Supporting Information

Integration of Trinuclear Triangle Copper(II) Secondary Building Units in Octacyanidometallates(IV)-based Frameworks

Olaf Stefanczyk,[†] Kunal Kumar,[†] Ting Yun Pai,[†] Guanping Li,[†] and Shin-ichi Ohkoshi*[†]*

Department of Chemistry, School of Science, The University of Tokyo, 7–3–1 Hongo,

Bunkyo–ku, Tokyo 113–0033, Japan.

*Corresponding authors: olaf@chem.s.u-tokyo.ac.jp, ohkoshi@chem.s.u–tokyo.ac.jp

Table of Contents

1. Infrared (IR) spectroscopy.....3

Figure S1. Infrared (IR) absorption spectra of 1 and 2, and reference samples.....3

Table S1. Interpretation of the IR spectra of **1** and **2**.....3

2. Solid-state UV–Vis–NIR spectroscopy.....**4**

Figure S2. Solid-state UV–Vis–IR absorption spectra of **1** and **2**.....4

Table S2. Comparison of the solid-state UV–Vis spectra of **1** and **2**, other 1D chains, 2D layers, and 3D Cu^{II}–$[M^{IV}(CN)_8]^{4-}$ (M^{IV} = Mo, W) networks.....4

3. Single-crystal X-ray diffraction (SXRD) analysis.....**5**

Table S3. Crystal data, data collection, and refinement parameters for **1** and **2**.....5

Figure S3. Thermal ellipsoids plot of the structural unit of **2** with selected atoms labeled. Arrangement of the TTC units. Crystal structure of **2** projected along the crystallographic *c*-axis.....6

Figure S4. Crystal structure of **1** projected along the crystallographic *a*- and *b*-axes.....7

Figure S5. Crystal structure of **2** projected along the crystallographic *a*- and *b*-axes.....8

Figure S6. *H* bonding and π interactions in the crystal structure of **1** projected along the crystallographic *a*-, *b*-, and *c*-axes.....9

Figure S7. *H* bonding and π interactions in the crystal structure of **2** projected along the crystallographic *a*-, *b*-, and *c*-axes, respectively.....10

Table S4. Selected distances and angles of **1** and **2**.....11

Table S5. Results of the continuous shape measurement analysis for the Cu^{II} and M^{IV} centers.....12

4. Computational details.....**13**

Table S6. Atomic coordinates for the optimized **[(TTC)(CN)$_3$]** model ($S = {}^1/_2$).....13

Table S7. Atomic coordinates for the optimized **[(TTC)(CN)$_3$]** model ($S = {}^3/_2$).....14

Table S8. Atomic coordinates for the **$[M(CN)_8]^{4-}$** model.....15

Results of broken symmetry magnetic coupling analysis for **[(TTC)(CN)$_3$]** panel.....16

Figure S8. The optimized geometry for the **[(TTC)(CN)$_3$]** panel, with determined intermetallic distances, assuming total spin $S = {}^3/_2$ and $S = {}^1/_2$.....17

Figure S9. Optimized models of the **[(TTC)(CN)$_3$]** units for the quantum chemical calculations assuming total spins of $^1/_2$ and $^3/_2$. Corresponding energy level diagrams.....17

Figure S10. Models of $[W(CN)_8]^{4-}$ and $[Mo(CN)_8]^{4-}$ used in the quantum chemical calculations, and corresponding energy level diagrams. 18

Figure S11. Projection of the calculated near HOMO–LUMO gap orbitals for $[(TTC)(CN)_3]$ units assuming total spins of $^1/_2$ and $^3/_2$. 19

Figure S12. Projection of the calculated near HOMO–LUMO gap orbitals for $[W(CN)_8]^{4-}$ and $[Mo(CN)_8]^{4-}$ units....20

Excitation energies and oscillator strengths (*f*) calculated for $[(TTC)(CN)_3]$ ($S = {}^1/_2$). 21

Excitation energies and oscillator strengths (*f*) calculated for $[W(CN)_8]^{4-}$. 28

Excitation energies and oscillator strengths (*f*) calculated for $[Mo(CN)_8]^{4-}$. 34

Figure S13. Comparison of the measured solid-state UV–Vis–NIR absorption spectra for **1** and **2** and calculated for $[(TTC)(CN)_3]$, $[W(CN)_8]^{4-}$, and $[Mo(CN)_8]^{4-}$ models. Enlarged spectra of **1** and **2** with absorption bands corresponding to the theoretically predicted HOMO–LUMO transition for β spins. 40

5. Powder X-ray diffraction (PXRD) studies 41

Figure S14. Experimental and simulated PXRD patterns based on the SXRD data for **1** and **2**....41

6. Thermogravimetric analysis 42

Figure S15. Thermogravimetric plots for powdered samples of **1** and **2** with the weight losses due to the removal of solvent indicated....42

7. References 43

1. Infrared (IR) spectroscopy

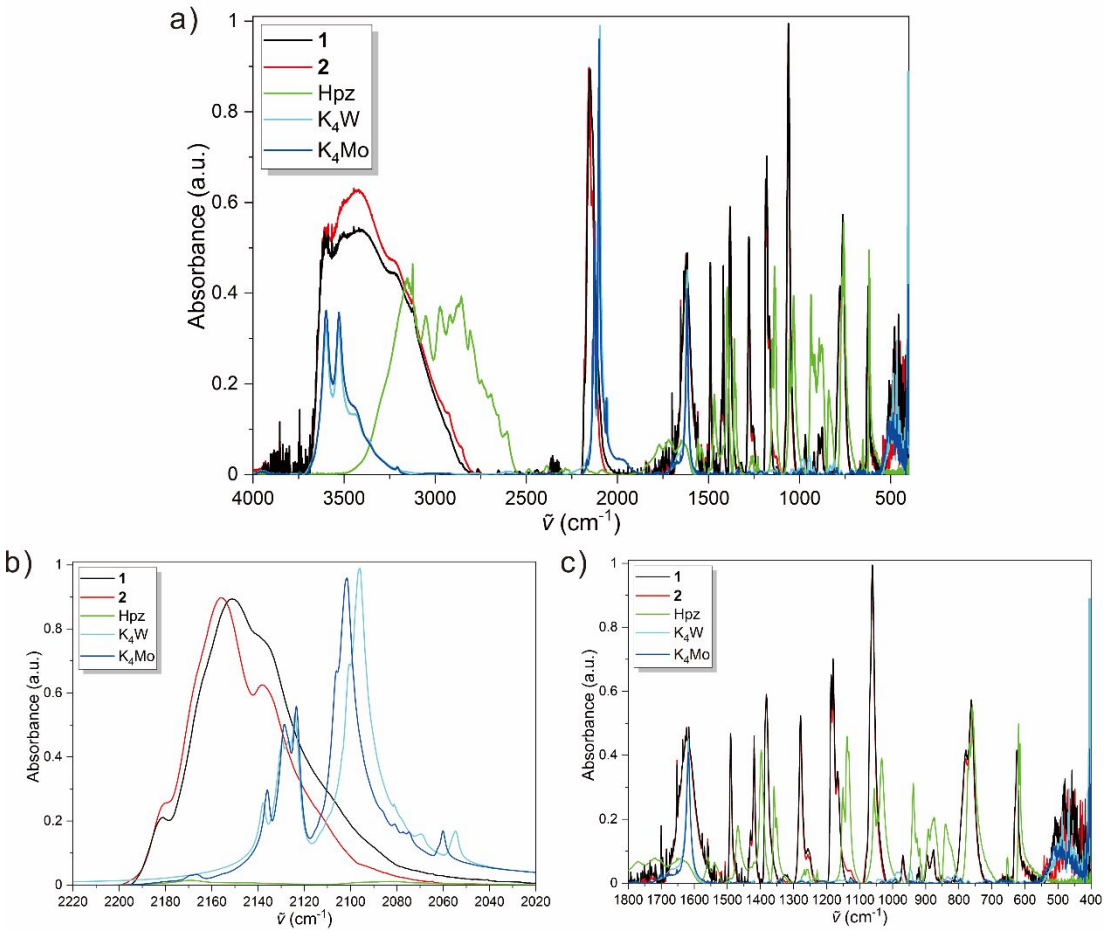

Figure S1. Infrared (IR) absorption spectra of **1** and **2**, and reference samples $K_4[W(CN)_8]\cdot 2H_2O$ (K_4W), $K_4[Mo(CN)_8]\cdot 2H_2O$ (K_4Mo), and pyrazole (Hpz) in (a) the full range, (b) the fingerprint, and (c) the CN^--stretching bands.

Table S1. Interpretation of the IR spectra of **1** and **2**.

1_W	2_Mo	Assignment [S1]
~3600m(br)	~3600m(br)	$[\nu(O–H)]_{water,OH}$
~3420s(br)	~3420s(br)	$[\nu(C–H)]_{pz}$
2181w(sh), 2151vs, 2138s(sh), 2114w(sh)	2180w(sh), 2156vs, 2138m(sh), 2114w(sh)	$[\nu(C\equiv N)]_{CN}$
1623m	1623m	$[\delta(O–H)]_{water}$
1490m, 1430vw(sh), 1419m	1490m, 1430vw(sh), 1419m	$[\delta(C–H)]_{pz}$
1382m, 1279m, 1256vw	1382m, 1279m, 1256vw	$[\nu(C=C)]_{pz}$, $[\nu(N–N)]_{pz}$, $[\nu(C–N)]_{pz}$, $[\nu(C–C)]_{pz}$

1186s, 1180s, 1166w, 969vw, 925vw, 921vw, 886vw, 877vw, 779w, 762m, 624m	1187m, 1180s, 1166w, 969vw, 925vw, 921vw, 886vw, 878vw, 779w, 762m, 624m	$[\delta$(H–ArC–H out–of–plane)$]_{pz}$, $[\delta$(ArC–H in–plane)$]_{pz}$, $[\pi$(ArC–H ring breathing)$]_{pz}$, $[\pi$(ArC–H ring deformation)$]_{pz}$ $[\delta$(M–OH)$]_{OH}$
509vw, 480w, 458w	533vw, 473w, 430w	$[\nu$(Mo–C/W–C)$]_{M(CN)8}$, $[\nu$(Cu–N/O)$]_{SBU}$

2. Solid-state UV–Vis–NIR spectroscopy

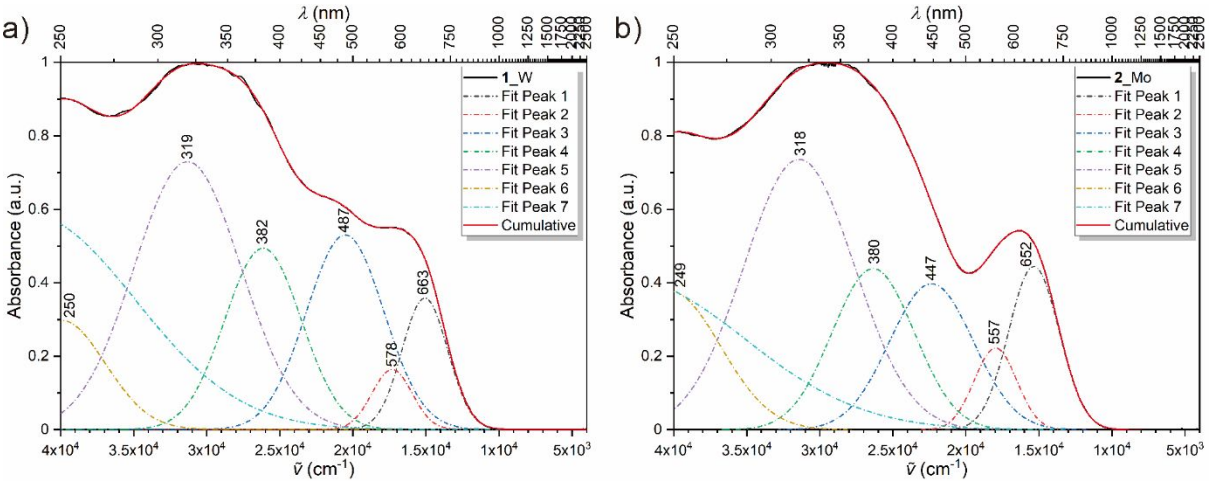

Figure S2. (a, b) Solid-state UV–Vis–IR absorption spectra of **1** and **2**. Colored peaks and red lines correspond to a Gaussian deconvolution of the experimental spectra.

Table S2. Comparison of the solid-state UV–Vis spectra of **1** and **2**, other 1D chains, 2D layers, and 3D Cu^{II}–$[M^{IV}(CN)_8]^{4-}$ (M^{IV} = Mo, W) networks.

