular areas and volumes of MT models.

	Molecular area ($Bohr^2$)						Molecular volume ($Bohr^3$)					
	RHS	UHS	RLS	ULS	RMS	UMS	RHS	UHS	RLS	ULS	RMS	UMS
Zn-Zn-Zn-Zn	2302	2166					6705	6514				
Cu-Zn-Zn-Zn	2325	2158					6723	6518				
Zn-Cu-Zn-Zn	2289	2164					6677	6577				
Zn-Zn-Cu-Zn	2315	2222					6686	6551				
Zn-Zn-Zn-Cu	2290	2213					6670	6568				
Cu-Cu-Zn-Zn	2292	2186	2263	2202			6642	6459	6560	6483		
Cu-Zn-Cu-Zn	2385	2218	2368	2257			6686	6553	6652	6631		
Cu-Zn-Zn-Cu	2317	2225	2321	2224			6712	6584	6717	6546		
Zn-Cu-Cu-Zn	2365	2326	2359	2321			6642	6590	6629	6510		
Zn-Cu-Zn-Cu	2308	2154	2312	2173			6699	6487	6688	6485		
Zn-Zn-Cu-Cu	2322	2195	2307	2204			6724	6482	6659	6510		
Cu-Cu-Cu-Zn	2379	2190	2380	2241			6652	6405	6647	6494		
Cu-Cu-Zn-Cu	2379	2150	2285	2135			6652	6470	6629	6411		
Cu-Zn-Cu-Cu	2376	2206	2370	2138			6680	6451	6677	6312		
Zn-Cu-Cu-Cu	2305	2186	2361	2162			6676	6493	6666	6376		
Cu-Cu-Cu-Cu	2371	2197	2185	2200	2197	2209	6645	6510	6496	6369	6505	6468

PART 4: MT3-beta

Table S44. Optimized M-S bond lengths of MT3-beta with restrained C-alpha; highest spin state (Å).

C6-Pos1	C8-Pos1	C8-Pos3	C14-Pos3	C16-Pos2	C16-Pos3	C20-Pos2	C22-Pos1	C25-Pos1	C25-Pos2	C27-Pos3	C30-Pos2	Metal 390-391-392	Energy	M_S
2.38	**2.47**	**2.45**	**2.38**	**2.41**	**2.44**	**2.36**	**2.37**	**2.49**	**2.47**	**2.37**	**2.40**	**Zn-Zn-Zn**	**-9692.239372**	**0**
2.32	2.46	2.43	2.38	2.41	2.45	2.37	2.33	2.41	2.45	2.37	2.40	Cu-Zn-Zn	-9551.212790	½
2.39	2.48	2.45	2.38	2.34	2.43	2.32	2.37	2.47	2.51	2.36	2.38	Zn-Cu-Zn	-9551.213963	
2.39	**2.42**	**2.41**	**2.33**	**2.42**	**2.45**	**2.36**	**2.36**	**2.46**	**2.45**	**2.36**	**2.40**	**Zn-Zn-Cu**	**-9551.218673**	
2.32	2.48	2.43	2.38	2.34	2.44	2.32	2.32	2.38	2.47	2.37	2.38	Cu-Cu-Zn	-9410.188345	1
2.30	2.40	2.38	2.33	2.42	2.44	2.37	2.34	2.39	2.45	2.38	2.39	Cu-Zn-Cu	-9410.194498	
2.39	**2.43**	**2.42**	**2.34**	**2.35**	**2.42**	**2.32**	**2.36**	**2.44**	**2.47**	**2.36**	**2.38**	**Zn-Cu-Cu**	**-9410.194740**	
2.31	**2.41**	**2.39**	**2.34**	**2.35**	**2.42**	**2.32**	**2.33**	**2.37**	**2.46**	**2.38**	**2.37**	**Cu-Cu-Cu**	**-9269.170869**	**3/2**

Table S45. Optimized M-S bond lengths of MT3-beta with unrestrained C-alpha; highest spin state (Å).

C6-Pos1	C8-Pos1	C8-Pos3	C14-Pos3	C16-Pos2	C16-Pos3	C20-Pos2	C22-Pos1	C25-Pos1	C25-Pos2	C27-Pos3	C30-Pos2	Metal 390-391-392	Energy	M_S
2.37	**2.42**	**2.42**	**2.36**	**2.45**	**2.43**	**2.35**	**2.33**	**2.43**	**2.42**	**2.38**	**2.37**	**Zn-Zn-Zn**	**-9692.267116**	**0**
2.32	2.39	2.41	2.36	2.45	2.43	2.35	2.31	2.37	2.40	2.38	2.35	Cu-Zn-Zn	-9551.242518	½
2.37	2.42	2.42	2.36	2.36	2.41	2.33	2.36	2.44	2.38	2.33	2.33	Zn-Cu-Zn	-9551.240620	
2.37	**2.42**	**2.40**	**2.34**	**2.43**	**2.36**	**2.36**	**2.35**	**2.41**	**2.42**	**2.32**	**2.35**	**Zn-Zn-Cu**	**-9551.246547**	
2.33	**2.40**	**2.40**	**2.36**	**2.37**	**2.41**	**2.31**	**2.31**	**2.36**	**2.36**	**2.37**	**2.33**	**Cu-Cu-Zn**	**-9410.220946**	**1**
2.33	2.39	2.37	2.34	2.42	2.37	2.38	2.31	2.36	2.42	2.32	2.36	Cu-Zn-Cu	-9410.220061	
2.38	2.43	2.40	2.34	2.36	2.37	2.31	2.36	2.42	2.40	2.31	2.33	Zn-Cu-Cu	-9410.220624	
2.34	**2.37**	**2.39**	**2.34**	**2.37**	**2.36**	**2.32**	**2.31**	**2.36**	**2.38**	**2.31**	**2.33**	**Cu-Cu-Cu**	**-9269.198050**	**3/2**

Table S46. Optimized M-S bond lengths of MT3-beta with restrained C-alpha; lowest spin state (Å).

C6-Pos1	C8-Pos1	C8-Pos3	C14-Pos3	C16-Pos2	C16-Pos3	C20-Pos2	C22-Pos1	C25-Pos1	C25-Pos2	C27-Pos3	C30-Pos2	Metal 390-391-392	Energy	M_S
2.30	2.45	2.44	2.39	2.32	2.44	2.31	2.32	2.38	2.57	2.37	2.39	Cu-Cu-Zn	-9410.174378	0
2.28	2.40	2.42	2.35	2.44	2.43	2.42	2.37	2.35	2.40	2.44	2.39	Cu-Zn-Cu	-9410.178204	
2.40	**2.44**	**2.41**	**2.33**	**2.36**	**2.36**	**2.31**	**2.36**	**2.43**	**2.48**	**2.35**	**2.35**	**Zn-Cu-Cu**	**-9410.191207**	
2.34	**2.38**	**2.39**	**2.34**	**2.38**	**2.33**	**2.32**	**2.31**	**2.36**	**2.37**	**2.31**	**2.32**	**Cu-Cu-Cu**	**-9269.197511**	½

Table S47. Optimized M-S bond lengths of MT3-beta with unrestrained C-alpha; lowest spin state (Å).

C6-Pos1	C8-Pos1	C8-Pos3	C14-Pos3	C16-Pos2	C16-Pos3	C20-Pos2	C22-Pos1	C25-Pos1	C25-Pos2	C27-Pos3	C30-Pos2	Metal 390-391-392	Energy	M_S
2.34	2.37	2.41	2.38	2.35	2.45	2.30	2.30	2.43	2.41	2.35	2.35	Cu-Cu-Zn	-9410.213690	0
2.32	2.33	2.41	2.35	2.48	2.36	2.37	2.32	2.41	2.40	2.30	2.36	Cu-Zn-Cu	-9410.218518	
2.38	**2.43**	**2.37**	**2.32**	**2.37**	**2.31**	**2.32**	**2.35**	**2.43**	**2.36**	**2.31**	**2.30**	**Zn-Cu-Cu**	**-9410.222395**	
2.34	**2.37**	**2.38**	**2.34**	**2.38**	**2.34**	**2.32**	**2.31**	**2.37**	**2.37**	**2.31**	**2.32**	**Cu-Cu-Cu**	**-9269.198102**	½

Table S48. Optimized angles of MT3-beta with restrained C-alpha; highest spin state (degrees).

C6-P1-C8	C6-P1-C22	C6-P1-C25	C8-P1-C22	C8-P1-C25	C22-P1-C25	C8-P3-C14	C8-P3-C16	C8-P3-C27	C14-P3-C16	C14-P3-C27	C16-P3-C27	C16-P2-C20	C16-P2-C25	C16-P2-C30	C20-P2-C25	C20-P2-C30	C25-P2-C30	Metal
109.2	**103.0**	**111.2**	**110.2**	**112.0**	**110.8**	**124.9**	**90.8**	**97.5**	**101.5**	**110.5**	**133.3**	**119.2**	**94.2**	**107.1**	**108.4**	**104.1**	**124.8**	**Zn-Zn-Zn**
97.0	94.3	129.5	124.7	111.7	101.2	128.0	91.9	96.9	99.7	109.1	134.4	119.9	93.7	107.8	108.5	103.3	124.9	Cu-Zn-Zn
108.7	104.0	113.3	108.4	112.5	109.4	125.1	92.6	97.8	101.7	110.4	131.2	130.0	91.9	101.6	105.9	100.4	131.3	Zn-Cu-Zn
110.6	**104.2**	**109.8**	**115.3**	**105.4**	**111.5**	**135.9**	**90.2**	**99.1**	**97.7**	**99.0**	**144.4**	**119.6**	**98.2**	**108.7**	**106.6**	**103.0**	**122.0**	**Zn-Zn-Cu**
97.5	95.1	132.1	122.8	111.5	99.5	128.4	93.3	97.0	100.1	108.9	132.4	128.8	91.7	102.0	105.5	100.6	132.5	Cu-Cu-Zn
96.0	97.8	128.4	130.9	105.2	102.2	136.5	89.9	97.7	98.3	100.2	143.2	119.7	97.1	108.9	106.5	102.6	123.4	Cu-Zn-Cu
110.3	**104.4**	**111.7**	**113.0**	**107.2**	**110.2**	**134.2**	**91.4**	**99.6**	**97.8**	**99.7**	**142.3**	**129.0**	**95.6**	**103.4**	**103.3**	**99.6**	**129.8**	**Zn-Cu-Cu**
96.7	**98.0**	**130.9**	**129.5**	**105.5**	**100.3**	**135.8**	**91.0**	**98.4**	**98.1**	**100.4**	**141.9**	**127.2**	**95.0**	**103.2**	**103.2**	**100.1**	**132.1**	**Cu-Cu-Cu**

Table S49. Optimized angles of MT3-beta with unrestrained C-alpha; highest spin state (degrees).

C6-P1-C8	C6-P1-C22	C6-P1-C25	C8-P1-C22	C8-P1-C25	C22-P1-C25	C8-P3-C14	C8-P3-C16	C8-P3-C27	C14-P3-C16	C14-P3-C27	C16-P3-C27	C16-P2-C20	C16-P2-C25	C16-P2-C30	C20-P2-C25	C20-P2-C30	C25-P2-C30	Metal
110.7	**109.0**	**107.0**	**115.0**	**104.5**	**109.8**	**106.7**	**115.1**	**107.6**	**107.4**	**113.4**	**106.9**	**111.8**	**113.6**	**105.7**	**107.3**	**111.1**	**107.2**	**Zn-Zn-Zn**
94.0	98.9	135.0	141.7	95.4	100.4	109.4	113.5	106.2	105.3	110.3	112.2	117.0	102.0	106.7	109.8	107.6	113.9	Cu-Zn-Zn
112.6	110.0	110.4	110.2	107.9	105.4	103.1	97.1	120.7	113.1	108.4	113.8	135.0	96.6	102.7	98.8	94.4	136.6	Zn-Cu-Zn
109.6	**108.8**	**105.5**	**114.2**	**106.9**	**111.5**	**132.9**	**95.3**	**99.2**	**95.5**	**98.8**	**143.3**	**112.5**	**105.0**	**108.1**	**111.4**	**110.6**	**109.3**	**Zn-Zn-Cu**
92.7	**98.4**	**135.1**	**144.2**	**95.7**	**99.9**	**108.8**	**111.1**	**106.9**	**107.2**	**110.4**	**112.3**	**137.4**	**93.6**	**101.6**	**98.3**	**98.5**	**134.6**	**Cu-Cu-Zn**
91.8	97.4	136.1	142.5	94.1	103.6	133.0	94.2	99.8	96.3	98.4	143.4	111.2	101.5	110.4	112.0	107.7	114.1	Cu-Zn-Cu
108.9	107.5	109.4	115.8	107.1	108.1	136.4	87.2	96.5	100.1	99.4	147.1	139.9	93.2	100.9	98.2	97.2	135.9	Zn-Cu-Cu
97.0	**99.2**	**129.7**	**141.1**	**93.4**	**102.7**	**130.4**	**89.2**	**97.5**	**99.8**	**100.7**	**146.3**	**143.7**	**89.0**	**101.1**	**96.3**	**96.8**	**141.5**	**Cu-Cu-Cu**

Table S50. Optimized angles of MT3-beta with restrained C-alpha; lowest spin state (degrees).