	Compound	Assignment				
		LMCT of Cu^{II} MLCT of $[M(CN)_8]^{4-}$	LF of $[M(CN)_8]^{4-}$	MMCT of Cu^{II}–M^{IV}	LF of Cu(II)	Ref.
1D chains	$[Cu(bapa)]_2[Mo(CN)_8]\cdot 7H_2O$ bapa = bis(3–aminopropyl)amine	n/d	302w	466m(sh)	686s(br), 906m(sh)	S2
	$[Cu((rac)\text{–chxn})_2]_2[Mo(CN)_8]\cdot 3H_2O$ (*rac*)–chxn = (*rac*)–1,2–diaminocyclohexane	n/d	≈ 340vs	436s	549m	S3
	$[Cu((S,S)\text{–chxn})_2]_2[Mo(CN)_8]\cdot H_2O$ (*S,S*)–chxn = (*S,S*)–1,2–diaminocyclohexane	n/d	≈ 320vs	436m	559w	S3
	$[Cu((R,R)\text{–chxn})_2]_2[Mo(CN)_8]\cdot H_2O$ (*R,R*)–chxn = (*R,R*)–1,2–diaminocyclohexane	n/d	≈ 320vs	436m	556w	S3
	$\{[Cu(en)_2]_2[Cu(en)_2][Mo(CN)_8]_2\}\cdot[Cu(en)_2(H_2O)_2]\cdot 8H_2O$ en = ethylenediamine	< 300vs	≈ 350s	≈ 525w(sh)	≈ 600s(br)	S4
	$[Cu(tren)][Cu(bapa)][Mo(CN)_8]\cdot 4H_2O$ tren = tris(2–aminoethyl)amine	< 300vs		≈ 450m	≈ 650s, ≈ 900m(br)	S5
2D layers	$[Cu(aepa)]_2[Mo(CN)_8]\cdot 6H_2O$ aepa = *N*–(2–aminoethyl)–1,3–propanediamine	n/d		≈ 450s(br)	≈ 600s(br)	S6
	$[Cu(bapa)]_3[Mo(CN)_8]_{1.5}\cdot 12.5H_2O$ bapa = bis(3–aminopropyl)amine	< 250vs	≈ 300s	≈ 450m	≈ 650s, ≈ 800m(br)	S7
	$[Cu(bapen)]_2[Mo(CN)_8]\cdot 4H_2O$ bapen = 1,2–bis(3–aminopropylamino)ethane	< 275vs	≈ 400m	≈ 450w(sh)	≈ 600m(br)	S7
	$K_4\{[Cu(ida)]_2[W(CN)_8]\}\cdot 4H_2O$ ida^{2-} = iminodiacetate	193m, 247vs, 284s,	322m, 335vw, 364vw, 430s(br)	511w	652m(br), 802w(br)	S8

	$K_4\{[Cu(ida)]_2[Mo(CN)_8]\}\cdot 4H_2O$ ida^{2-} = iminodiacetate	204m, 244vs, 273w,	301vs, 337vw, 360vw, 414m(br)	498vw	610w, 720m(br)	**S8**
3D networks	$\{[Cu^{II}_3(\mu_3\text{–}OH)(\mu\text{–}pz)_3(H_2O)_3]_2[W^{IV}(CN)_8]\}\cdot nH_2O$ (**1**), pz = pyrazole anion	237m(br), 250w	319s, 382m	487m	578vw, 663w	**This work**
	$\{[Cu^{II}_3(\mu_3\text{-}OH)(\mu\text{-}pz)_3(H_2O)_3]_2[Mo^{IV}(CN)_8]\}\cdot nH_2O$ (**2**), pz = pyrazole anion	231w(br), 249w	318vs, 380m	447m	557w, 652m	**This work**
	$[Cu(tn)_2][Cu(tn)][Mo(CN)_8]\cdot 3.5H_2O$ tn = 1,3–propanediamine	< 275vs	≈ 350m	≈ 450m	≈ 650s(br)	**S7**
	$Cu_2[Mo(CN)_8]_2(PVP)_7\cdot 11H_2O$ PVP = polyvinylpyrrolidone	n/d		529s	n/d	**S9**
	$Cu_2[Mo(CN)_8]\cdot 2H_2O$	< 300vs		≈ 520s	n/d	**S10**
	$Cu_2[Mo(CN)_8]\cdot 7.6H_2O$	n/d	≈ 400m	≈ 600m(br)	n/d	**S11**
	$Cs_2Cu_7[Mo(CN)_8]_4\cdot 6H_2O$	n/d		≈ 525s, ≈ 650m(br)	n/d	**S12**

3. Single-crystal X-ray diffraction (SXRD) analysis

Table S3. Crystal data, data collection, and refinement parameters for **1** and **2**.

	1_W	**2**_Mo
Empirical formula	$C_{52}H_{40}Cu_{12}N_{40}O_{19}W_2$	$C_{52}H_{82}Cu_{12}Mo_2N_{40}O_{25}$
M (g mol^{-1})	2659.42	2621.93
T (K)	120	120
Crystal system, space group	Orthorhombic, *P*bcn	Orthorhombic, *P*bcn
a (Å)	19.6219(10)	19.919(5)
b (Å)	15.9763(8)	14.988(4)
c (Å)	15.6324(7)	15.696(4)
$\alpha = \beta = \gamma$ (°)	90	90
V (Å^3)	4900.5(4)	4686(2)
Z, ρ_{calc} (g cm^{-3})	4, 1.802	4, 1.858
μ (mm^{-1})	4.954	3.005
F(000)	2560	2612
Crystal size (mm^3)	0.08 x 0.06 x 0.04	0.10 x 0.03 x 0.02
Radiation, λ (Å)	Mo Kα (0.71073)	Mo Kα (0.71073)
2θ range for data collection (°)	5.526 to 55.81	5.58 to 55.108
Index ranges	−25 ≤ h ≤ 25 −20 ≤ k ≤ 20 −20 ≤ l ≤ 20	−25 ≤ h ≤ 25 −19 ≤ k ≤ 19 −18 ≤ l ≤ 20
Unique reflections collected	70555 / 5767 [R_{int} = 0.0739, R_σ = 0.0360]	49523 / 5381 [R_{int} = 0.4179, R_σ = 0.2906]
Refinement method	Full matrix least squares on F^2	Full matrix least squares on F^2
Data / restraints / parameters	5767 / 18 / 312	5381 / 18 / 329

Goodness-of-fit on F^2	1.065	0.998
Final R indexes [$I \geq 2\sigma(I)$]	$R_1 = 0.0393,\ wR_2 = 0.0984$	$R_1 = 0.0767,\ wR_2 = 0.1500$
Final R indexes [all data]	$R_1 = 0.0641,\ wR_2 = 0.1105$	$R_1 = 0.2622,\ wR_2 = 0.2200$
Largest diff. peak / hole ($e \cdot Å^{-3}$)	1.50 / −1.45	2.06 / −1.79
CCDC Number	2166440	2166441

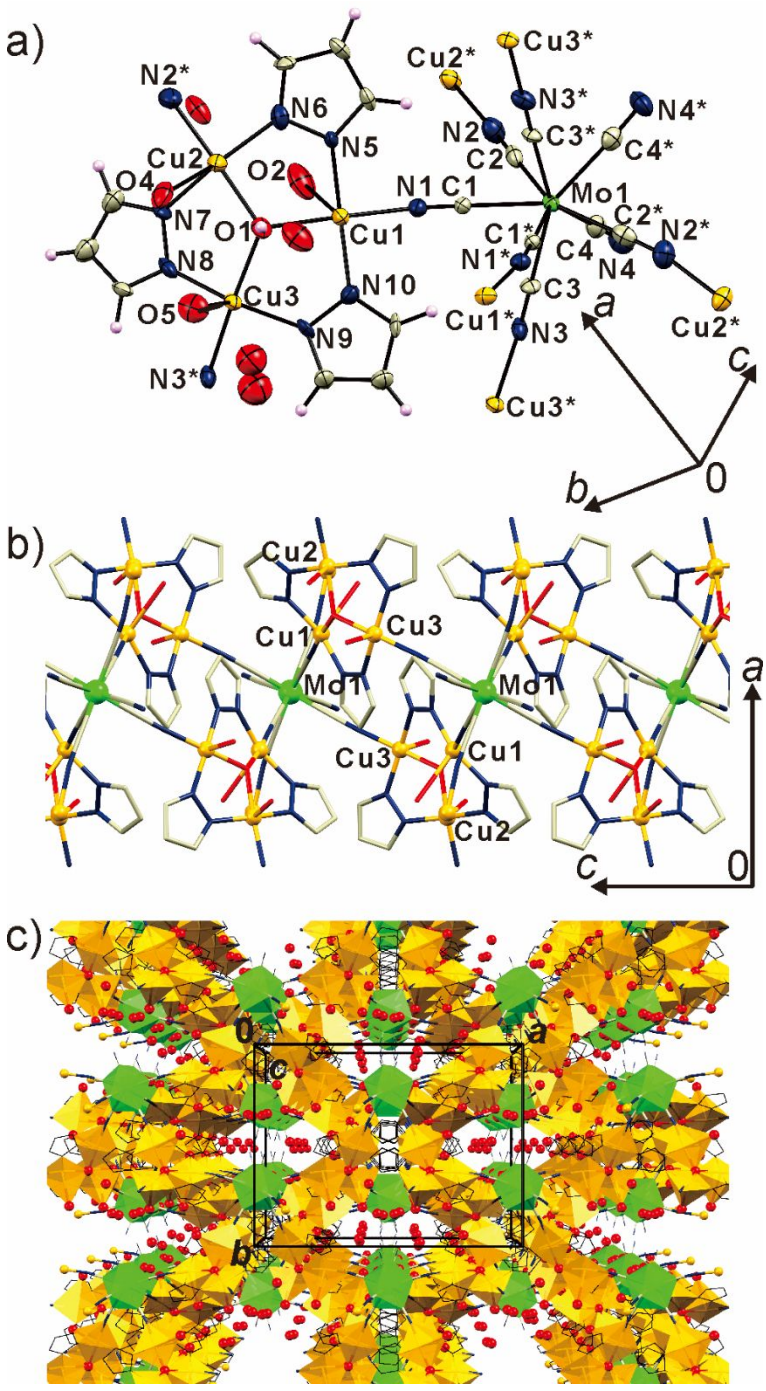

Figure S3. (a) Thermal ellipsoids plot of the structural unit of **2** with selected atoms labeled. (b) Arrangement of the TTC units. (c) Crystal structure of **2** projected along the crystallographic *c*-

axis. Yellow, green, black, and red indicate Cu, Mo, pyrazole, and H_2O, respectively. Hydrogen atoms in (b) and (c) are omitted for clarity.

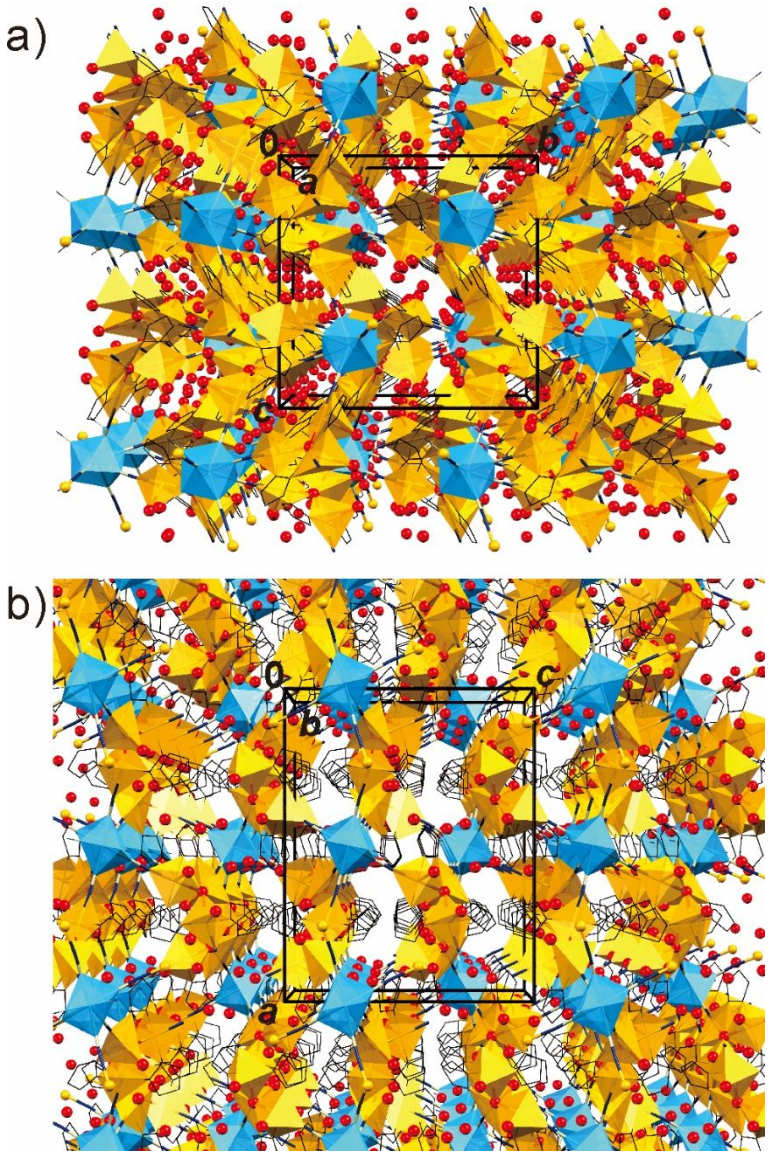

Figure S4. Crystal structure of **1** projected along the crystallographic (a) *a*- and (b) *b*-axes. Yellow, blue, black, and red indicate Cu, W, pyrazole, and H_2O, respectively. Hydrogen atoms are omitted for clarity.

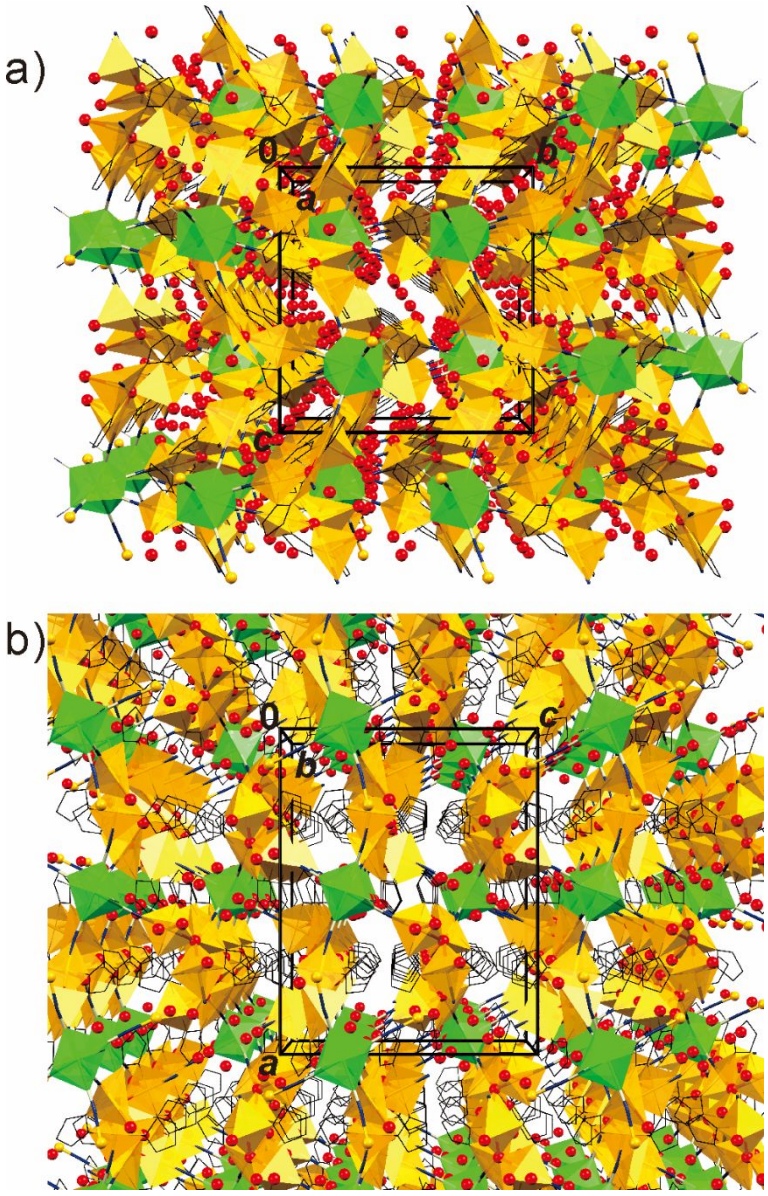

Figure S5. Crystal structure of **2** projected along the crystallographic (a) *a*- and (b) *b*-axes. Yellow, green, black, and red indicate Cu, Mo, pyrazole, and H_2O, respectively. Hydrogen atoms are omitted for clarity.

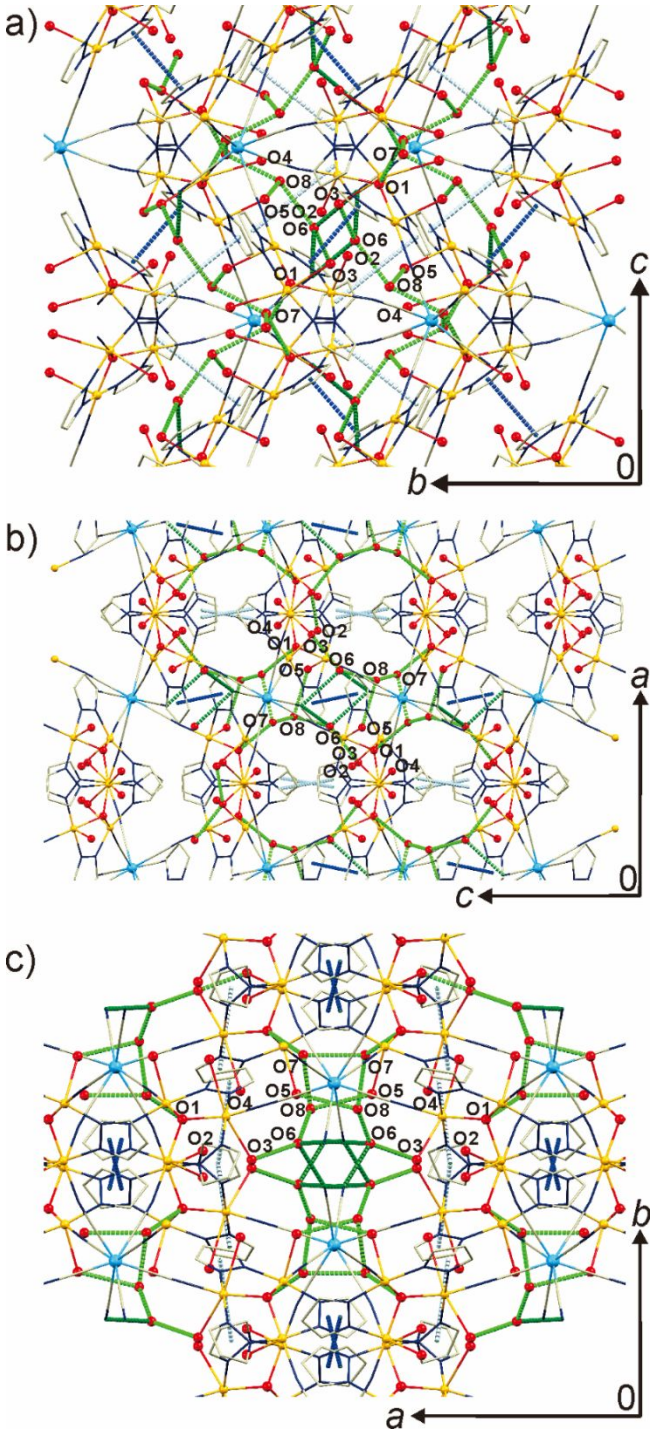

Figure S6. *H* bonding (H_2O–H_2O/OH^- and H_2O–terminal CN^-: green and dark green dotted sticks, respectively) and π interactions (edge-to-face and face-to-face: light blue and blue dotted sticks, respectively) in the crystal structure of **1** projected along the crystallographic (a) *a*-, (b)

b-, and (c) *c*-axes, respectively. Yellow, blue, gray, light blue, and red indicate Cu, W, C, N, and O, respectively. Hydrogen atoms are omitted for clarity.

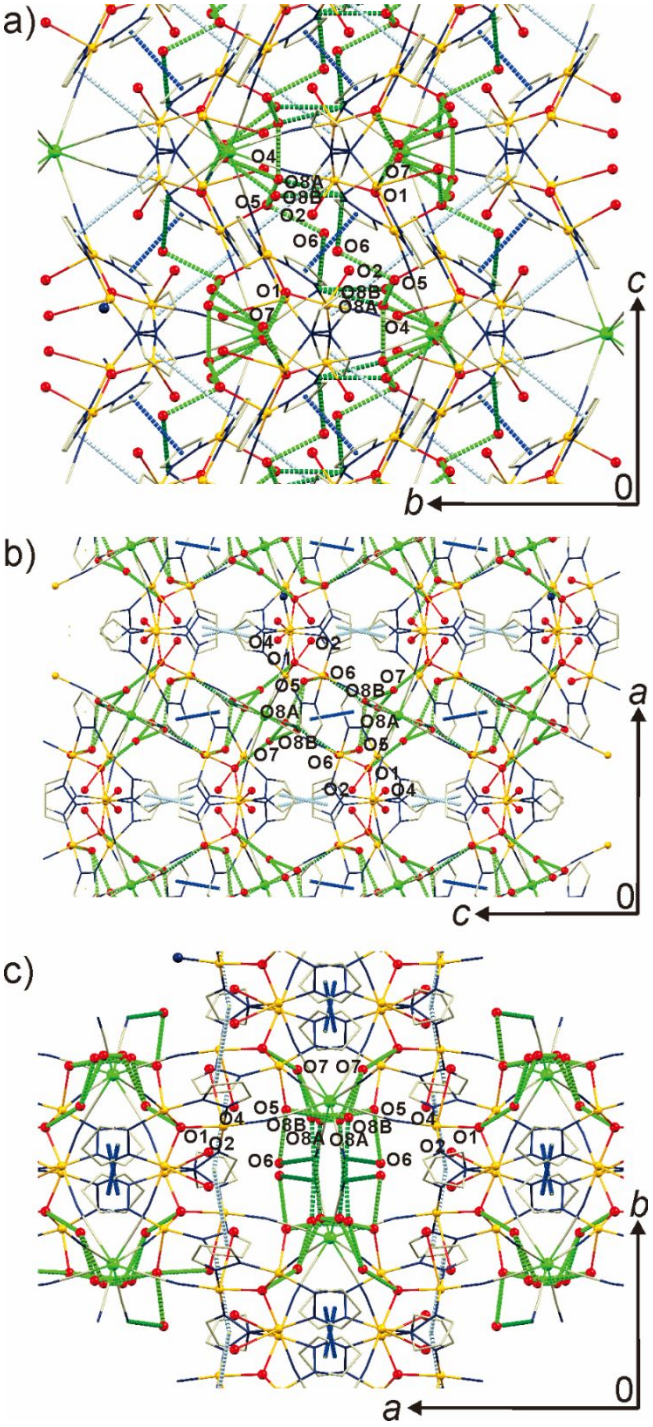

Figure S7. *H* bonding (H_2O–H_2O/OH^- and H_2O-terminal CN^-: green and dark green dotted sticks, respectively) and π interactions (edge-to-face and face-to-face: light blue and blue dotted sticks, respectively) in the crystal structure of **2** projected along the crystallographic (a) *a*-, (b) *b*-, and (c) *c*-axes, respectively. Yellow, green, gray, light blue, and red indicate Cu, Mo, C, N, and O, respectively. Hydrogen atoms are omitted for clarity.

Table S4. Selected distances and angles of **1** and **2**.

Bond lengths (Å)					
	1_W	**2**_Mo		**1**_W	**2**_Mo
M1–$C1_{CN}$	2.151(6)	2.174(12)	Cu3–$N3_{CN}^{\#2}$	1.976(5)	2.004(10)
M1–$C2_{CN}$	2.145(5)	2.136(14)	Cu3–$N8_{pz}$	1.940(5)	1.938(10)
M1–$C3_{CN}$	2.145(5)	2.155(12)	Cu3–$N9_{pz}$	1.937(5)	1.940(10)
M1–$C4_{CN}$	2.159(7)	2.172(14)	Cu3–$O1_{OH}$	1.980(4)	1.980(7)
<M–C_{CN}>	2.150(6)	2.159(13)	Cu3–$O5_{H2O}$	2.426(9)	2.333(9)
C1≡$N1_{CN}$	1.143(7)	1.112(13)	<Cu–N_{CN}>	1.972(5)	1.988(11)
C2≡$N2_{CN}$	1.142(7)	1.161(15)	<Cu–N_{pz}>	1.943(5)	1.942(10)
C3≡$N3_{CN}$	1.151(7)	1.127(13)	<Cu–O_{OH}>	1.995(4)	1.984(8)
C4≡$N4_{CN}$	1.138(9)	1.137(15)	<Cu–O_{H2O}>	2.540(15)	2.609(13)
<C≡N_{CN}>	1.144(8)	1.134(14)	$N5_{pz}$–$N6_{pz}$	1.364(6)	1.372(12)
Cu1–$N1_{CN}$	1.976(5)	1.991(10)	$N7_{pz}$–$N8_{pz}$	1.365(7)	1.382(13)
Cu1–$N5_{pz}$	1.944(5)	1.930(9)	$N9_{pz}$–$N10_{pz}$	1.368(7)	1.366(13)
Cu1–$N10_{pz}$	1.941(5)	1.942(10)	<N_{pz}–N_{pz}>	1.366(7)	1.373(13)
Cu1–$O1_{OH}$	2.014(4)	1.992(8)	Cu1···Cu2	3.296(1)	3.280(2)
Cu1–$O2_{H2O}$	2.401(14)	2.682(18)	Cu1···Cu3	3.320(1)	3.350(2)
Cu2–$N2_{CN}^{\#1}$	1.965(5)	1.970(12)	Cu2···Cu3	3.404(1)	3.410(2)
Cu2–$N6_{pz}$	1.951(5)	1.953(10)	<Cu···Cu>	3.340(1)	3.347(2)
Cu2–$N7_{pz}$	1.946(5)	1.950(9)	Cu1···M1	5.232(1)	5.249(1)
Cu2–$O1_{OH}$	1.990(4)	1.981(8)	Cu2···M1	5.248(1)	5.241(1)
Cu2–$O3_{H2O}$	2.561(17)	——	Cu3···M1	5.268(1)	5.283(1)
Cu2–$O4_{H2O}$	2.77(2)	2.812(12)	<Cu···M>	5.249(1)	5.258(1)
Angles (˚)					
	1_W	**2**_Mo		**1**_W	**2**_Mo
C1–M1–$C1^{\#3}$	73.8(3)	72.1(6)	Cu1–N1≡C1	168.2(5)	170.2(10)
C1–M1–C2	72.0(2)	70.4(4)	Cu2–N2≡C2	175.2(5)	169.4(11)
C1–M1–$C2^{\#3}$	141.8(2)	141.6(4)	Cu3–N3≡C3	174.5(5)	176.1(10)
C1–M1–C3	74.2(2)	76.2(4)	<Cu–N≡C>	172.6(5)	171.9(10)
C1–M1–$C3^{\#3}$	77.3(2)	75.5(4)	Cu1–$O1_{OH}$–Cu2	110.8(2)	111.3(4)
C1–M1–C4	120.6(2)	125.0(4)	Cu1–$O1_{OH}$–Cu3	112.4(2)	115.0(4)
C1–M1–$C4^{\#3}$	138.9(2)	135.2(4)	Cu2–$O1_{OH}$–Cu3	118.0(2)	118.8(4)
C2–M1–$C2^{\#3}$	145.4(3)	147.8(6)	<Cu–O–Cu>	113.7(2)	115.0(4)
C2–M1–C3	108.6(2)	102.9(4)	N1–Cu1–$O1_{OH}$	177.1(2)	174.8(4)
C2–M1–$C3^{\#3}$	82.1(2)	86.8(4)	$N2^{\#1}$–Cu2–$O1_{OH}$	164.8(2)	172.6(4)
C2–M1–C4	75.1(2)	75.6(5)	$N3^{\#2}$–Cu3–$O1_{OH}$	176.8(2)	175.7(4)