C6-P1-C8	C6-P1-C22	C6-P1-C25	C8-P1-C22	C8-P1-C25	C22-P1-C25	C8-P3-C14	C8-P3-C16	C8-P3-C27	C14-P3-C16	C14-P3-C27	C16-P3-C27	C16-P2-C20	C16-P2-C25	C16-P2-C30	C20-P2-C25	C20-P2-C30	C25-P2-C30	Metal
98.4	99.0	135.7	125.7	108.2	93.6	128.8	93.5	96.4	99.6	108.3	133.7	131.6	88.1	103.5	99.0	103.0	137.1	Cu-Cu-Zn
93.7	100.4	130.0	133.6	100.9	103.1	140.5	89.2	95.1	98.5	99.1	146.3	117.9	98.8	108.9	106.3	102.8	123.1	Cu-Zn-Cu
109.7	**104.0**	**112.5**	**112.0**	**107.7**	**111.0**	**133.7**	**94.8**	**100.0**	**96.9**	**100.0**	**138.8**	**132.6**	**93.7**	**97.2**	**104.4**	**103.7**	**129.4**	**Zn-Cu-Cu**
96.7	**99.0**	**130.0**	**139.7**	**96.1**	**101.5**	**129.8**	**89.8**	**98.8**	**99.9**	**100.9**	**144.1**	**148.7**	**87.3**	**97.2**	**97.2**	**97.8**	**142.2**	**Cu-Cu-Cu**

Table S51. Optimized angles of MT3-beta with unrestrained C-alpha; lowest spin state (degrees).

C6-P1-C8	C6-P1-C22	C6-P1-C25	C8-P1-C22	C8-P1-C25	C22-P1-C25	C8-P3-C14	C8-P3-C16	C8-P3-C27	C14-P3-C16	C14-P3-C27	C16-P3-C27	C16-P2-C20	C16-P2-C25	C16-P2-C30	C20-P2-C25	C20-P2-C30	C25-P2-C30	Metal
102.0	101.2	132.4	136.5	98.8	91.8	117.2	93.3	107.3	105.9	111.7	120.8	157.2	80.6	97.5	89.9	95.9	167.9	Cu-Cu-Zn
91.1	104.0	128.3	141.6	90.5	106.3	168.0	84.6	89.3	93.4	93.6	172.2	113.8	103.4	105.3	110.8	107.8	115.7	Cu-Zn-Cu
108.7	**107.8**	**109.5**	**114.1**	**108.5**	**108.2**	**136.3**	**90.3**	**99.0**	**97.3**	**98.9**	**144.9**	**139.8**	**93.0**	**94.2**	**102.9**	**100.5**	**133.5**	**Zn-Cu-Cu**
96.5	**98.5**	**129.2**	**139.4**	**96.4**	**102.6**	**131.3**	**89.1**	**97.9**	**100.1**	**100.6**	**145.1**	**147.0**	**87.5**	**98.2**	**98.1**	**97.4**	**141.2**	**Cu-Cu-Cu**

Table S52. Mulliken charges for High-Spin Models (a.u.).

	1		2		3	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Zn-Zn-Zn	0.45	0.47	0.44	0.44	0.47	0.46
Cu-Zn-Zn	0.12	0.09	0.44	0.43	0.47	0.47
Zn-Cu-Zn	0.45	0.47	0.14	0.09	0.47	0.44
Zn-Zn-Cu	0.45	0.45	0.44	0.46	0.15	0.08
Cu-Cu-Zn	0.11	0.07	0.13	0.09	0.47	0.47
Cu-Zn-Cu	0.10	0.08	0.44	0.49	0.15	0.09
Zn-Cu-Cu	0.44	0.45	0.13	0.10	0.15	0.09
Cu-Cu-Cu	0.10	0.09	0.13	0.08	0.15	0.09

Table S53. Mulliken charges for Low-Spin Models (a.u.).

	1		2		3	
	Restrained	Unrestrained	Restrained	Unrestrained	Restrained	Unrestrained
Cu-Cu-Zn	0.11	0.08	0.13	0.07	0.47	0.50
Cu-Zn-Cu	0.11	0.07	0.44	0.43	0.15	0.02
Zn-Cu-Cu	0.45	0.45	0.12	0.06	0.12	0.08
Cu-Cu-Cu	0.09	0.09	0.08	0.08	0.08	0.08

Table S54. Molecular areas and volumes of MT models.

	Molecular area ($Bohr^2$)				Molecular volume ($Bohr^3$)			
	RHS	UHS	RLS	ULS	RHS	UHS	RLS	ULS
Zn-Zn-Zn	1923	1825			5379	5377		
Cu-Zn-Zn	1943	1925			5381	5359		
Zn-Cu-Zn	1919	1872			5371	5354		
Zn-Zn-Cu	1923	1950			5387	5343		
Cu-Cu-Zn	1928	1918	1921	1948	5338	5273	5338	5274
Cu-Zn-Cu	1957	1926	1939	1915	5388	5279	5365	5297
Zn-Cu-Cu	1933	1930	1915	1923	5405	5365	5357	5309
Cu-Cu-Cu	1949	1927	1919	1949	5364	5262	5235	5305

PART 5: Comparative data

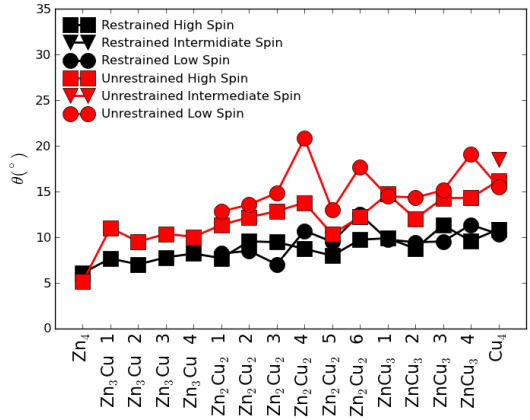
Table S55. Comparing MT2α/MT3α and MT2β/MT3β computed average metal-sulfur bond lengths (Å).

Cluster α	Formula	No.	M_S	Zn-S^t_{avg}	Cu-S^t_{avg}	Zn-S^b_{avg}	Cu-S^b_{avg}	Position[1] 63^t-65^b-64^b62^t
	Zn_4	1	0	2.35 / 2.35	- / -	2.42 / 2.41	- / -	Zn-Zn-Zn-Zn
	Zn_3Cu	2	½	2.35 / 2.35	2.33 / 2.33	2.41 / 2.41	2.38 / 2.37	Cu-Zn-Zn-Zn
		3	½	2.36 / 2.36	2.31 / 2.31	2.42 / 2.42	2.34 / 2.36	Zn-Cu-Zn-Zn
		4	½	2.36 / 2.36	2.31 / 2.31	2.43 / 2.42	2.36 / 2.37	Zn-Zn-Cu-Zn
		5	½	2.36 / 2.35	2.31 / 2.31	2.41 / 2.41	2.38 / 2.39	Zn-Zn-Zn-Cu
	Zn_2Cu_2	6	0 / 0	2.35 / 2.36	2.31 / 2.34	2.41 / 2.41	2.35 / 2.39	Cu-Cu-Zn-Zn
		7	0 / 1*	2.36 / 2.35	2.31 / 2.31	2.41 / 2.41	2.37/ 2.37	Cu-Zn-Cu-Zn
		8	1 / 0	2.36 / 2.36	2.32 / 2.32	2.40 / 2.41	2.39 / 2.39	Cu-Zn-Zn-Cu
		9	0 / -	2.36 / -	2.32 / -	2.43 / -	2.36 / -	Zn-Cu-Cu-Zn
		10	0 / 1	2.36 / 2.36	2.30/ 2.30	2.41 / 2.41	2.34 / 2.35	Zn-Cu-Zn-Cu
		11	0 / 0	2.35 / 2.36	2.32 / 2.32	2.43 / 2.42	2.42 / 2.42	Zn-Zn-Cu-Cu
	$ZnCu_3$	12	0 / -	2.37 / -	2.31 / -	2.41 / -	2.35 / -	Cu-Cu-Cu-Zn
		13	½ / ½	2.37 / 2.35	2.31 / 2.31	2.40 / 2.40	2.35 / 2.35	Cu-Cu-Zn-Cu
		14	½ / ½*	2.36 / 2.35	2.31 / 2.32	2.41 / 2.41	2.37 / 2.37	Cu-Zn-Cu-Cu
		15	½ /3/2*	2.37 / 2.37	2.32 / 2.32	2.42 / 2.45	2.35 / 2.35	Zn-Cu-Cu-Cu
	Cu_4	16	1 / 2*	- / -	2.31 / 2.32	- / -	2.35 / 2.35	Cu-Cu-Cu-Cu
Cluster β	Formula	No.	M_S	Zn-S^t_{avg}	Cu-S^t_{avg}	Zn-S^b_{avg}	Cu-S^b_{avg}	Position 66^t-67^t-68^t
	Zn_3		0	2.36 / 2.36	- / -	2.43 / 2.43	- / -	Zn-Zn-Zn
	Zn_2Cu	1	½	2.36 / 2.36	2.33 / 2.32	2.42 / 2.42	2.38 / 2.38	Cu-Zn-Zn
		2	½	2.37 / 2.36	2.33 / 2.33	2.42 / 2.42	2.37 / 2.37	Zn-Cu-Zn
		3	½	2.36 / 2.36	2.32 / 2.33	2.42 / 2.42	2.39 / 2.38	Zn-Zn-Cu
	$ZnCu_2$	4	0 / 1	2.36 / 2.37	2.31 / 2.32	2.44 / 2.41	2.38 / 2.37	Cu-Cu-Zn
		5	0 / 1	2.36 / 2.37	2.31 / 2.33	2.42 / 2.42	2.40 / 2.37	Cu-Zn-Cu
		6	0	2.36 / 2.37	2.31 / 2.31	2.44 / 2.43	2.36 / 2.35	Zn-Cu-Cu
	Cu_3		½	- / -	2.32 / 2.32	- / -	2.37 / 2.37	Cu-Cu-Cu

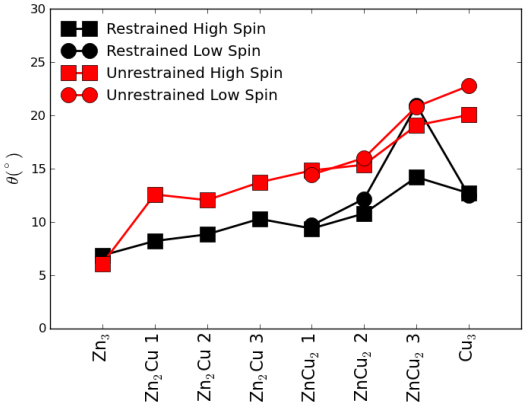
[1] t and b refers to metal sites being in "terminal" and "bridging" positions where terminal denotes metal ions have two terminal sulfur atoms while in the bridge the metal ions are only coordinated by one terminal sulfur.

Figure S1. Angular distortion in computed metallothionein clusters (degrees).

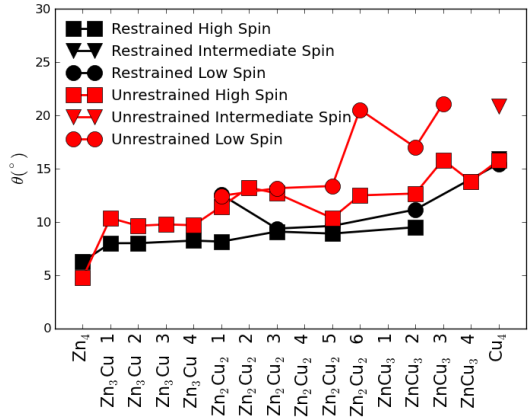
MT2 a



MT2 b



MT3 a



MT3b

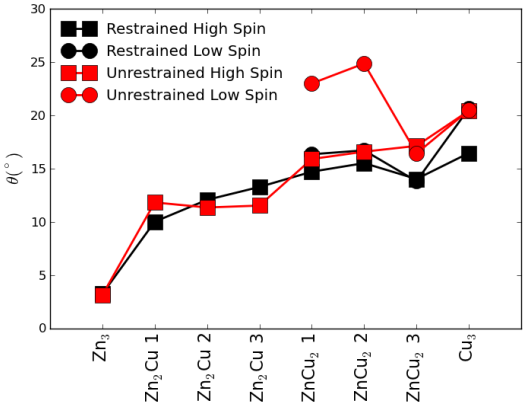


Figure S2. Volumes of α-domain clusters.

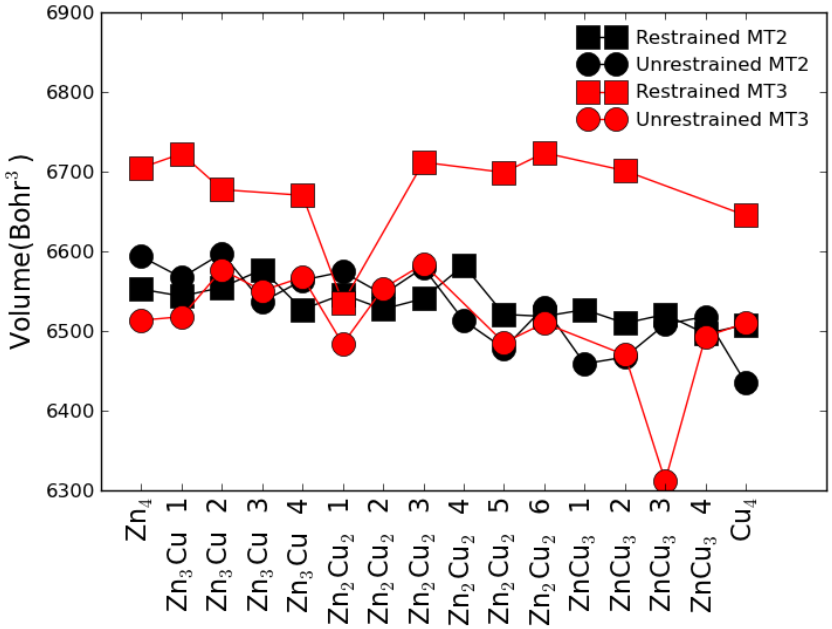


Figure S3. Volumes of β-domain clusters.

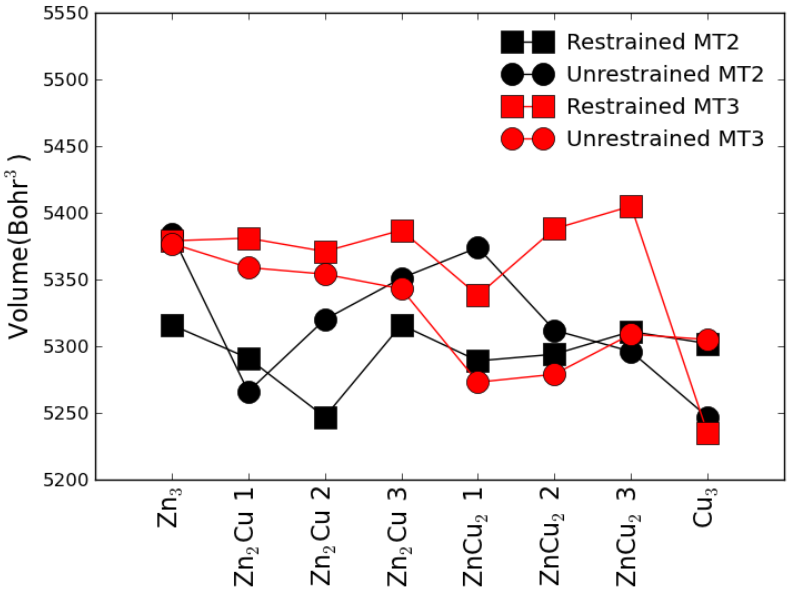


Figure S4. Bond lengths (Å). MT2α: terminal S^t (left) and bridging S^b bonds (right).

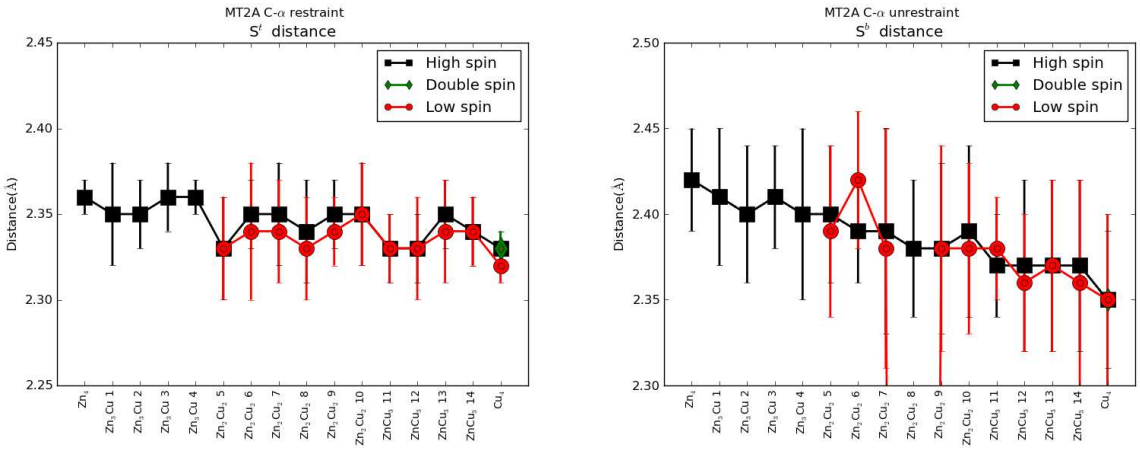


Figure S5. Bond lengths (Å). MT2β: terminal S^t (left) and bridging S^b bonds (right).

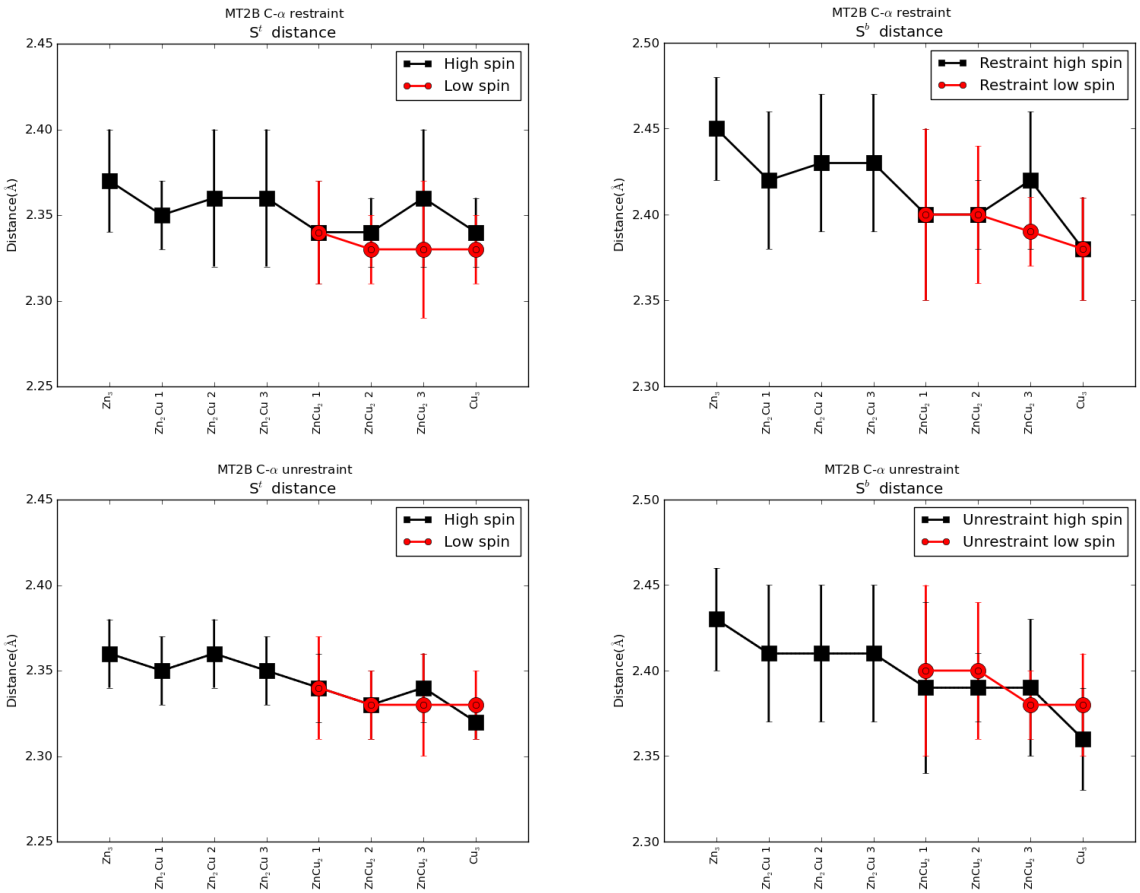


Figure S6. Bond lengths (Å). MT3α: terminal S^t (left) and bridging S^b bonds (right).

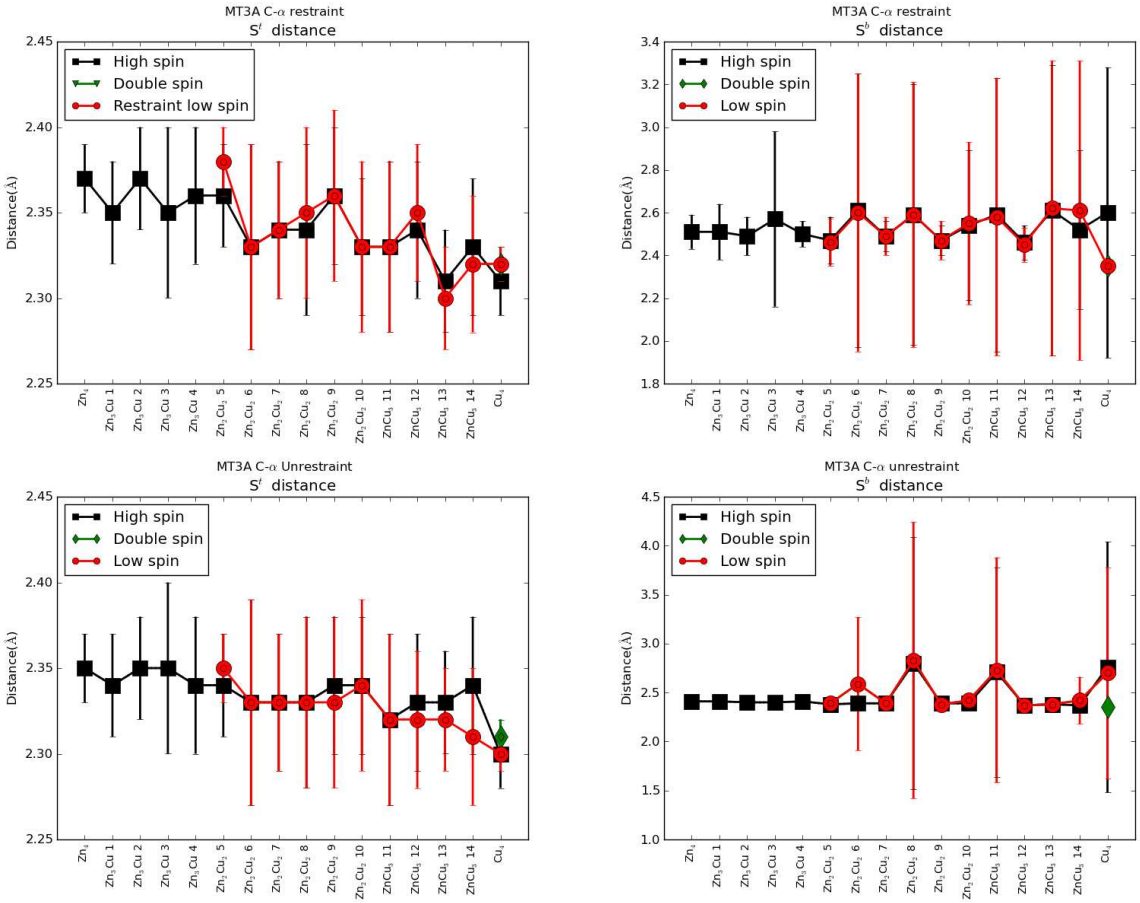


Figure S7. Bond lengths (Å). MT3β: terminal S^t (left) and bridging S^b bonds (right).