C2–M1–C4$^{\#3}$	77.8(2)	79.0(4)	<N–Cu–O>	172.9(2)	174.4(4)
C3–M1–C3$^{\#3}$	144.2(3)	144.9(6)	$N5_{pz}$–Cu1–$N10_{pz}$	160.1(2)	160.5(4)
C3–M1–C4	71.5(3)	70.8(5)	$N6_{pz}$–Cu2–$N7_{pz}$	170.9(2)	169.4(4)
C3–M1–C4$^{\#3}$	143.3(2)	143.9(5)	$N8_{pz}$–Cu3–$N9_{pz}$	171.3(2)	166.5(4)
C4–M1–C4$^{\#3}$	75.8(4)	75.0(7)	<N_{pz}–Cu–N_{pz}>	167.4(2)	165.5(4)
M1–C1≡N1	176.3(5)	176.9(10)			
M1–C2≡N2	178.9(6)	178.3(11)			
M1–C3≡N3	178.2(6)	178.8(11)			
M1–C4≡N4	179.0(6)	176.5(12)			
<M–C≡N>	178.1(6)	177.6(11)			

$^{\#1}$ $^{3}/_{2}$-x,½+y,+z; $^{\#2}$ 1-x,1-y,-z; $^{\#3}$ 1-x,+y, ½-z

Table S5. Results of the continuous shape measurement analysis for the Cu^{II} and M^{IV} centers.

Center	Geometry			Center	Geometry		
	S_{TBPY-5}	S_{SPY-5}	S_{vOC-5}		S_{BTPR-8}	S_{SAPR-8}	S_{TDD-8}
ideal TBPY–5	0.000	5.384	7.342	ideal BTPR	0	2.262	2.709
ideal SPY–5	5.384	0.000	1.741	ideal SAPR	2.267	0	2.848
ideal vOC–5	7.342	1.741	0.000	ideal TDD	2.717	2.848	0
1_Cu1	3.470	1.792	2.350	**1**_W1	1.803	0.416	1.367
1_Cu2	3.739	2.265	2.784	**2**_Mo1	1.856	1.218	0.653
1_Cu3	5.210	0.840	1.145				
2_Cu1	4.741	3.559	4.677				
2_Cu2	5.235	2.910	3.810				
2_Cu3	4.501	0.747	1.143				

S_{TBPY-5}: the shape measure relative to a trigonal bipyramid; S_{SPY-5}: the shape measure relative to a square pyramid; S_{vOC-5}: the shape measure relative to a vacant octahedron; S_{BTPR-8}: the shape measure relative to a bicapped trigonal prism; S_{SAPR-8}: the shape measure relative to a square antiprism; S_{TDD-8}: the shape measure relative to a triangular dodecahedron; a smaller S value reflects a better match with the ideal geometry ($S = 0$).[S13, S14]

4. Computational details

Table S6. Atomic coordinates for the optimized **[(TTC)(CN)$_3$]** model ($S = {}^1/_2$).

Center number	Atomic number		Atomic type	Coordinates (Å)		
				X	Y	Z
1	Cu	29	0	1.76220	0.80342	–0.31391
2	Cu	29	0	–0.41842	–1.96150	–0.49396
3	Cu	29	0	–1.72744	1.24424	0.17620
4	N	7	0	–0.42954	2.74847	0.02329
5	N	7	0	0.87912	2.58307	–0.16142
6	O	8	0	–0.07726	0.07172	0.11381
7	H	1	0	0.02595	–0.16746	1.06412
8	N	7	0	–2.29455	–1.62802	0.02780
9	C	6	0	–4.07925	–0.53094	0.70606
10	H	1	0	–4.65383	0.35990	0.98358
11	N	7	0	–2.77504	–0.40878	0.35846
12	C	6	0	3.66408	–1.50806	–0.36947
13	H	1	0	4.49115	–0.79378	–0.28882
14	C	6	0	1.47516	3.79877	–0.22729
15	H	1	0	2.55777	3.86277	–0.38442
16	C	6	0	0.49270	4.79094	–0.07412
17	H	1	0	0.62938	5.87789	–0.07916
18	C	6	0	–4.22303	3.12373	0.34750
19	N	7	0	–3.26846	2.41970	0.26579
20	C	6	0	–3.28990	–2.53435	0.16242
21	H	1	0	–3.09652	–3.58621	–0.07978
22	C	6	0	–4.45322	–1.88005	0.60210
23	H	1	0	–5.43514	–2.31997	0.80681
24	N	7	0	1.55138	–2.11391	–0.50452
25	C	6	0	2.29430	–3.23975	–0.60880
26	H	1	0	1.79278	–4.20428	–0.75237
27	C	6	0	3.65594	–2.90371	–0.52081
28	H	1	0	4.51880	–3.57644	–0.57087
29	N	7	0	2.38506	–1.06011	–0.35903
30	C	6	0	–0.70552	4.07439	0.08060
31	H	1	0	–1.74026	4.40604	0.22248
32	N	7	0	–0.74728	–3.72625	–1.26011
33	N	7	0	3.50628	1.56323	–0.69480

34	C	6	0	4.58816	2.00928	–0.90483
35	C	6	0	–0.94482	–4.81431	–1.69696

Table S7. Atomic coordinates for the optimized **[(TTC)(CN)$_3$]** model ($S = {}^3/_2$).

Center number	Atomic number		Atomic type	Coordinates (Å)		
				X	Y	Z
1	Cu	29	0	1.74323	–1.05848	–0.30853
2	Cu	29	0	0.03430	1.94682	0.07898
3	Cu	29	0	–1.77941	–0.99668	–0.30860
4	N	7	0	–0.72422	–2.59010	0.18531
5	N	7	0	0.63272	–2.61377	0.18586
6	O	8	0	–1.75E–4	–0.00402	–0.65539
7	H	1	0	0.00333	0.18034	–1.61775
8	N	7	0	–1.93931	1.91855	–0.09928
9	C	6	0	–3.95729	1.09721	–0.41880
10	H	1	0	–4.69393	0.30394	–0.59048
11	N	7	0	–2.64614	0.77947	–0.31464
12	C	6	0	3.99303	0.95784	–0.42362
13	H	1	0	4.70127	0.13911	–0.59526
14	C	6	0	1.04217	–3.84803	0.56133
15	H	1	0	2.11397	–4.07225	0.60846
16	C	6	0	–0.08173	–4.64964	0.81917
17	H	1	0	–0.10009	–5.69479	1.14609
18	C	6	0	–4.44061	–2.57810	–0.79578
19	N	7	0	–3.43345	–1.97928	–0.59433
20	C	6	0	–2.80284	2.96008	–0.06013
21	H	1	0	–2.42372	3.97130	0.12668
22	C	6	0	–4.10865	2.48508	–0.26500
23	H	1	0	–5.03672	3.06606	–0.29417
24	N	7	0	2.00543	1.84957	–0.10237
25	C	6	0	2.90487	2.86032	–0.06560
26	H	1	0	2.56159	3.88437	0.12062
27	C	6	0	4.19300	2.33975	–0.27168
28	H	1	0	5.14076	2.88790	–0.30279
29	N	7	0	2.67172	0.68623	–0.31732
30	C	6	0	–1.17678	–3.80930	0.56049
31	H	1	0	–2.25580	–3.99596	0.60684
32	N	7	0	0.06610	3.72718	0.86355
33	N	7	0	3.36203	–2.09867	–0.59244
34	C	6	0	4.34775	–2.73252	–0.79275
35	C	6	0	0.08566	4.81690	1.33856

Table S8. Atomic coordinates for the $[M(CN)_8]^{4-}$ model.

Center number	Atomic number		Atomic type	Coordinates (Å)		
				X	Y	Z
1	Mo/W	42/74	0	0.01409	0.00608	0
2	N	7	0	1.69782	–2.86452	0
3	N	7	0	–0.01868	2.69791	1.95479
4	N	7	0	–3.08852	1.14214	0
5	N	7	0	0.08031	–1.00478	–3.14542
6	N	7	0	3.21921	0.87501	0
7	N	7	0	–2.06141	–2.57261	0
8	C	6	0	–1.34799	–1.66106	0
9	C	6	0	0.03568	–0.63229	–2.05214
10	C	6	0	1.14711	–1.85242	0
11	C	6	0	–2.01982	0.70608	0
12	C	6	0	0.03568	1.75664	1.2898
13	C	6	0	2.10729	0.55551	0
14	N	7	0	–0.01868	2.69791	–1.95479
15	N	7	0	0.08031	–1.00478	3.14542
16	C	6	0	0.03568	–0.63229	2.05214
17	C	6	0	0.03568	1.75664	–1.2898

Results of broken symmetry magnetic coupling analysis for **[(TTC)(CN)$_3$]** panel

Calculation output for the spin–flip operation for Cu1.

$\hat{S}_{HS}$ = 1.5
$\langle \hat{S}_{HS}{}^2 \rangle$ = 3.7581
$\langle \hat{S}_{BS}{}^2 \rangle$= 1.7274
E_{HS} = -5951.460530 E_h
E_{BS} = -5951.461847 E_h
$E_{HS} - E_{BS}$ = 0.0358 eV 288.941 cm^{-1} (ANTIFERROMAGNETIC coupling)

| Spin-Hamiltonian Analysis based on $\hat{H}_{HDvV} = -2J_{HS-BS}\hat{S}_{HS}\hat{S}_{BS}$ |

| J_1 = -128.42 cm^{-1} (from -($E_{HS} - E_{BS}$)/$S_{max}{}^2$) |S15,S16,S17
| J_2 = -77.05 cm^{-1} (from -($E_{HS} - E_{BS}$)/(S_{max}·(S_{max}+1))) |S18
| J_3 = -142.29 cm^{-1} (from -($E_{HS} - E_{BS}$)/($\langle \hat{S}_{HS}{}^2 \rangle - \langle \hat{S}_{BS}{}^2 \rangle$)) |S19,S20

Calculation output for the spin–flip operation for Cu2.

$\hat{S}_{HS}$ = 1.5
$\langle \hat{S}_{HS}{}^2 \rangle$ = 3.7581
$\langle \hat{S}_{BS}{}^2 \rangle$ = 1.7240
E_{HS} = -5951.461143 E_h
E_{BS} = -5951.462627 E_h
$E_{HS} - E_{BS}$ = 0.0404 eV 325.708 cm^{-1} (ANTIFERROMAGNETIC coupling)

| Spin-Hamiltonian Analysis based on $\hat{H}_{HDvV} = -2J_{HS-BS}\hat{S}_{HS}\hat{S}_{BS}$ |

| J_1 = -144.76 cm^{-1} (from -($E_{HS} - E_{BS}$)/$S_{max}{}^2$) |S15,S16,S17
| J_2 = -86.86 cm^{-1} (from -($E_{HS} - E_{BS}$)/(S_{max}·(S_{max}+1))) |S18
| J_3 = -160.12 cm^{-1} (from -($E_{HS} - E_{BS}$)/($\langle \hat{S}_{HS}{}^2 \rangle - \langle \hat{S}_{BS}{}^2 \rangle$)) |S19,S20

Calculation output for the spin–flip operation for Cu3.