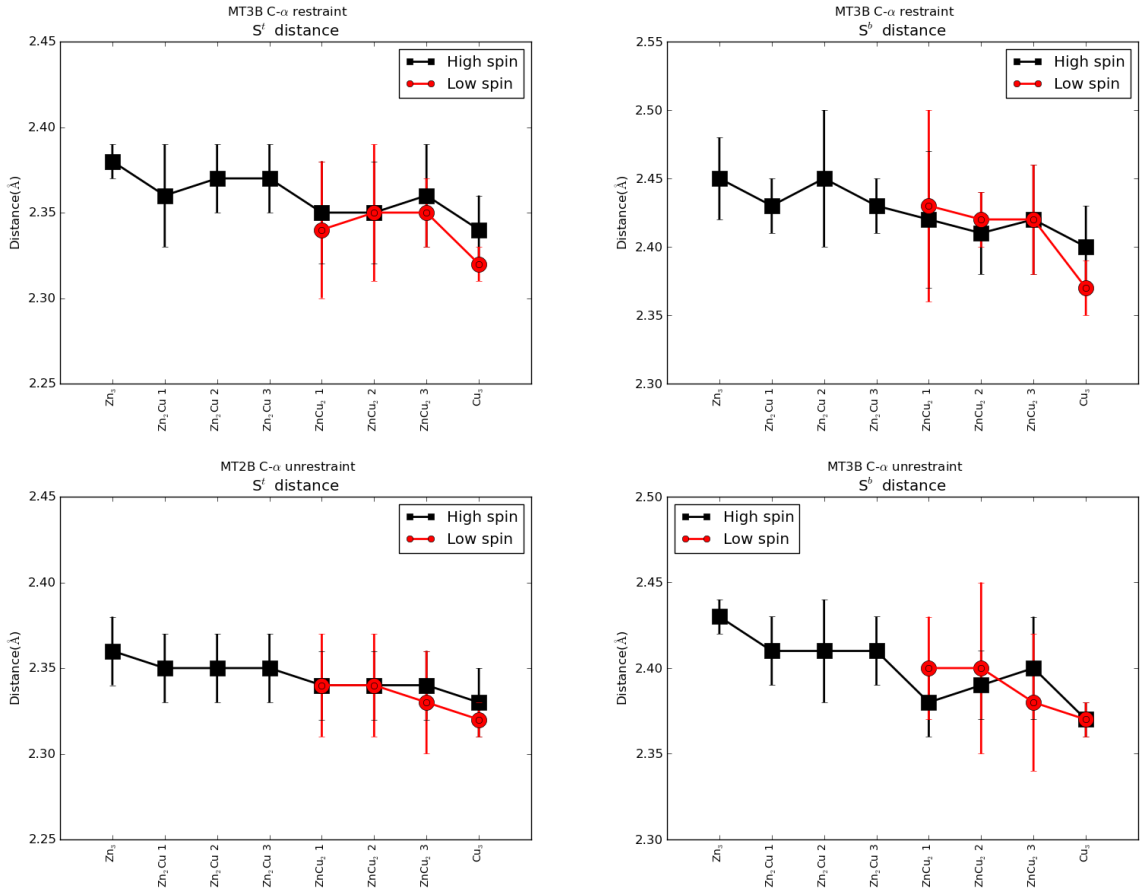
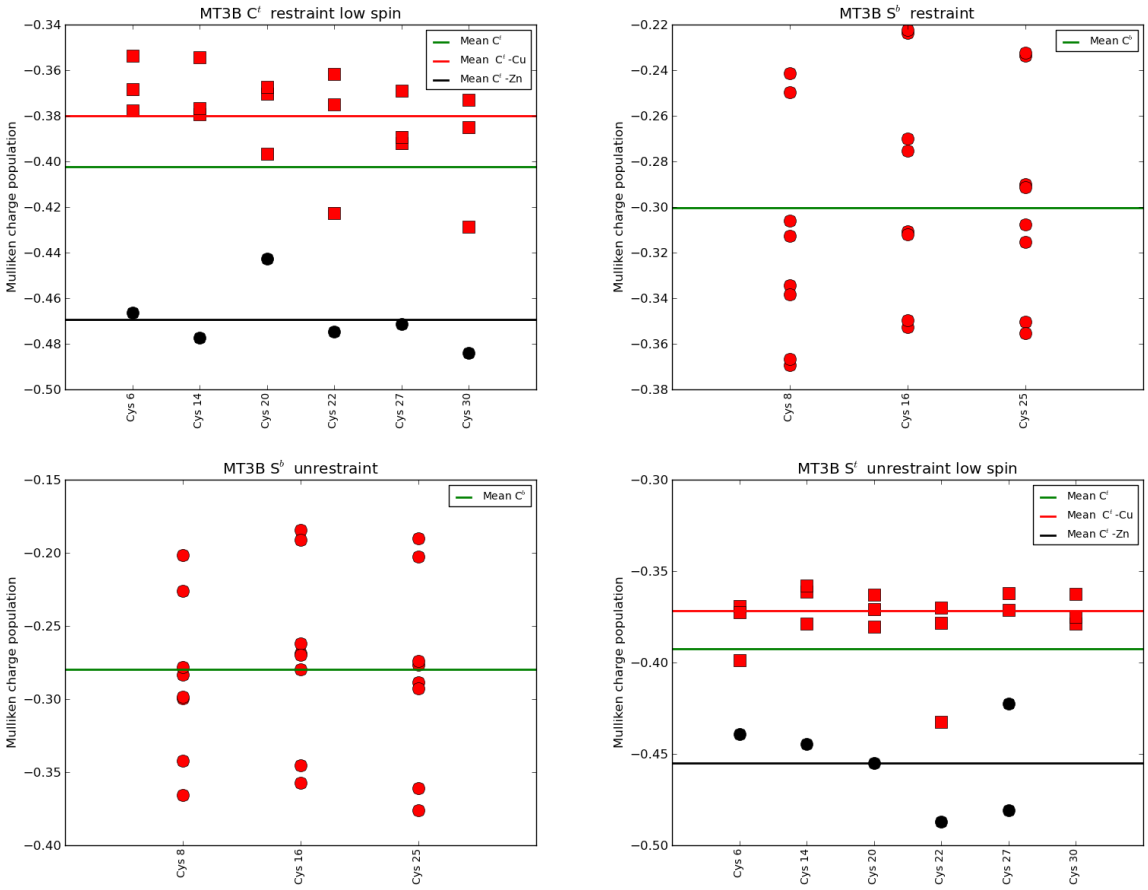
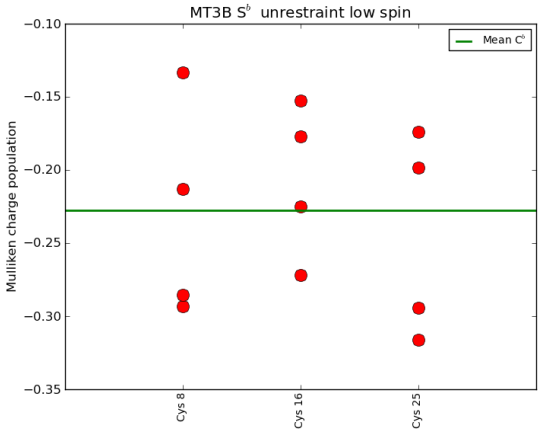
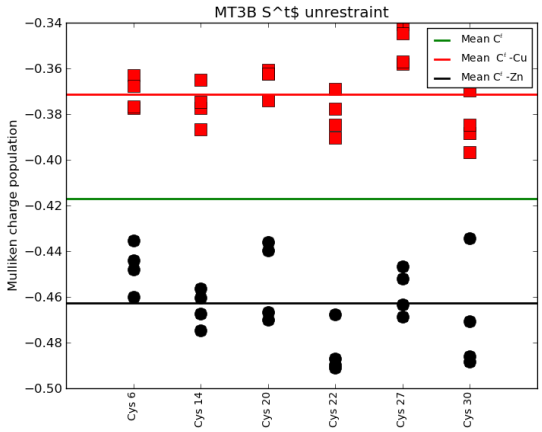
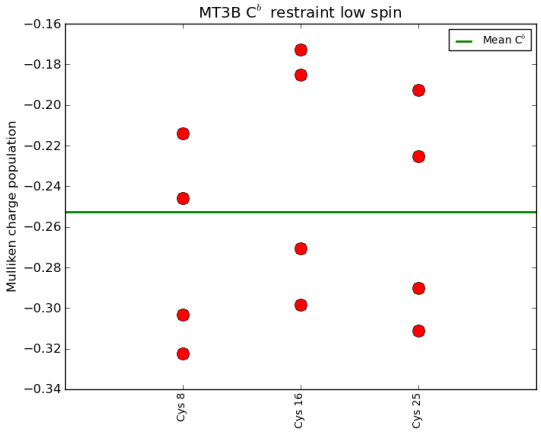


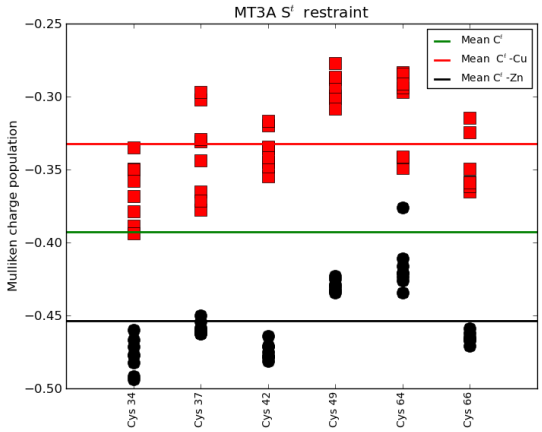
Figure S8. Mulliken charges.