$\hat{S}_{HS}$ = 1.5

$\langle \hat{S}_{HS}{}^2 \rangle$ = 3.7581

$\langle \hat{S}_{BS}{}^2 \rangle$ = 1.7240

E_{HS} = -5951.461143 E_h

E_{BS} = -5951.462627 E_h

$E_{HS} - E_{BS}$ = 0.0404 eV 325.681 cm^{-1} (ANTIFERROMAGNETIC coupling)

| Spin-Hamiltonian Analysis based on $\hat{H}_{HDvV} = -2J_{HS-BS}\hat{S}_{HS}\hat{S}_{BS}$ |

| J_1 = -144.75 cm^{-1} (from -($E_{HS} - E_{BS}$)/$S_{max}{}^2$) |S15,S16,S17

| J_2 = -86.85 cm^{-1} (from -($E_{HS} - E_{BS}$)/(S_{max}·(S_{max}+1))) |S18

| J_3 = -160.11 cm^{-1} (from -($E_{HS} - E_{BS}$)/($\langle \hat{S}_{HS}{}^2 \rangle - \langle \hat{S}_{BS}{}^2 \rangle$)) |S19,S20

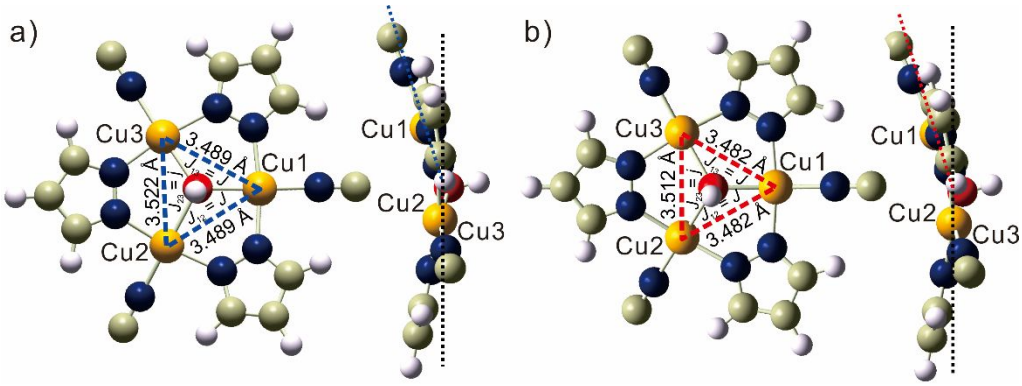

Figure S8. The optimized geometry for the **[(TTC)(CN)$_3$]** panel, with determined intermetallic distances, assuming total spin (a) $S = {}^1/_2$ and (b) $S = {}^3/_2$. The isosceles distribution of Cu(II) centers in the TTC panels is a consequence of Cu1 projecting beyond the plane described by the other two centers and their cyanides.

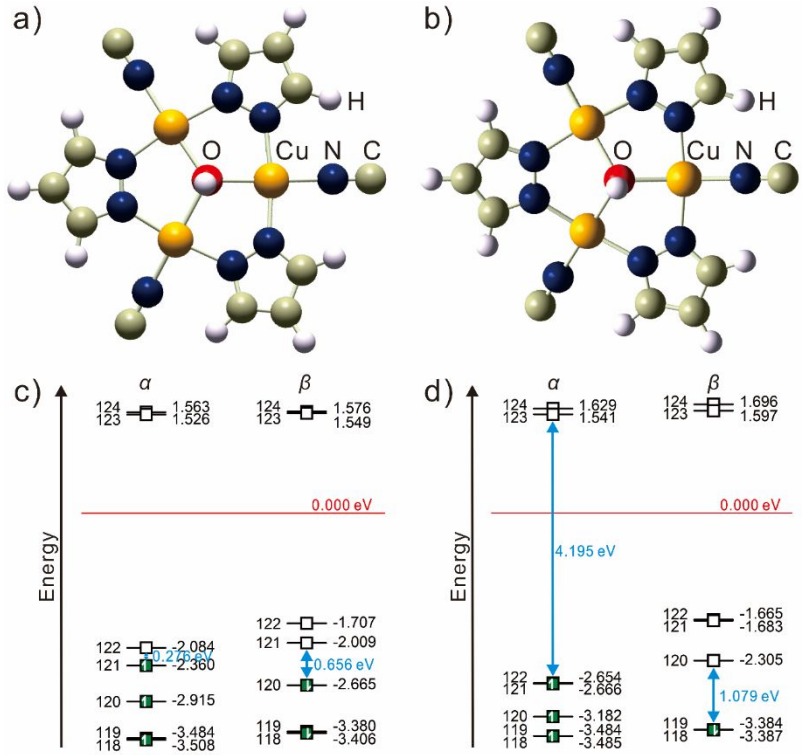

Figure S9. Optimized models of the **[(TTC)(CN)$_3$]** units for the quantum chemical calculations assuming total spins of (a) $^1/_2$ and (b) $^3/_2$. (c, d) Corresponding energy level diagrams (eV). Blue arrows and numbers correspond to the positions and values of the HOMO–LUMO gaps, respectively.

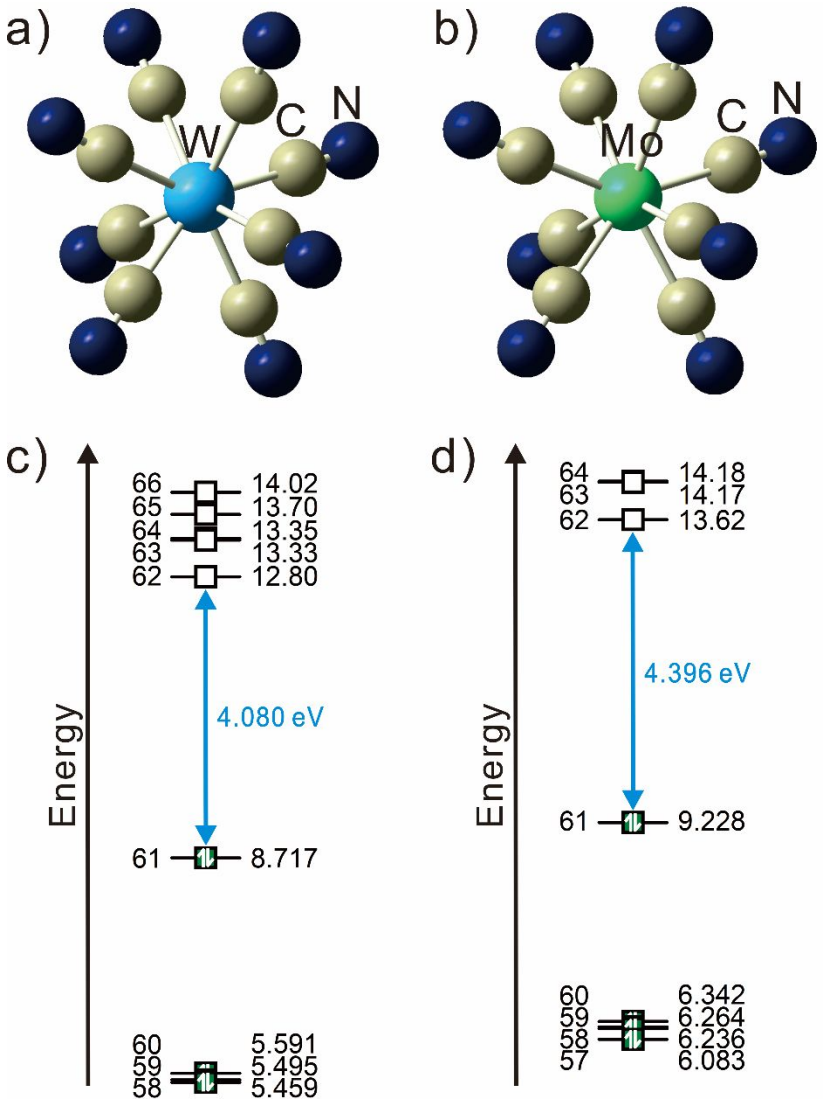

Figure S10. Models of (a) $[W(CN)_8]^{4-}$ and (b) $[Mo(CN)_8]^{4-}$ used in the quantum chemical calculations, and (c, d) corresponding energy level diagrams (eV). Blue arrows and numbers correspond to the positions and values of the HOMO–LUMO gaps, respectively.

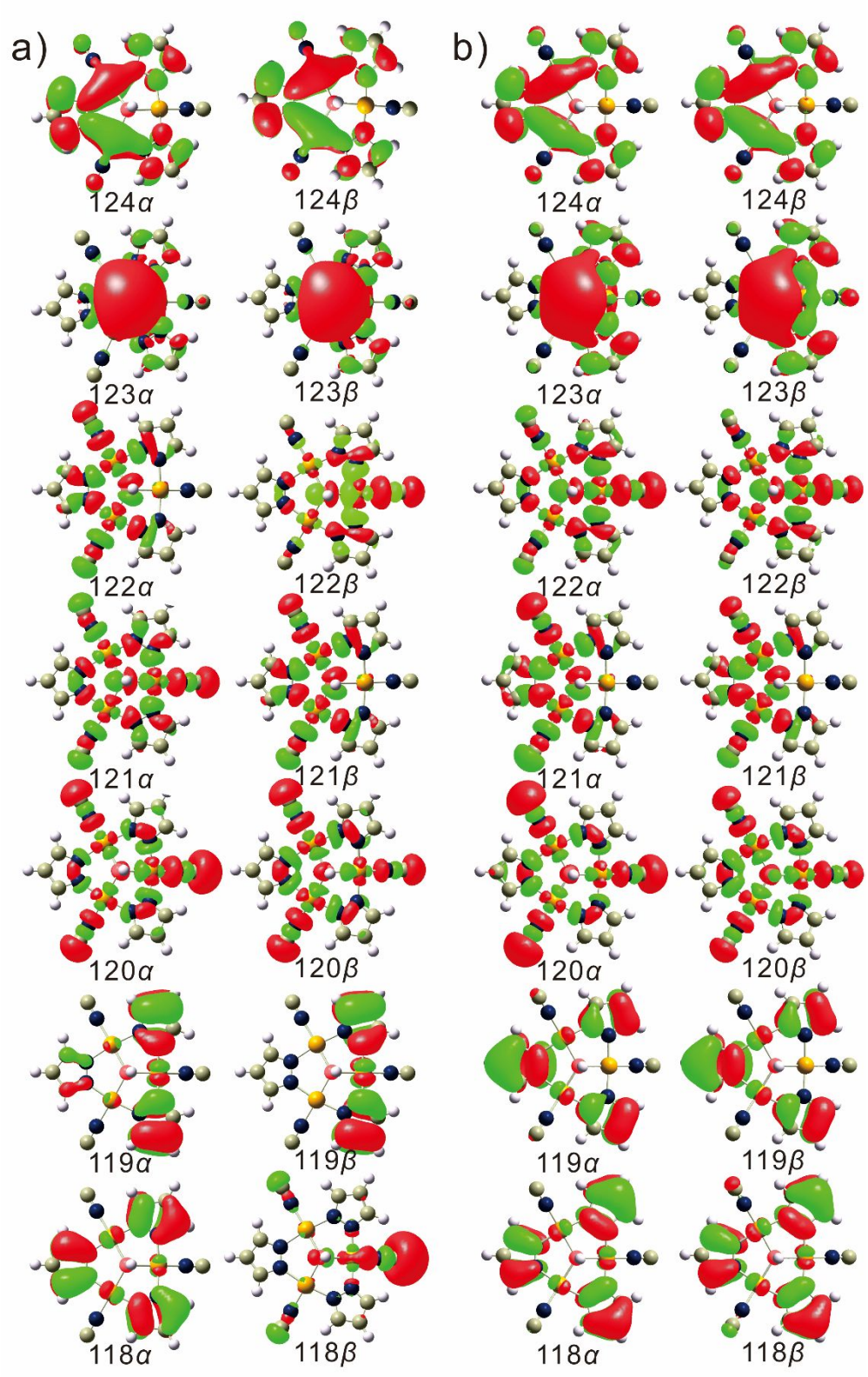

Figure S11. Projection of the calculated near HOMO–LUMO gap orbitals for **[(TTC)(CN)$_3$]** units assuming total spins of (a) $^1/_2$ and (b) $^3/_2$.