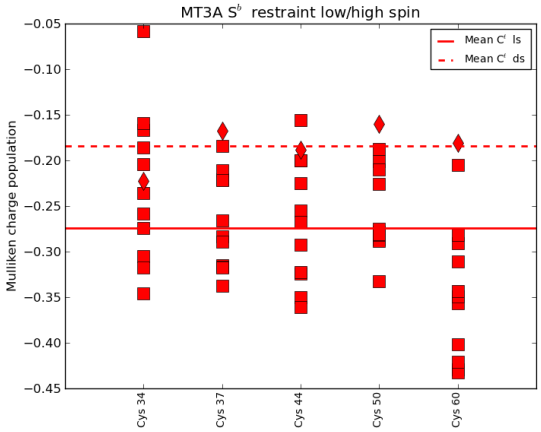



MT3B S^b unrestraint low spin
Mean C^b
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
Cys 8
Cys 16
Cys 25

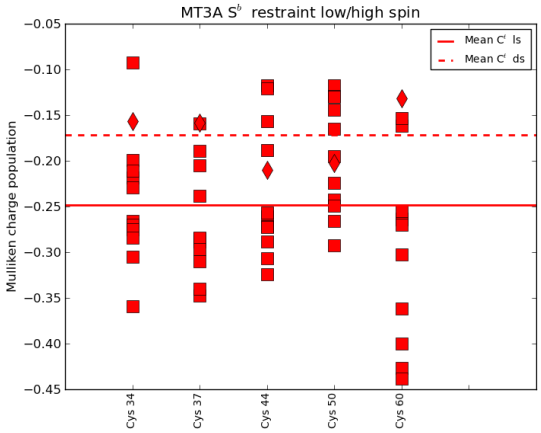

MT3B S^t$ unrestraint
Mean C^t
Mean C^t-Cu
Mean C^t-Zn
Mulliken charge population
−0.34
−0.36
−0.38
−0.40
−0.42
−0.44
−0.46
−0.48
−0.50
Cys 6
Cys 14
Cys 20
Cys 22
Cys 27
Cys 30

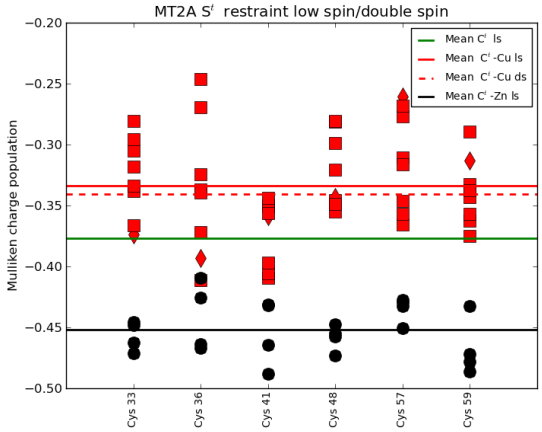

MT3B C^b restraint low spin
Mean C^b
Mulliken charge population
−0.16
−0.18
−0.20
−0.22
−0.24
−0.26
−0.28
−0.30
−0.32
−0.34
Cys 8
Cys 16
Cys 25

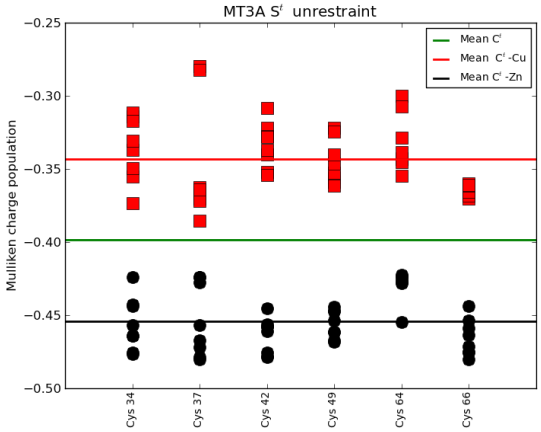

MT3A S^t restraint
Mean C^t
Mean C^t-Cu
Mean C^t-Zn
Mulliken charge population
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 34
Cys 37
Cys 42
Cys 49
Cys 64
Cys 66

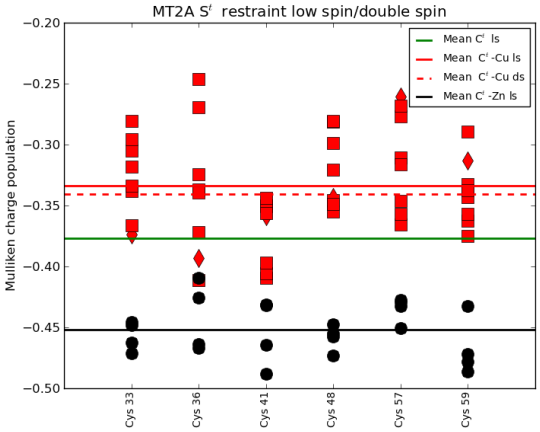

MT3A S^b restraint low/high spin
Mean C^t ls
Mean C^t ds
Mulliken charge population
−0.05
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60

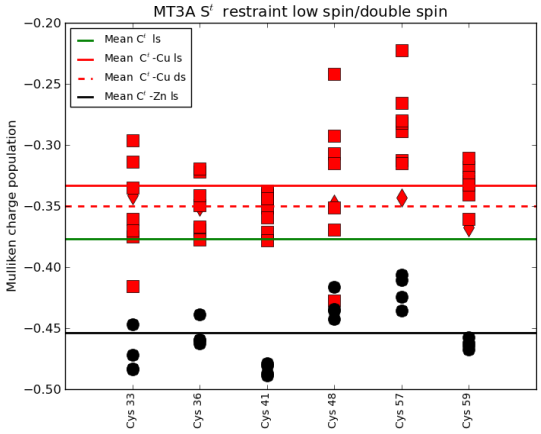

MT3A S^b restraint
Mean C^t
Mulliken charge population
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
Cys 35
Cys 38
Cys 45
Cys 51
Cys 67

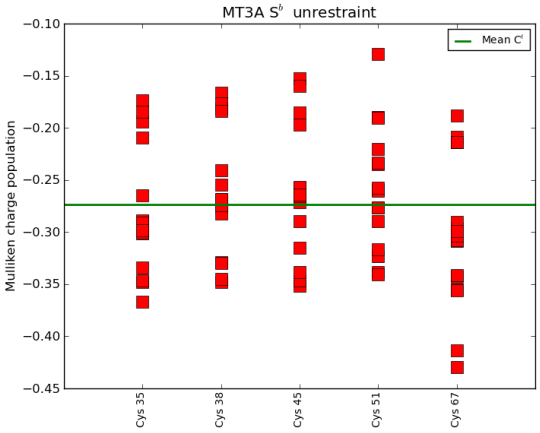

MT3A S^b restraint low/high spin
Mean C^t ls
Mean C^t ds
Mulliken charge population
−0.05
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60

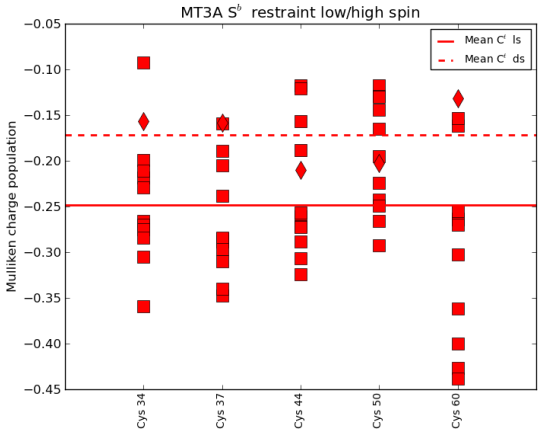

MT2A S^t restraint low spin/double spin
Mean C^t ls
Mean C^t-Cu ls
Mean C^t-Cu ds
Mean C^t-Zn ls
Mulliken charge population
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59

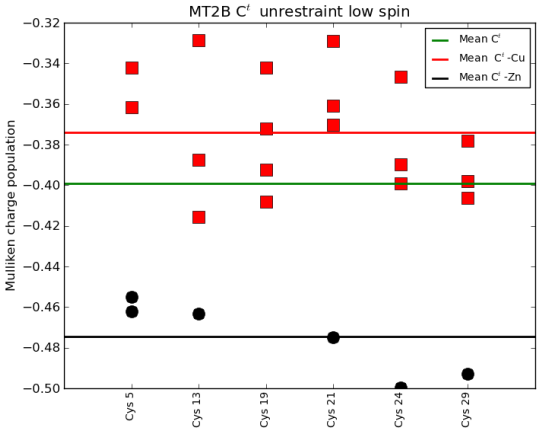

MT3A S^{t} unrestraint
Mean C^{t}
Mean C^{t} -Cu
Mean C^{t} -Zn
Mulliken charge population
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 34
Cys 37
Cys 42
Cys 49
Cys 64
Cys 66

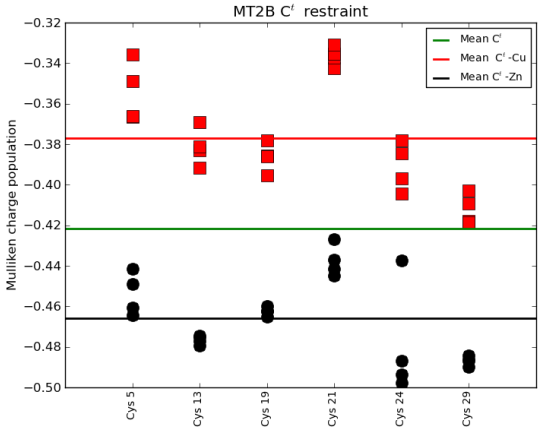

MT2A S^{t} restraint low spin/double spin
Mean C^{t} ls
Mean C^{t} -Cu ls
Mean C^{t} -Cu ds
Mean C^{t} -Zn ls
Mulliken charge population
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59

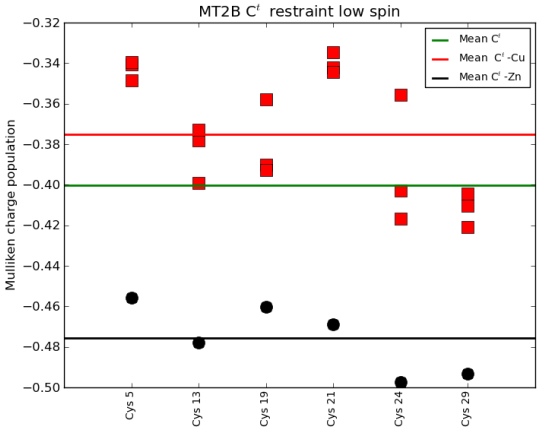

MT3A S^{t} restraint low spin/double spin
Mean C^{t} ls
Mean C^{t} -Cu ls
Mean C^{t} -Cu ds
Mean C^{t} -Zn ls
Mulliken charge population
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59

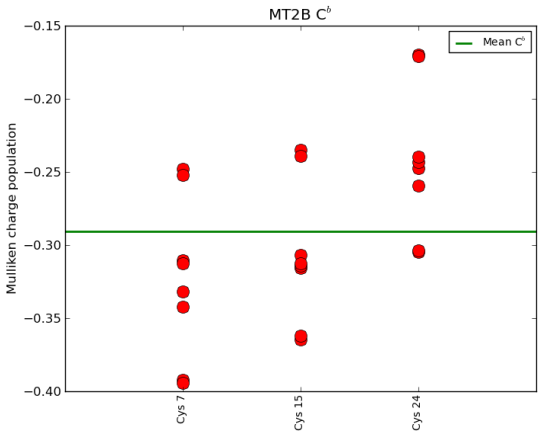

MT3A S^{b} unrestraint
Mean C^{t}
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
Cys 35
Cys 38
Cys 45
Cys 51
Cys 67

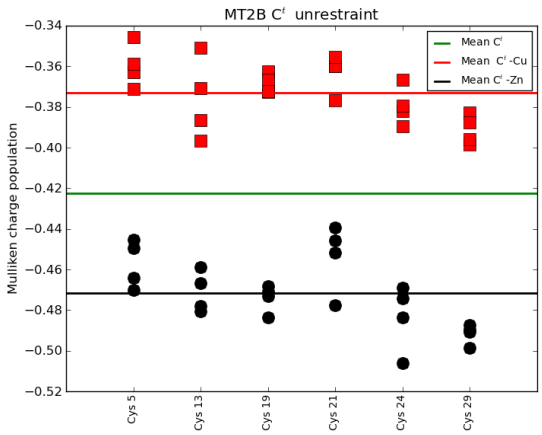

MT3A S^{b} restraint low/high spin
Mean C^{t} ls
Mean C^{t} ds
Mulliken charge population
−0.05
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
−0.45
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60