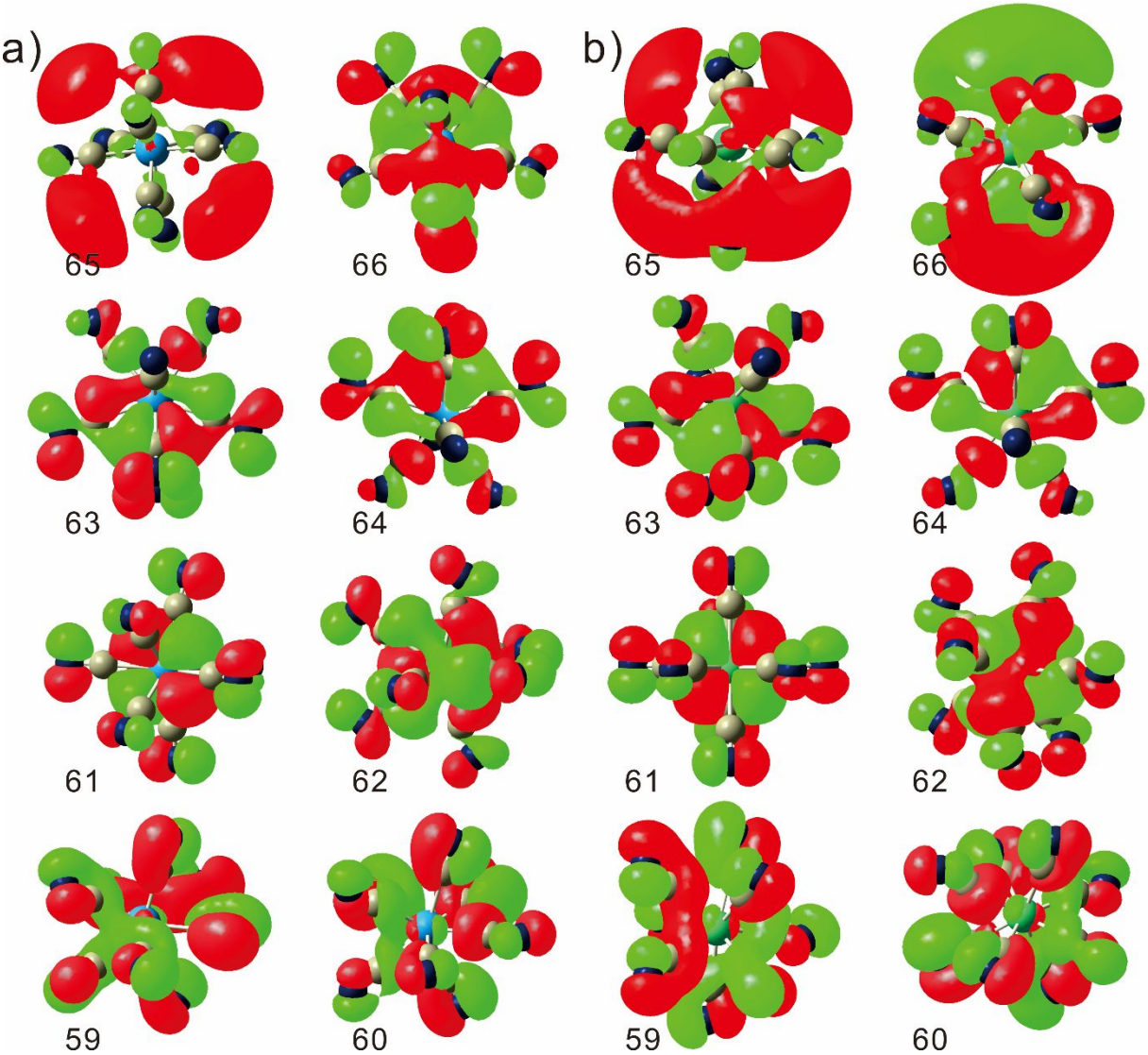

Figure S12. (a, b) Projection of the calculated near HOMO–LUMO gap orbitals for $[W(CN)_8]^{4-}$ and $[Mo(CN)_8]^{4-}$ units.

Excitation energies and oscillator strengths (*f*) calculated for **[(TTC)(CN)$_3$]** ($S = {}^1/_2$).

Excited State 1: 2.103-A 0.6191 eV 2002.78 nm *f* = 0.0039
<S^2> = 0.855
121A → 122A 0.94681
120B → 121B -0.23553
120B → 122B 0.21967

Excited State 2: 2.677-A 0.8874 eV 1397.17 nm *f* = 0.0040
<S^2> = 1.542
120A → 122A 0.64919
121A → 122A 0.22400
120B → 121B 0.72247

Excited State 3: 2.066-A 1.3295 eV 932.57 nm *f* = 0.0313
<S^2> = 0.817
120A → 122A -0.37290
117B → 121B -0.12631
120B → 121B 0.42210
120B → 122B 0.79128

Excited State 4: 2.950-A 1.4357 eV 863.57 nm *f* = 0.0002
<S^2> = 1.926
119A → 122A -0.18672
119B → 121B 0.97490

Excited State 5: 2.893-A 1.4557 eV 851.72 nm *f* = 0.0000
<S^2> = 1.842
117A → 122A 0.13031
118B → 121B 0.98585

Excited State 6: 2.953-A 1.5167 eV 817.44 nm *f* = 0.0142
<S^2> = 1.930
111A → 122A -0.11063
118A → 122A 0.19364
119A → 122A 0.56718
120A → 122A 0.29573
115B → 121B -0.23431
116B → 121B 0.57882
119B → 121B 0.11530
120B → 121B -0.21899
120B → 122B 0.24030

Excited State 7: 2.448-A 1.5252 eV 812.91 nm *f* = 0.0516
<S^2> = 1.248
111A → 122A -0.19939
116A → 122A -0.10578
118A → 122A -0.29849
119A → 122A -0.26302
120A → 122A 0.52385
121A → 122A -0.17310
111B → 121B -0.12189
116B → 121B -0.30510
117B → 121B -0.12489
117B → 122B 0.13554
120B → 121B -0.37281
120B → 122B 0.43071
120B <-121B 0.10143

Excited State 8: 2.752-A 1.5884 eV 780.56 nm *f* = 0.0017
<S^2> = 1.643
118A → 122A 0.65001
119A → 122A -0.61497
116B → 121B 0.30234
117B → 121B -0.10677
119B → 121B -0.16088
119B → 122B -0.18730

Excited State 9: 2.894-A 1.6045 eV 772.75 nm *f* = 0.0237
<S^2> = 1.844
115A → 122A 0.18824
116A → 122A 0.19194
118A → 122A 0.12296
115B → 121B -0.61862
116B → 121B -0.21178
117B → 121B 0.63565
117B → 122B 0.11566
120B → 122B 0.18100

Excited State 10: 2.681-A 1.6246 eV 763.16 nm *f* = 0.0001
<S^2> = 1.547
117A → 122A 0.98136
118B → 121B -0.12975

Excited State 11: 2.341-A 1.6450 eV 753.72 nm *f* = 0.0006
<S^2> = 1.120
118A → 122A 0.62628
119A → 122A 0.40630
115B → 121B 0.27027
116B → 121B -0.58820

Excited State 12: 3.160-A 1.6817 eV 737.28 nm *f* = 0.0235
<S^2> = 2.247
115A → 122A 0.28509
116A → 122A -0.32373
111B → 121B -0.26837
113B → 121B 0.10113
115B → 121B 0.47697
115B → 122B -0.13706
116B → 121B 0.20125
117B → 121B 0.57220
117B → 122B -0.28004

Excited State 13: 2.916-A 1.7541 eV 706.81 nm *f* = 0.0015
<S^2> = 1.876
114A → 122A 0.11612
116A → 122A -0.17369
114B → 121B 0.94299
115B → 121B -0.13383
117B → 122B 0.11306

Excited State 14: 2.615-A 1.7779 eV 697.36 nm *f* = 0.0036
<S^2> = 1.460

115A → 122A 0.71380
116A → 122A 0.20984
113B → 121B -0.21635
115B → 122B 0.25624
116B → 122B 0.11701
117B → 121B -0.15487
118B → 122B -0.27087
119B → 122B 0.44059

Excited State 15: 2.517-A 1.7805 eV 696.36 nm f= 0.0034
<S^2> = 1.333
115A → 122A -0.37190
116A → 122A -0.17123
118A → 122A 0.11335
119A → 122A -0.10344
113B → 121B 0.14091
114B → 121B -0.12816
115B → 122B -0.14397
117B → 121B 0.12884
118B → 122B -0.10449
119B → 122B 0.83599

Excited State 16: 2.535-A 1.7907 eV 692.36 nm f= 0.0001
<S^2> = 1.357
115A → 122A 0.13520
113B → 121B -0.12941
118B → 122B 0.94134
119B → 122B 0.19919

Excited State 17: 2.899-A 1.7935 eV 691.30 nm f= 0.0021
<S^2> = 1.852
113A → 122A 0.14431
115A → 122A 0.25445
111B → 121B -0.13008
113B → 121B 0.88347
114B → 121B -0.10137
115B → 122B 0.14987
117B → 121B -0.15336

Excited State 18: 3.147-A 1.8411 eV 673.41 nm f= 0.0022
<S^2> = 2.227
112A → 122A -0.24300
113A → 122A 0.42187
114A → 122A 0.31957
116A → 122A 0.10070
112B → 121B 0.77388
117B → 122B -0.15590

Excited State 19: 2.961-A 1.8456 eV 671.79 nm f= 0.0277
<S^2> = 1.941
111A → 122A -0.24850
115A → 122A -0.10858
116A → 122A 0.38284
120A → 122A 0.16898
111B → 121B 0.59837
112B → 121B -0.21755
113B → 121B 0.29540
114B → 121B 0.20434
115B → 121B 0.16199
117B → 122B -0.39156

Excited State 20: 2.717-A 1.8634 eV 665.37 nm f= 0.0002
<S^2> = 1.595
113A → 122A -0.20796
114A → 122A 0.90840
111B → 121B -0.13656
112B → 121B -0.24077
114B → 121B -0.13463

Excited State 21: 2.588-A 1.9030 eV 651.51 nm f= 0.0003
<S^2> = 1.424
111A → 122A 0.10390
112A → 122A 0.27346
113A → 122A 0.76563
111B → 121B -0.17906
112B → 121B -0.38526
113B → 121B -0.11916
115B → 122B 0.12482
116B → 122B -0.31407

Excited State 22: 2.507-A 1.9136 eV 647.91 nm f= 0.0011
<S^2> = 1.321
113A → 122A 0.31521
115B → 122B -0.37255
116B → 122B 0.85614

Excited State 23: 2.549-A 1.9473 eV 636.71 nm f= 0.0007
<S^2> = 1.375
111A → 122A -0.12004
112A → 122A 0.91083
113A → 122A -0.12527
112B → 121B 0.33740

Excited State 24: 2.662-A 1.9819 eV 625.57 nm f= 0.0569
<S^2> = 1.522
111A → 122A 0.52344
113A → 122A -0.20124
114A → 122A -0.15732
115A → 122A -0.17433
116A → 122A 0.35683
120A → 122A 0.10054
111B → 121B -0.39604
117B → 122B -0.51482
120B → 121B -0.13330

Excited State 25: 2.493-A 2.0725 eV 598.23 nm f= 0.0391
<S^2> = 1.304
115A → 122A -0.24858
116A → 122A 0.22218
110B → 121B 0.13963
111B → 122B -0.30147

114B → 122B 0.16339
115B → 121B 0.22462
115B → 122B 0.63014
116B → 122B 0.26059
117B → 121B 0.32987
117B → 122B 0.29450

Excited State 26: 2.482-A 2.0932 eV 592.32 nm f = 0.0027
$<S^2>$ = 1.291
114B → 122B 0.97757

Excited State 27: 2.521-A 2.1291 eV 582.33 nm f = 0.0003
$<S^2>$ = 1.339
109B → 121B 0.11329
113B → 122B 0.98661

Excited State 28: 2.879-A 2.1556 eV 575.16 nm f = 0.0111
$<S^2>$ = 1.822
110A → 122A 0.20144
111A → 122A -0.10784
107B → 121B -0.19929
108B → 121B -0.11055
109B → 121B 0.83188
110B → 121B -0.26820
111B → 122B -0.24519
113B → 122B -0.12932

Excited State 29: 2.506-A 2.1926 eV 565.47 nm f = 0.0033
$<S^2>$ = 1.320
116A → 122A 0.23151
110B → 121B 0.22919
112B → 122B 0.86474
115B → 121B 0.14269
115B → 122B -0.18340
117B → 122B 0.23977

Excited State 30: 2.627-A 2.2070 eV 561.77 nm f = 0.0072
$<S^2>$ = 1.476
111A → 122A 0.18071
116A → 122A 0.22843
109B → 121B 0.24612
110B → 121B 0.59256
111B → 122B 0.33059
112B → 122B -0.44677
115B → 121B 0.15286
115B → 122B -0.25440
116B → 122B -0.10180
117B → 122B 0.25403

Excited State 31: 2.717-A 2.2249 eV 557.27 nm f = 0.0047
$<S^2>$ = 1.596
109A → 122A 0.14860
116A → 122A -0.40148
107B → 121B 0.12448
108B → 121B -0.28193
109B → 121B 0.22649
110B → 121B 0.55152
111B → 121B 0.16831
112B → 122B 0.18456
115B → 121B -0.22593
115B → 122B 0.25025
117B → 122B -0.35207