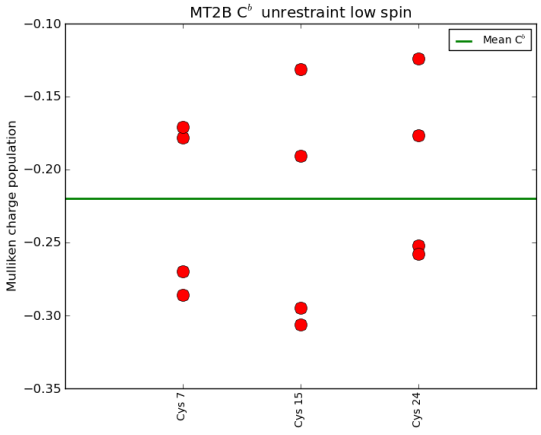

MT2B C^{t} unrestraint low spin
Mean C^{t}
Mean C^{t} -Cu
Mean C^{t} -Zn
Mulliken charge population
−0.32
−0.34
−0.36
−0.38
−0.40
−0.42
−0.44
−0.46
−0.48
−0.50
Cys 5
Cys 13
Cys 19
Cys 21
Cys 24
Cys 29

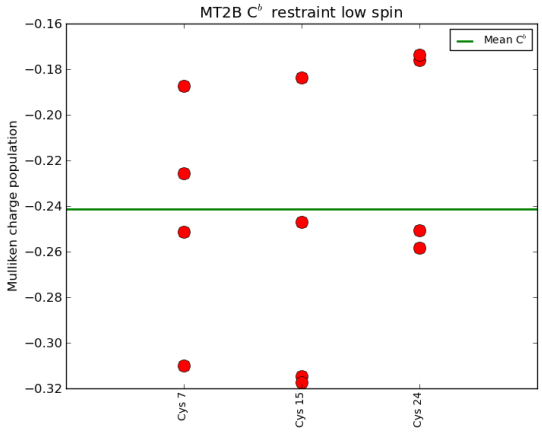

MT2B C^{t} restraint
Mean C^{t}
Mean C^{t} -Cu
Mean C^{t} -Zn
Mulliken charge population
−0.32
−0.34
−0.36
−0.38
−0.40
−0.42
−0.44
−0.46
−0.48
−0.50
Cys 5
Cys 13
Cys 19
Cys 21
Cys 24
Cys 29

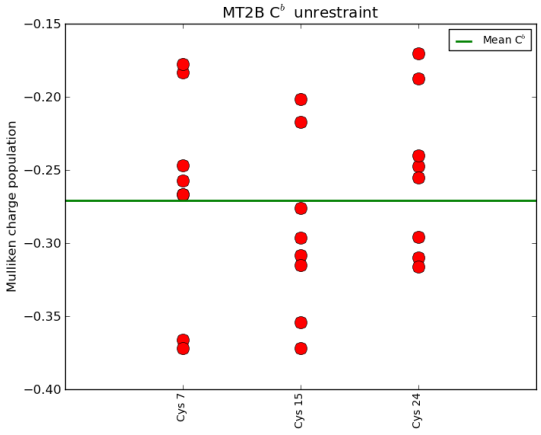

MT2B C^{t} restraint low spin
Mean C^{t}
Mean C^{t} -Cu
Mean C^{t} -Zn
Mulliken charge population
−0.32
−0.34
−0.36
−0.38
−0.40
−0.42
−0.44
−0.46
−0.48
−0.50
Cys 5
Cys 13
Cys 19
Cys 21
Cys 24
Cys 29

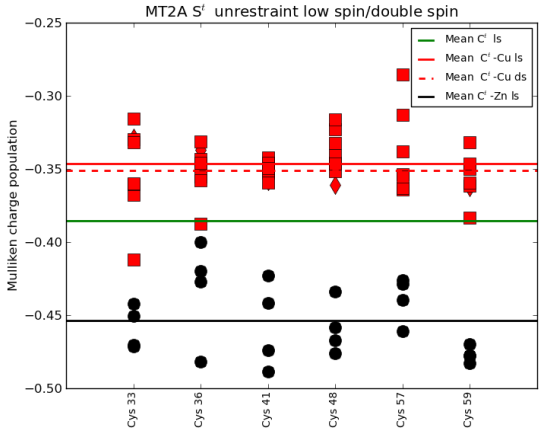

MT2B C^b
Mean C^b
Mulliken charge population
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
Cys 7
Cys 15
Cys 24

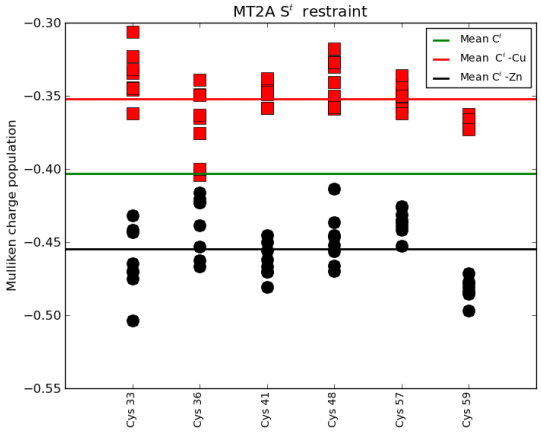

MT2B C^t unrestraint
Mean C^t
Mean C^t-Cu
Mean C^t-Zn
Mulliken charge population
−0.34
−0.36
−0.38
−0.40
−0.42
−0.44
−0.46
−0.48
−0.50
−0.52
Cys 5
Cys 13
Cys 19
Cys 21
Cys 24
Cys 29

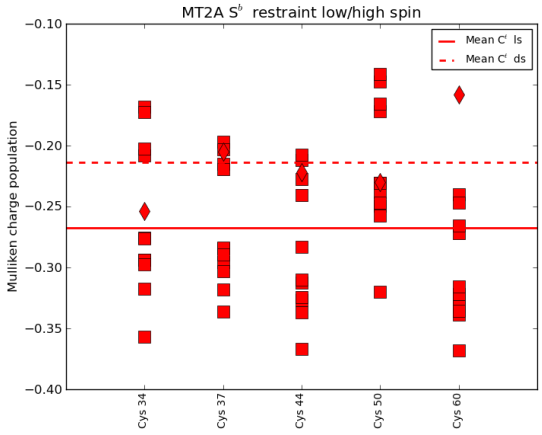

MT2B C^b unrestraint low spin
Mean C^b
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
Cys 7
Cys 15
Cys 24

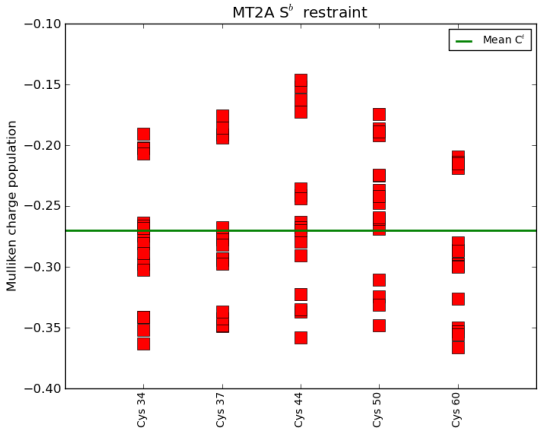

MT2B C^b restraint low spin
Mean C^b
Mulliken charge population
−0.16
−0.18
−0.20
−0.22
−0.24
−0.26
−0.28
−0.30
−0.32
Cys 7
Cys 15
Cys 24

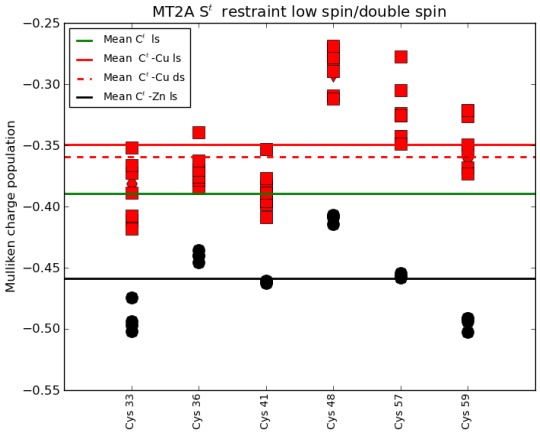

MT2B C^b unrestraint
Mean C^b
Mulliken charge population
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
Cys 7
Cys 15
Cys 24

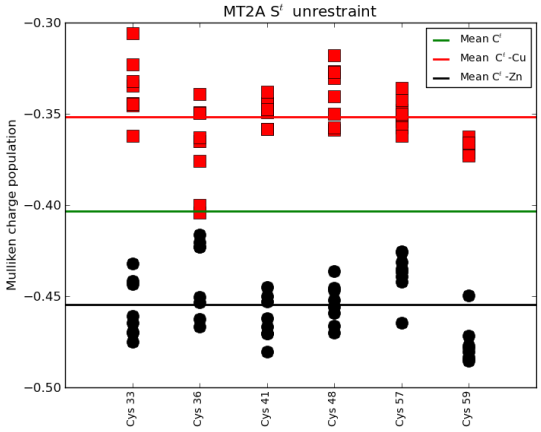

MT2A S^t unrestraint low spin/double spin
Mean C^t ls
Mean C^t-Cu ls
Mean C^t-Cu ds
Mean C^t-Zn ls
Mulliken charge population
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59

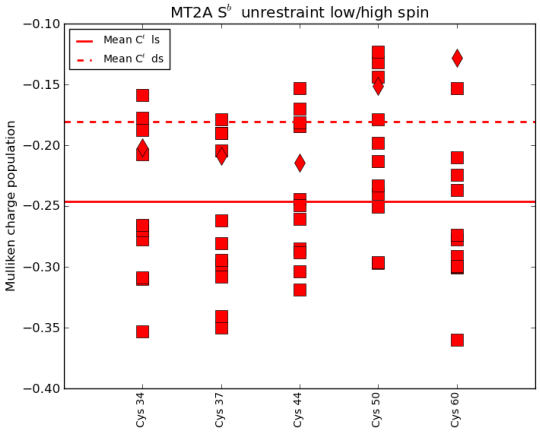

MT2A S^t restraint
Mean C^t
Mean C^t-Cu
Mean C^t-Zn
Mulliken charge population
−0.30
−0.35
−0.40
−0.45
−0.50
−0.55
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59


MT2A S^b restraint low/high spin
Mean C^t ls
Mean C^t ds
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60


MT2A S^b restraint
Mean C^l
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60


MT2A S^l restraint low spin/double spin
Mean C^l ls
Mean C^l -Cu ls
Mean C^l -Cu ds
Mean C^l -Zn ls
Mulliken charge population
−0.25
−0.30
−0.35
−0.40
−0.45
−0.50
−0.55
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59


MT2A S^l unrestraint
Mean C^l
Mean C^l -Cu
Mean C^l -Zn
Mulliken charge population
−0.30
−0.35
−0.40
−0.45
−0.50
Cys 33
Cys 36
Cys 41
Cys 48
Cys 57
Cys 59


MT2A S^b unrestraint low/high spin
Mean C^l ls
Mean C^l ds
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60

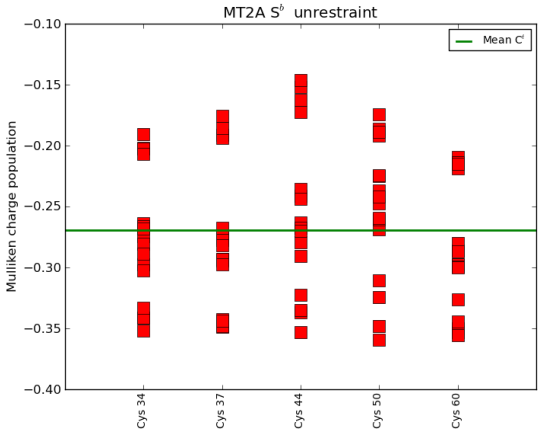
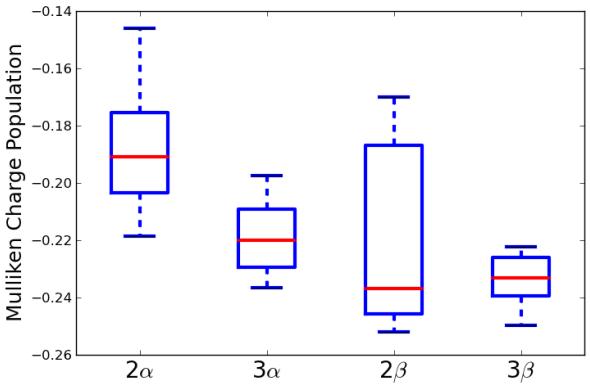
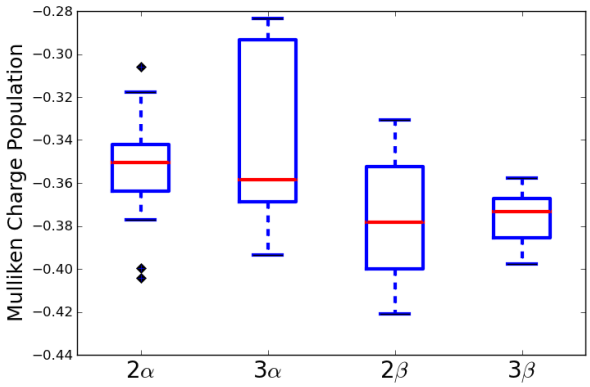

MT2A S^b unrestraint
Mean C^l
Mulliken charge population
−0.10
−0.15
−0.20
−0.25
−0.30
−0.35
−0.40
Cys 34
Cys 37
Cys 44
Cys 50
Cys 60

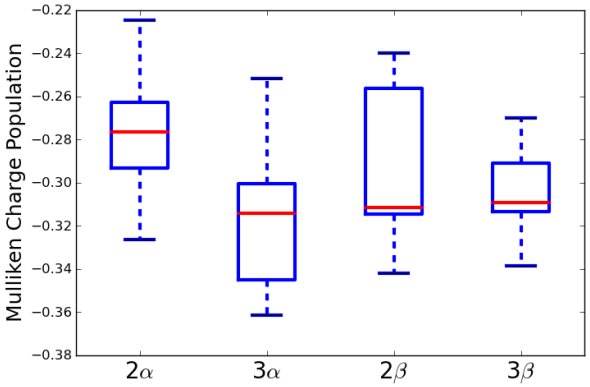
Figure S9. Mulliken charges separated into metal ion types, restrained models. Red signifies average values, blue spreads in charges.



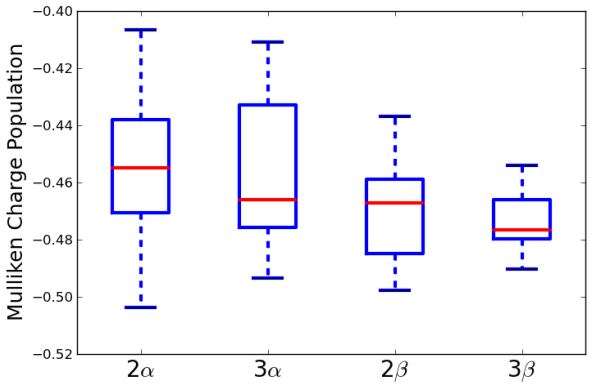
Restrained, S^b bound between Cu/Cu



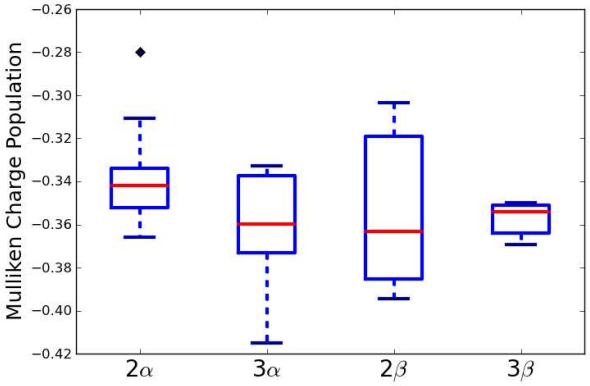
Restrained, S^t bound to Cu



Restrained, S^b bound between Cu/Zn

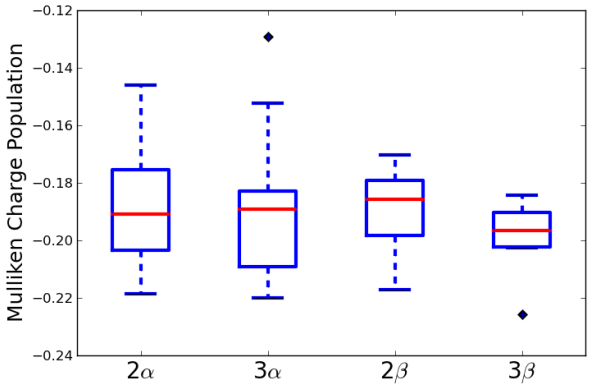


Restrained, S^t bound to Zn

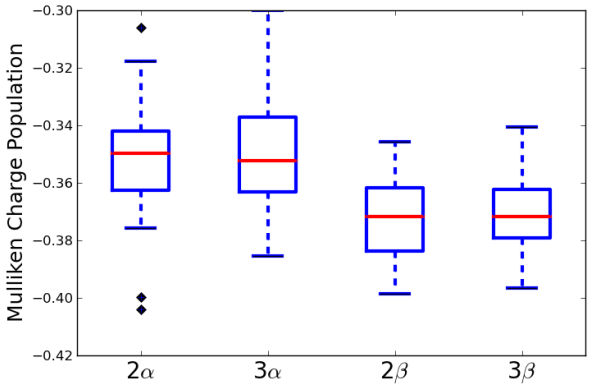


Restrained, S^b bound between Zn/Zn

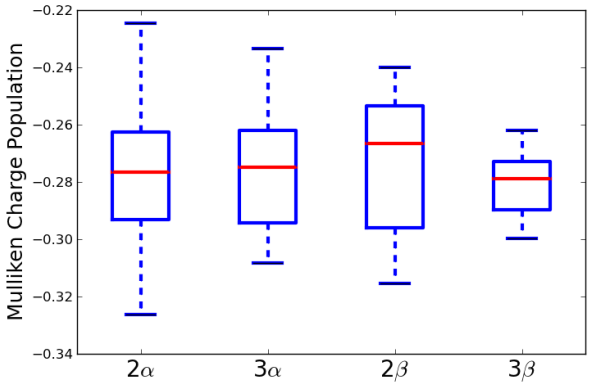
Figure S10. Mulliken charges separated into metal ion types, unrestrained models. Red signifies average values, blue spreads in charges.



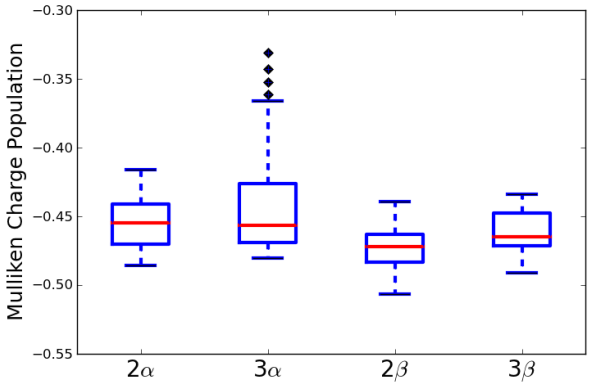
Unrestrained, S^b bound between Cu/Cu



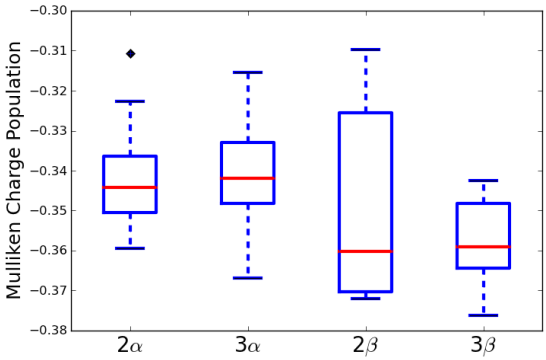
Unrestrained, S^t bound to Cu



Unrestrained, S^b bound between Cu/Zn



Unrestrained, S^t bound to Zn



Unrestrained, S^b bound between Zn/Zn

Table S56. Energies for metal substitution between cluster types and isomers, MT2. Favored form in bold.

Isomer	**E(α)**	**E(β)**		**E(α)**	**E(β)**	**ΔE (kJ / mol)**
Zn_4 + $CuZn_2$	-12444.556672	-9551.238469	$CuZn_3$ + Zn_3	-12303.527613	-9692.264201	-8.74
Zn_4 + Cu_2Zn	-12444.556672	-9410.221564	$CuZn_3$ + $CuZn_2$	-12303.527613	-9551.238469	-31.91
$CuZn_3$ + $CuZn_2$	-12303.527613	-9551.238469	**Cu_2Zn_2 + Zn_3**	-12162.510495	-9692.264201	22.62
Zn_4 + Cu_3	-12444.556672	-9269.194671	$CuZn_3$ + Cu_2Zn	-12303.527613	-9410.221564	-5.69
Zn_4 + Cu_3	-12444.556672	-9269.194671	Cu_2Zn_2 +$CuZn_2$	-12162.510495	-9551.238469	-6.25
Zn_4 + Cu_3	-12444.556672	-9269.194671	Cu_3Zn + Zn_3	-12021.482491	-9692.264201	-12.21
$CuZn_3$ + Cu_2Zn	-12303.527613	-9410.221564	Cu_2Zn_2 + $CuZn_2$	-12162.510495	-9551.238469	-0.56
Cu_3Zn + Zn_3	-12021.482491	-9692.264201	**Cu_2Zn_2 + $CuZn_2$**	-12162.510495	-9551.238469	5.97
Cu_2Zn_2 + $CuZn_2$	-12162.510495	-9551.238469	**$CuZn_3$ + Cu_2Zn**	-12303.527613	-9410.221564	0.56
Cu_3Zn + $CuZn_2$	-12021.482491	-9551.238469	**$CuZn_3$ + Cu_3**	-12303.527613	-9269.194671	3.48
Cu_4 + Zn_3	-11880.458628	-9692.264201	$CuZn_3$ + Cu_3	-12303.527613	-9269.194671	-1.43
Cu_4 + Zn_3	-11880.458628	-9692.264201	**Cu_2Zn_2 + Cu_2Zn**	-12162.510495	-9410.221564	24.23
Cu_4 + Zn_3	-11880.458628	-9692.264201	Cu_3Zn + $CuZn_2$	-12021.482491	-9551.238469	-4.91
Cu_3Zn + $CuZn_2$	-12021.482491	-9551.238469	**Cu_2Zn_2 + Cu_2Zn**	-12162.510495	-9410.221564	29.14
Cu_2Zn_2 + Cu_2Zn	-12162.510495	-9410.221564	$CuZn_3$ + Cu_3	-12303.527613	-9269.194671	-25.66
Cu_4 + $CuZn_2$	-11880.458628	-9551.238469	**Cu_3Zn + Cu_2Zn**	-12021.482491	-9410.221564	18.27
Cu_3Zn + Cu_2Zn	-12021.482491	-9410.221564	**Cu_2Zn_2 + Cu_3**	-12162.510495	-9269.194671	2.92
Cu_4 + Cu_2Zn	-11880.458628	-9410.221564	Cu_3Zn + Cu_3	-12021.482491	-9269.194671	-7.96

Table S57. Energies for metal substitution between cluster types and isomers, MT3. Favored form in bold.