Excited State 32: 2.943-A 2.2454 eV 552.17 nm f = 0.0012
$<S^2>$ = 1.915
106A → 122A 0.10892
109A → 122A -0.13847
110A → 122A 0.11916
111A → 122A 0.13745
116A → 122A -0.14842
107B → 121B 0.27005
108B → 121B 0.85366
109B → 121B 0.20365
111B → 121B 0.14388

Excited State 33: 2.915-A 2.2758 eV 544.79 nm f = 0.0007
$<S^2>$ = 1.875
107A → 122A -0.11696
110A → 122A 0.26151
111A → 122A 0.27439
107B → 121B 0.69752
108B → 121B -0.34563
109B → 122B -0.12635
110B → 121B -0.25799
111B → 121B 0.19061
115B → 121B 0.12369
117B → 122B 0.16365

Excited State 34: 2.687-A 2.3092 eV 536.92 nm f = 0.0053
$<S^2>$ = 1.555
107A → 122A 0.14403
109A → 122A -0.25023
110A → 122A 0.20223
111A → 122A 0.19006
116A → 122A -0.16098
106B → 121B 0.16339
107B → 121B -0.36274
109B → 122B 0.16142
110B → 121B -0.16935
111B → 121B 0.20695
111B → 122B 0.64618
115B → 122B 0.23211
117B → 121B 0.10990

Excited State 35: 3.053-A 2.3577 eV 525.87 nm f = 0.0051
$<S^2>$ = 2.080
105A → 122A -0.20352
107A → 122A -0.18985
108A → 122A -0.16518
109A → 122A 0.55125
110A → 122A 0.45320
111A → 122A -0.11601

103B → 121B 0.19610
104B → 121B -0.36286
105B → 121B -0.22032
106B → 121B -0.22663
108B → 121B 0.11547
111B → 122B 0.18333

Excited State 36: 3.015-A 2.3861 eV 519.60 nm f = 0.0048

$<S^2>$ = 2.023

105A → 122A 0.43419
106A → 122A 0.26810
109A → 122A 0.31665
110A → 122A -0.41911
103B → 121B 0.34181
104B → 121B 0.12233
105B → 121B -0.34697
106B → 121B 0.13877
107B → 121B 0.21656
109B → 121B 0.15311
110B → 121B -0.16354
111B → 122B 0.19941

Excited State 37: 2.842-A 2.4216 eV 511.99 nm f = 0.0036

$<S^2>$ = 1.769

105A → 122A 0.15565
106A → 122A -0.20910
108A → 122A 0.38560
110A → 122A 0.27203
111A → 122A -0.30219
116A → 122A 0.10285
103B → 121B -0.15925
105B → 121B -0.10812
106B → 121B 0.61644
107B → 121B 0.14634
108B → 122B 0.13986
109B → 122B -0.10779
111B → 121B -0.18546
111B → 122B 0.15281

Excited State 38: 2.821-A 2.4446 eV 507.18 nm f = 0.0010

$<S^2>$ = 1.740

103A → 122A 0.12834
105A → 122A -0.35568
108A → 122A -0.39267
111A → 122A 0.23333
104B → 121B -0.13297
105B → 121B -0.39853
106B → 121B 0.54890
110B → 122B -0.14314
111B → 121B 0.10917
111B → 122B -0.23549

Excited State 39: 2.838-A 2.4649 eV 502.99 nm f = 0.0019

$<S^2>$ = 1.763

102A → 122A 0.10889
104A → 122A -0.18681
106A → 122A -0.22943
107A → 122A 0.20697
109A → 122A 0.42031
110A → 122A -0.15718
111A → 122A 0.11903
103B → 121B 0.18109
104B → 121B -0.14111
105B → 121B 0.62170
106B → 121B 0.21232
108B → 121B 0.10969
108B → 122B 0.12528
109B → 122B 0.16759
110B → 121B -0.14415
110B → 122B -0.17537

Excited State 40: 2.782-A 2.4771 eV 500.52 nm f = 0.0040

$<S^2>$ = 1.684

105A → 122A 0.38125
107A → 122A 0.35999
108A → 122A -0.26366
110A → 122A 0.50469
111A → 122A 0.12671
103B → 121B 0.20980
104B → 121B 0.39385
107B → 121B -0.12476
107B → 122B -0.10413
109B → 121B -0.16749
109B → 122B -0.12039
110B → 122B -0.21755
111B → 122B -0.16468

Excited State 41: 2.762-A 2.4882 eV 498.29 nm f = 0.0014

$<S^2>$ = 1.657

104A → 122A -0.13197
106A → 122A 0.44447
107A → 122A -0.32604
108A → 122A -0.12360
103B → 121B 0.27588
105B → 121B 0.37539
106B → 121B 0.33308
108B → 122B -0.34189
109B → 122B -0.29310
110B → 122B 0.25815

Excited State 42: 2.561-A 2.5209 eV 491.83 nm f = 0.0014

$<S^2>$ = 1.390

105A → 122A -0.30166
106A → 122A 0.44386
107A → 122A 0.45686
108A → 122A 0.43319
109A → 122A 0.15154

110A → 122A 0.16936
104B → 121B 0.11412
107B → 122B 0.10431
109B → 121B -0.10362
109B → 122B 0.23330
110B → 122B 0.30931
111B → 122B -0.16279

Excited State 43: 2.527-A 2.5514 eV 485.94 nm f= 0.0084
$\langle S^2 \rangle$ = 1.346
106A → 122A 0.13562
107A → 122A -0.24816
108A → 122A -0.14256
109A → 122A -0.11646
110A → 122A 0.16454
111A → 122A -0.13081
104B → 121B 0.10934
104B → 122B 0.11572
107B → 121B 0.22241
108B → 122B -0.10685
109B → 122B 0.79125
110B → 122B -0.19777

Excited State 44: 2.594-A 2.5682 eV 482.76 nm f= 0.0024
$\langle S^2 \rangle$ = 1.432
104A → 122A 0.21948
105A → 122A 0.11272
106A → 122A 0.25091
107A → 122A 0.46356
108A → 122A -0.21578
111A → 122A -0.20360
101B → 121B 0.12904
102B → 121B 0.10483
103B → 121B -0.22242
104B → 121B -0.35946
104B → 122B 0.11500
105B → 121B 0.14805
107B → 121B 0.22016
107B → 122B 0.30073
108B → 122B -0.14228
109B → 122B -0.19477
110B → 122B -0.24773
111B → 121B -0.13936

Excited State 45: 2.666-A 2.5757 eV 481.37 nm f= 0.0009
$\langle S^2 \rangle$ = 1.527
104A → 122A 0.10924
106A → 122A -0.21680
107A → 122A 0.14082
108A → 122A -0.43143
111A → 122A -0.12200
101B → 121B 0.10854
104B → 121B 0.15159
107B → 121B 0.10609
107B → 122B 0.10436
108B → 122B 0.31512
110B → 122B 0.69844

Excited State 46: 2.846-A 2.6033 eV 476.25 nm f= 0.0001
$\langle S^2 \rangle$ = 1.775
104A → 122A 0.54744
105A → 122A -0.25863
109A → 122A 0.24455
101B → 121B 0.30087
102B → 121B 0.26457
103B → 121B -0.11983
104B → 121B 0.32864
105B → 121B 0.13317
107B → 122B -0.30045
108B → 122B -0.37776

Excited State 47: 2.690-A 2.6232 eV 472.65 nm f= 0.0003
$\langle S^2 \rangle$ = 1.559
102A → 122A 0.10844
104A → 122A 0.43974
106A → 122A 0.18626
107A → 122A -0.13560
108A → 122A 0.17307
109A → 122A -0.21814
111A → 122A 0.13951
101B → 121B 0.29311
102B → 121B 0.15071
103B → 121B 0.32895
104B → 121B -0.16771
108B → 122B 0.56489
110B → 122B -0.11100

Excited State 48: 2.429-A 2.6468 eV 468.43 nm f= 0.0010
$\langle S^2 \rangle$ = 1.225
105A → 122A -0.35024
106A → 122A 0.12658
107A → 122A -0.11376
104B → 121B 0.55032
107B → 122B 0.49771
108B → 122B 0.19872
109B → 121B 0.11797
109B → 122B -0.23388
110B → 122B -0.26767
111B → 122B 0.15891

Excited State 49: 2.374-A 2.6601 eV 466.08 nm f= 0.0010
$\langle S^2 \rangle$ = 1.159
104A → 122A 0.12851
106A → 122A -0.41160
107A → 122A 0.13226
108A → 122A 0.23427
109A → 122A -0.21960
103B → 121B 0.60648
105B → 121B -0.10358

105B → 122B -0.11229
107B → 122B 0.29941
108B → 122B -0.39118
Excited State 50: 2.427-A 2.6659 eV 465.07 nm f= 0.0002
<S^2> = 1.223
102A → 122A -0.13089
104A → 122A 0.14883
105A → 122A 0.34881
107A → 122A -0.26196
109A → 122A 0.24321
111A → 122A 0.18657
103B → 121B -0.27041
104B → 122B -0.15665
106B → 122B -0.14399
107B → 121B -0.13012
107B → 122B 0.61035
108B → 122B -0.14255
110B → 122B 0.15298
111B → 121B 0.12445
111B → 122B -0.12491
Excited State 51: 2.749-A 2.7222 eV 455.45 nm f= 0.0006
<S^2> = 1.640
102A → 122A -0.41882
103A → 122A 0.48649
103B → 121B 0.11375
105B → 122B 0.68027
106B → 122B -0.19444
Excited State 52: 2.430-A 2.7488 eV 451.05 nm f= 0.0008
<S^2> = 1.227
102A → 122A 0.19587
103A → 122A 0.33634
106A → 122A -0.11918
105B → 122B 0.16461
106B → 122B 0.87938
Excited State 53: 2.921-A 2.8422 eV 436.22 nm f= 0.0033
<S^2> = 1.883
101A → 122A 0.15083
104A → 122A -0.12830
101B → 121B -0.41218
102B → 121B 0.86703
Excited State 54: 2.663-A 2.8650 eV 432.75 nm f= 0.0012
<S^2> = 1.522
102A → 122A 0.41353
103A → 122A 0.67838
104A → 122A -0.10128
101B → 121B 0.12072
103B → 122B 0.24286
104B → 122B -0.24305
105B → 122B -0.32198
106B → 121B -0.14461
106B → 122B -0.27657
Excited State 55: 2.552-A 2.9032 eV 427.07 nm f= 0.0003
<S^2> = 1.378
102A → 122A -0.26267
103A → 122A -0.19915
103B → 122B 0.80683
104B → 122B -0.42122
106B → 122B 0.15079
Excited State 56: 2.459-A 2.9219 eV 424.33 nm f= 0.0117
<S^2> = 1.261
102A → 122A -0.13680
103A → 122A 0.13677
105A → 122A 0.11152
111A → 122A 0.14497
103B → 122B 0.40250
104B → 122B 0.79973
105B → 122B -0.22779
Excited State 57: 3.059-A 2.9716 eV 417.23 nm f= 0.0004
<S^2> = 2.089
100A → 122A 0.24876
101A → 122A 0.26153
100B → 121B 0.91437
Excited State 58: 2.418-A 3.0290 eV 409.32 nm f= 0.0014
<S^2> = 1.212
101A → 122A -0.18170
102A → 122A 0.55687
103A → 122A -0.27398
104A → 122A -0.24708
101B → 121B 0.22236
102B → 122B -0.11844
103B → 122B 0.28418
104B → 122B 0.12478
105B → 121B -0.15130
105B → 122B 0.51988
106B → 122B -0.12554
Excited State 59: 2.439-A 3.0751 eV 403.18 nm f= 0.0007
<S^2> = 1.237
101A → 122A -0.23332
102A → 122A -0.34713
104A → 122A -0.42876
101B → 121B 0.64473
101B → 122B 0.17358
102B → 121B 0.27797
102B → 122B 0.22058
105B → 122B -0.12231
Excited State 60: 2.962-A 3.1026 eV 399.61 nm f= 0.0001
<S^2> = 1.943
99A → 122A -0.16823
100A → 122A 0.34063
101A → 122A 0.65283

104A → 122A -0.13968
95B → 121B -0.10444
96B → 121B 0.30194
97B → 121B 0.32865
100B → 121B -0.32336
101B → 121B 0.22812

Excited State 61: 2.460-A 3.1538 eV 393.13 nm f = 0.0002
$\langle S^2 \rangle$ = 1.263
99A → 122A 0.17930
100A → 122A -0.13995
101A → 122A 0.11378
102A → 122A 0.15414
96B → 121B -0.10168
98B → 121B 0.12080
101B → 122B -0.11278
102B → 122B 0.90920

Excited State 62: 2.688-A 3.1715 eV 390.94 nm f = 0.0002
$\langle S^2 \rangle$ = 1.556
99A → 122A 0.11808
100A → 122A 0.73464
101A → 122A -0.42299
100B → 121B -0.12858
100B → 122B -0.39341
101B → 121B -0.18294
101B → 122B -0.11809
102B → 122B 0.11872

Excited State 63: 3.029-A 3.2399 eV 382.68 nm f = 0.0018
$\langle S^2 \rangle$ = 2.044
98A → 122A 0.28976
99A → 122A -0.21402
95B → 121B -0.18866
98B → 121B 0.83198
99B → 121B -0.28820
100B → 122B 0.11519