Isomer	**E(α)**	**E(β)**		**E(α)**	**E(β)**	**ΔE (kJ/mol)**
Zn_4 + $CuZn_2$	-12444.551488	-9551.246547	$CuZn_3$ + Zn_3	-12303.529559	-9692.267116	-3.57
Zn_4 + Cu_2Zn	-12444.551488	-9410.222395	**$CuZn_3$ + $CuZn_2$**	-12303.529559	-9551.246547	5.84
$CuZn_3$ + $CuZn_2$	-12303.529559	-9551.246547	Cu_2Zn_2 + Zn_3	-12162.506764	-9692.267116	-5.84
Zn_4 + Cu_3	-12444.551488	-9269.198102	**$CuZn_3$ + Cu_2Zn**	-12303.529559	-9410.222395	6.21
Zn_4 + Cu_3	-12444.551488	-9269.198102	**Cu_2Zn_2 +$CuZn_2$**	-12162.506764	-9551.246547	9.77
Zn_4 + Cu_3	-12444.551488	-9269.198102	**Cu_3Zn + Zn_3**	-12021.483555	-9692.267116	2.84
$CuZn_3$ +Cu_2Zn	-12303.529559	-9410.222395	**Cu_2Zn_2 + $CuZn_2$**	-12162.506764	-9692.267116	3.56
Cu_3Zn + Zn_3	-12021.483555	-9692.267116	**Cu_2Zn_2 + $CuZn_2$**	-12162.506764	-9551.246547	6.93
Cu_2Zn_2 + $CuZn_2$	-12162.506764	-9551.246547	$CuZn_3$ + Cu_2Zn	-12303.529559	-9410.222395	-3.56
Cu_3Zn + $CuZn_2$	-12021.483555	-9551.246547	$CuZn_3$ + Cu_3	-12303.529559	-9269.198102	-6.41
Cu_4 + Zn_3	-11880.459435	-9692.267116	**$CuZn_3$ + Cu_3**	-12303.529559	-9269.198102	2.91
Cu_4 + Zn_3	-11880.459435	-9692.267116	**Cu_2Zn_2 + Cu_2Zn**	-12162.506764	-9410.222395	6.85
Cu_4 + Zn_3	-11880.459435	-9692.267116	**Cu_3Zn + $CuZn_2$**	-12021.483555	-9551.246547	9.32
Cu_3Zn + $CuZn_2$	-12021.483555	-9551.246547	Cu_2Zn_2 + Cu_2Zn	-12162.506764	-9410.222395	-2.48
Cu_2Zn_2 + Cu_2Zn	-12162.506764	-9410.222395	$CuZn_3$ + Cu_3	-12303.529559	-9269.198102	-3.93
Cu_4 + $CuZn_2$	-11880.459435	-9551.246547	Cu_3Zn + Cu_2Zn	-12021.483555	-9410.222395	-0.08
Cu_3Zn + Cu_2Zn	-12021.483555	-9410.222395	Cu_2Zn_2 + Cu_3	-12162.506764	-9269.198102	-2.85
Cu_4 + Cu_2Zn	-11880.459435	-9410.222395	Cu_3Zn + Cu_3	-12021.483555	-9269.198102	-0.45

Table S58. Bond orders from the MO overlap computed with the Mulliken population analysis.

Cluster α	**Formula**	**No.**	**M_S**	**Zn-S^t_{avg}**	**Cu-S^t_{avg}**	**Zn-S^b_{avg}**	**Cu-S^b_{avg}**	**Composition**
	Zn_4	1	0	1.00 / 1.01	-- / --	0.80 / 0.81	-- / --	Zn-Zn-Zn-Zn
	Zn_3Cu	2	½	1.00 / 1.00	0.95 / 0.96	0.82 / 0.82	0.94 / 0.76	Cu-Zn-Zn-Zn
		3	½	0.98 / 0.98	1.01 / 0.96	0.80 / 0.79	0.83 / 0.82	Zn-Cu-Zn-Zn
		4	½	0.99 / 0.98	0.98 / 0.99	0.81 / 0.78	0.98 / 0.85	Zn-Zn-Cu-Zn
		5	½	0.96 / 0.97	0.94 / 0.92	0.80 / 0.80	0.94 / 0.81	Zn-Zn-Zn-Cu
	Zn_2Cu_2	6	0 / 0	1.02 / 0.96	0.96 / 0.94	0.86 / 0.84	0.81 / 0.76	Cu-Cu-Zn-Zn
		7	0 / 1	0.97 / 0.99	0.99 / 1.00	0.81 / 0.83	0.80 / 0.83	Cu-Zn-Cu-Zn
		8	1 / 0	0.96 / 0.96	0.94 / 0.94	0.83 / 0.83	0.77 / 0.78	Cu-Zn-Zn-Cu
		9	0 / -	0.97 / -	0.96 / -	0.76 / -	0.83 / -	Zn-Cu-Cu-Zn
		10	0 / 0	0.96 / 0.97	1.01 / 1.01	0.81 / 0.80	0.80 / 0.81	Zn-Cu-Zn-Cu
		11	0 / 0	0.95 / 0.96	0.91 / 0.92	0.74 / 0.73	0.65 / 0.64	Zn-Zn-Cu-Cu
	$ZnCu_3$	12	½ / -	0.97 / -	0.99 / -	0.80 / -	0.82 / -	Cu-Cu-Cu-Zn
		13	½ / ½	0.94 / 1.00	1.00 / 1.00	0.87 / 0.83	0.81 / 0.79	Cu-Cu-Zn-Cu
		14	0 / ½	0.91 / 0.94	0.96 / 0.94	0.79 / 0.78	0.80 / 0.80	Cu-Zn-Cu-Cu
		15	½/3/2	0.97 / 0.93	0.94 / 0.95	0.76 / 0.76	0.84 / 0.81	Zn-Cu-Cu-Cu
	Cu_4	16	1 / 2	-- / --	0.99 / 0.94	-- / --	0.81 / 0.84	Cu-Cu-Cu-Cu
Cluster β	**Formula**	**No.**	**M_S**	**Zn-S^t_{avg}**	**Cu-S^t_{avg}**	**Zn-S^b_{avg}**	**Cu-S^b_{avg}**	**Composition**
	Zn_3		0	1.01 / 0.97	-- / --	0.81 / 0.79	-- / --	Zn-Zn-Zn
	Zn_2Cu	1	½	1.00 / 0.97	0.92 / 0.94	0.80 / 0.80	0.77 / 0.75	Cu-Zn-Zn
		2	½	0.99 / 0.99	0.97 / 0.90	0.83 / 0.83	0.80 / 0.80	Zn-Cu-Zn
		3	½	0.97 / 0.97	0.94 / 0.96	0.80 / 0.83	0.79 / 0.80	Zn-Zn-Cu
	$ZnCu_2$	4	0 / 1	0.97 / 0.97	1.01 / 0.92	0.85 / 0.82	0.83 / 0.78	Cu-Cu-Zn
		5	0 / 1	1.01 / 0.93	0.96 / 0.94	0.83 / 0.79	0.76 / 0.80	Cu-Zn-Cu
		6	0 / 0	0.95 / 0.97	1.00 / 0.96	0.83 / 0.84	0.77 / 0.78	Zn-Cu-Cu
	Cu_3		½ / ½	-- / --	0.94 / 0.93	-- / --	0.77 / 0.79	Cu-Cu-Cu

Table S59. Spin Populations for MT2α clusters.

Cys 33	Cys 34	Cys 36	Cys 37	Cys 41	Cys 44	Cys 48	Cys 50	Cys 57	Cys 59	Cys 60	Metal (62-63-64-65)	M_S
											Zn-Zn-Zn-Zn	0
0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.09	0.23	0.22	0.09	Cu-Zn-Zn-Zn	½
0.22	0.11	0.00	0.00	0.00	0.11	0.22	0.00	0.00	0.00	0.00	Zn-Cu-Zn-Zn	½
0.00	0.01	0.00	0.11	0.20	0.17	0.01	0.00	0.01	0.01	0.12	Zn-Zn-Cu-Zn	½
0.01	0.11	0.20	0.09	0.00	0.00	0.01	0.20	0.01	0.01	0.00	Zn-Zn-Zn-Cu	½
0.22	0.10	0.00	0.00	0.00	0.08	0.25	0.10	0.22	0.22	0.09	Cu-Cu-Zn-Zn	1
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Cu-Zn-Cu-Zn	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Cu-Zn-Zn-Cu	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Zn-Cu-Cu-Zn	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Zn-Cu-Zn-Cu	0
0.01	0.12	0.15	-0.03	-0.15	-0.15	-0.01	0.16	0.00	0.00	-0.10	Zn-Zn-Cu-Cu	0
0.00	0.00	0.00	0.01	0.00	0.00	-0.01	0.10	0.20	0.20	0.13	Cu-Cu-Cu-Zn	½
0.20	0.10	-0.04	-0.01	0.00	0.10	0.20	0.01	0.06	0.03	0.04	Cu-Cu-Zn-Cu	½
0.01	0.12	0.15	0.25	0.17	0.15	0.01	0.10	-0.21	-0.20	0.06	Cu-Zn-Cu-Cu	½
0.20	0.02	-0.14	0.02	0.18	0.28	0.16	-0.14	0.00	0.00	0.08	Zn-Cu-Cu-Cu	½
0.22	0.10	-0.03	0.07	0.20	0.29	0.18	0.00	0.08	0.03	0.13	Cu-Cu-Cu-Cu	1

Table S60. Spin Populations for MT3α clusters.

Cys 34	Cys 35	Cys 37	Cys 38	Cys 42	Cys 45	Cys 49	Cys 51	Cys 64	Cys 66	Cys 67	Metal (69-70-71-72)	M_S
											Zn-Zn-Zn-Zn	0
0.22	0.11	0.00	0.00	0.00	0.11	0.21	0.00	0.00	0.00	0.00	Cu-Zn-Zn-Zn	½
0.02	0.13	0.17	0.11	0.00	0.00	0.00	0.19	0.00	0.01	0.00	Zn-Cu-Zn-Zn	½
0.00	0.00	0.00	0.14	0.19	0.15	0.02	0.00	0.00	0.01	0.12	Zn-Zn-Cu-Zn	½
0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.08	0.24	0.22	0.09	Zn-Zn-Zn-Cu	½
-0.16	0.09	0.13	0.05	0.00	-0.04	0.16	0.10	0.00	0.00	0.00	Cu-Cu-Zn-Zn	0
0.22	0.11	0.01	0.12	0.21	0.30	0.20	0.00	0.00	0.01	0.10	Cu-Zn-Cu-Zn	1
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Cu-Zn-Zn-Cu	0
N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	Zn-Cu-Cu-Zn	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Zn-Cu-Zn-Cu	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Zn-Zn-Cu-Cu	0
N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D	Cu-Cu-Cu-Zn	
0.20	0.08	-0.04	-0.02	0.00	0.12	0.18	0.01	0.06	0.38	0.04	Cu-Cu-Zn-Cu	½
-0.03	-0.02	0.00	0.01	0.06	0.01	-0.04	0.13	0.19	0.17	0.13	Cu-Zn-Cu-Cu	½
0.02	0.09	0.23	0.28	0.19	0.11	0.01	0.37	0.24	0.19	0.23	Zn-Cu-Cu-Cu	3/2
0.23	0.26	0.17	0.27	0.18	0.29	0.20	0.25	0.24	0.23	0.25	Cu-Cu-Cu-Cu	2

Table S61. Spin Populations for MT2β clusters.

Cys 5	Cys 7	Cys 13	Cys 15	Cys 19	Cys 21	Cys 24	Cys 26	Cys 29	Metal (66-67-68)	M_S
0.20	0.14	0.00	0.00	0.01	0.18	0.13	0.00	0.00	Cu-Zn-Zn	½
0.02	0.00	0.00	0.12	0.17	0.00	0.12	0.00	0.17	Zn-Cu-Zn	½
0.00	0.10	0.20	0.11	0.00	0.01	0.00	0.20	0.00	Zn-Zn-Cu	½
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Cu-Cu-Zn	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Cu-Zn-Cu	0
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Zn-Cu-Cu	0
0.01	0.00	0.00	0.15	0.18	0.00	0.12	0.00	0.16	Cu-Cu-Cu	½

Table S62. Spin Populations for MT3β clusters.

Cys 6	Cys 8	Cys 14	Cys 16	Cys 20	Cys 22	Cys 25	Cys 27	Cys 30	Metal (390-391-392)	M_S
0.20	0.09	0.00	0.00	0.00	0.19	0.14	0.01	0.01	Cu-Zn-Zn	½
0.01	0.00	0.00	0.13	0.20	0.00	0.11	0.00	0.19	Zn-Cu-Zn	½
0.00	0.11	0.21	0.09	0.01	0.00	0.00	0.20	0.00	Zn-Zn-Cu	½
0.19	0.10	0.01	0.13	0.21	0.19	0.26	0.01	0.19	Cu-Cu-Zn	1
0.20	0.23	0.21	0.10	0.01	0.17	0.13	0.21	0.00	Cu-Zn-Cu	1
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	Zn-Cu-Cu	0
0.19	0.23	0.17	-0.03	0.17	0.17	0.02	0.18	-0.15	Cu-Cu-Cu	½