Excited State 64: 2.853-A 3.2537 eV 381.06 nm f = 0.0004
$\langle S^2 \rangle$ = 1.785
97A → 122A 0.13904
98B → 121B 0.25480
99B → 121B 0.86217
101B → 122B -0.36797

Excited State 65: 2.556-A 3.2570 eV 380.66 nm f = 0.0005
$\langle S^2 \rangle$ = 1.383
99A → 122A -0.22852
100A → 122A 0.10369
101A → 122A -0.17544
104A → 122A 0.13993
96B → 121B 0.10499
98B → 121B 0.11054
99B → 121B 0.31736
100B → 122B 0.15408
101B → 121B -0.12727
101B → 122B 0.81984
102B → 122B 0.14613

Excited State 66: 2.798-A 3.2867 eV 377.22 nm f = 0.0019
$\langle S^2 \rangle$ = 1.707
98A → 122A -0.14536
99A → 122A -0.48181
101A → 122A -0.29346
96B → 121B 0.19207
97B → 121B 0.23327
98B → 121B -0.19659
99B → 121B -0.13274
100B → 121B 0.10174
100B → 122B 0.57485
101B → 122B -0.33200
102B → 122B 0.19260

Excited State 67: 2.940-A 3.3348 eV 371.79 nm f = 0.0008
$\langle S^2 \rangle$ = 1.911
95A → 122A 0.10297
98A → 122A 0.28605
100A → 122A -0.18699
94B → 121B -0.13901
95B → 121B 0.44131
96B → 121B -0.22405
97B → 121B 0.72884
100B → 122B -0.14635

Excited State 68: 2.981-A 3.3507 eV 370.03 nm f = 0.0003
$\langle S^2 \rangle$ = 1.971
94A → 122A -0.15887
96A → 122A 0.10248
98A → 122A 0.19674
94B → 121B -0.11073
95B → 121B 0.60371
96B → 121B 0.60626
97B → 121B -0.32773
98B → 122B -0.14416

Excited State 69: 2.945-A 3.4248 eV 362.01 nm f = 0.0018
$\langle S^2 \rangle$ = 1.918
93A → 122A 0.40457
96A → 122A -0.12868
97A → 122A -0.27832
98A → 122A -0.32618
99A → 122A 0.21064
100A → 122A 0.16593
94B → 121B 0.41732
95B → 121B 0.44157
95B → 122B -0.13462
96B → 121B -0.16643
98B → 121B 0.14190
100B → 122B 0.26820

Excited State 70: 2.648-A 3.4320 eV 361.26 nm f = 0.0020
$\langle S^2 \rangle$ = 1.502

93A → 122A -0.25971
96A → 122A -0.15723
98A → 122A 0.41601
99A → 122A 0.33209
100A → 122A 0.32566
101A → 122A 0.11486
93B → 121B -0.15210
94B → 121B -0.25939
96B → 121B -0.20683
97B → 121B -0.11055
98B → 121B -0.11394
100B → 122B 0.54423

Excited State 71: 3.013-A 3.4669 eV 357.62 nm f = 0.0008
$\langle S^2 \rangle$ = 2.019
95A → 122A 0.22647
96A → 122A 0.36618
97A → 122A 0.66153
98A → 122A -0.13569
99A → 122A 0.11369
100A → 122A 0.10988
93B → 121B 0.46536
95B → 121B 0.11181
96B → 121B -0.15737
99B → 121B -0.10516
100B → 122B 0.18224

Excited State 72: 2.525-A 3.5122 eV 353.01 nm f = 0.0001
$\langle S^2 \rangle$ = 1.344
93A → 122A 0.21864
94A → 122A 0.15038
98A → 122A 0.30118
99A → 122A 0.48167
100A → 122A -0.15688
101A → 122A -0.18264
94B → 121B 0.30533
95B → 121B -0.27577
96B → 121B 0.45232
97B → 121B 0.29328
97B → 122B 0.10785
100B → 122B 0.16110

Excited State 73: 2.671-A 3.5173 eV 352.49 nm f = 0.0093
$\langle S^2 \rangle$ = 1.533
92A → 122A 0.16369
93A → 122A 0.11252
95A → 122A -0.18693
96A → 122A -0.28545
97A → 122A 0.39320
98A → 122A 0.34814
99A → 122A -0.30797
93B → 121B -0.10639
94B → 121B 0.47170
95B → 122B -0.14612
96B → 121B -0.18923
96B → 122B -0.14995
97B → 121B -0.13040
98B → 121B -0.23583
98B → 122B -0.15543
99B → 122B 0.17938

Excited State 74: 2.713-A 3.5222 eV 352.00 nm f = 0.0094
$\langle S^2 \rangle$ = 1.590
93A → 122A 0.13296
95A → 122A 0.15559
96A → 122A 0.55398
97A → 122A -0.43229
98A → 122A 0.36753
99A → 122A -0.19561
92B → 121B 0.11980
93B → 121B 0.25723
94B → 121B 0.10680
96B → 121B -0.18082
97B → 121B -0.14669
97B → 122B -0.10544
98B → 121B -0.15014
98B → 122B -0.19255
99B → 121B 0.13644
99B → 122B -0.10672

Excited State 75: 2.814-A 3.5593 eV 348.34 nm f = 0.0001
$\langle S^2 \rangle$ = 1.730
92A → 122A -0.26872
93A → 122A 0.45602
94A → 122A 0.36024
95A → 122A -0.32369
97A → 122A 0.13954
92B → 121B 0.28695
93B → 121B -0.14668
94B → 121B -0.31997
95B → 122B 0.21729
96B → 121B -0.12053
97B → 122B 0.28213
98B → 122B -0.10523
99B → 122B -0.27716

Excited State 76: 2.710-A 3.5690 eV 347.39 nm f = 0.0017
$\langle S^2 \rangle$ = 1.586
92A → 122A -0.15356
93A → 122A 0.24949
94A → 122A 0.21316
95A → 122A 0.54272
96A → 122A -0.31176
92B → 121B 0.22036
94B → 121B -0.19941
95B → 122B 0.13702

96B → 122B 0.17933
97B → 121B -0.13472
97B → 122B -0.24917
99B → 122B 0.45714

Excited State 77: 2.578-A 3.6008 eV 344.33 nm f= 0.0023
$<S^2>$ = 1.411
94A → 122A -0.18740
95A → 122A -0.27636
96A → 122A 0.21521
97A → 122A -0.13100
98A → 122A -0.14601
94B → 121B -0.18304
94B → 122B -0.15704
95B → 121B -0.13285
96B → 121B -0.10136
97B → 122B 0.26839
98B → 122B -0.32105
99B → 122B 0.71321

Excited State 78: 2.531-A 3.6214 eV 342.36 nm f= 0.0068
$<S^2>$ = 1.351
94A → 122A 0.22958
95A → 122A -0.18408
96A → 122A 0.19095
98A → 122A 0.10569
94B → 122B 0.19632
95B → 121B 0.17617
95B → 122B 0.11865
96B → 122B -0.16502
97B → 122B 0.13298
98B → 122B 0.75561
99B → 122B 0.34243

Excited State 79: 2.681-A 3.6646 eV 338.33 nm f= 0.0031
$<S^2>$ = 1.547
93A → 122A -0.43694
94A → 122A 0.73367
98A → 122A -0.10611
92B → 121B -0.14503
94B → 121B 0.13801
95B → 121B 0.14916
96B → 122B 0.14103
98B → 122B -0.33937
99B → 122B 0.10410

Excited State 80: 2.469-A 3.6925 eV 335.77 nm f= 0.0007
$<S^2>$ = 1.274
95A → 122A 0.29929
99A → 122A -0.11290
94B → 121B 0.17301
95B → 122B 0.10030
96B → 122B 0.44055
97B → 122B 0.77690

Excited State 81: 2.382-A 3.7190 eV 333.38 nm f= 0.0002
$<S^2>$ = 1.168
94A → 122A 0.15220
96A → 122A -0.41669
97A → 122A -0.22732
93B → 121B 0.60397
94B → 121B -0.14749
95B → 122B -0.14400
96B → 122B -0.47957
97B → 122B 0.26308

Excited State 82: 2.420-A 3.7280 eV 332.57 nm f= 0.0015
$<S^2>$ = 1.214
94A → 122A -0.22773
95A → 122A -0.22869
96A → 122A -0.11361
92B → 121B 0.10026
93B → 121B 0.35291
94B → 121B 0.26289
95B → 122B 0.76799
97B → 122B -0.13796
98B → 122B -0.10824

Excited State 83: 2.412-A 3.7551 eV 330.18 nm f= 0.0021
$<S^2>$ = 1.204
92A → 122A 0.11264
93A → 122A 0.11105
95A → 122A -0.44469
96A → 122A -0.14116
93B → 121B 0.36699
94B → 121B -0.15596
95B → 122B -0.26828
96B → 122B 0.66985
97B → 122B -0.17379
98B → 122B 0.11927

Excited State 84: 2.811-A 3.7674 eV 329.09 nm f= 0.0006
$<S^2>$ = 1.725
92A → 122A -0.41485
93A → 122A -0.39100
92B → 121B 0.69015
93B → 121B 0.11041
94B → 121B 0.13878
95B → 122B -0.34880

Excited State 85: 2.524-A 3.8584 eV 321.34 nm f= 0.0121
$<S^2>$ = 1.343
92A → 122A -0.44088
91B → 121B -0.20192
92B → 121B -0.42390
92B → 122B -0.12712
94B → 122B 0.70262
95B → 122B -0.13571

Excited State 86: 2.828-A 3.9398 eV 314.69 nm f= 0.0188
$<S^2>$ = 1.749

91A → 122A 0.27393
92A → 122A 0.34331
90B → 121B -0.46018
91B → 121B 0.47414
92B → 121B 0.26330
94B → 122B 0.47458
98B → 122B -0.11483

Excited State 87: 2.486-A 3.9675 eV 312.50 nm f= 0.0001
$\langle S^2 \rangle$ = 1.295
93B → 122B 0.98994

Excited State 88: 2.924-A 3.9854 eV 311.10 nm f= 0.0095
$\langle S^2 \rangle$ = 1.888
91A → 122A 0.53870
92A → 122A -0.37127
90B → 121B 0.11262
91B → 121B 0.58235
91B → 122B -0.13565
92B → 121B -0.18905
92B → 122B -0.17888
94B → 122B -0.29087

Excited State 89: 2.516-A 4.0447 eV 306.53 nm f= 0.0010
$\langle S^2 \rangle$ = 1.333
121A → 123A 0.97347
121A → 124A -0.16659

Excited State 90: 2.796-A 4.0821 eV 303.73 nm f= 0.0156
$\langle S^2 \rangle$ = 1.704
90A → 122A -0.32626
92A → 122A -0.13255
121A → 124A 0.65606
90B → 121B -0.51856
91B → 121B -0.16071
91B → 122B -0.10250
92B → 122B -0.26180
94B → 122B -0.16096

Excited State 91: 2.774-A 4.0846 eV 303.54 nm f= 0.0111
$\langle S^2 \rangle$ = 1.674
90A → 122A 0.30332
92A → 122A 0.10322
121A → 123A 0.14240
121A → 124A 0.72627
90B → 121B 0.52188
91B → 121B 0.12706
92B → 122B 0.16300
94B → 122B 0.10339

Excited State 92: 2.527-A 4.1263 eV 300.48 nm f= 0.0004
$\langle S^2 \rangle$ = 1.347
121A → 125A 0.99095

Excited State 93: 2.507-A 4.1438 eV 299.20 nm f= 0.0073
$\langle S^2 \rangle$ = 1.322
90A → 122A -0.45062
91A → 122A 0.13315
92A → 122A -0.18646
91B → 122B -0.17415
92B → 122B 0.82583

Excited State 94: 2.450-A 4.2985 eV 288.43 nm f= 0.0501
$\langle S^2 \rangle$ = 1.251
91A → 122A 0.64824
92A → 122A 0.16337
90B → 121B 0.21300
90B → 122B 0.48995
91B → 121B -0.39920
91B → 122B 0.20431

Excited State 95: 2.519-A 4.3378 eV 285.82 nm f= 0.0187
$\langle S^2 \rangle$ = 1.337
90A → 122A -0.60477
91A → 122A -0.11577
90B → 121B 0.22214
90B → 122B -0.11981
91B → 121B 0.24506
91B → 122B 0.64834
92B → 122B -0.16730
120B → 123B -0.10988

Excited State 96: 2.971-A 4.3901 eV 282.42 nm f= 0.0006
$\langle S^2 \rangle$ = 1.957
120A → 123A -0.12104
91B → 122B 0.10582
120B → 123B 0.95972
120B → 124B -0.17547

Excited State 97: 2.962-A 4.4353 eV 279.54 nm f= 0.0052
$\langle S^2 \rangle$ = 1.943
120A → 124A -0.10115
91B → 122B 0.17928
120B → 123B 0.14578
120B → 124B 0.95199

Excited State 98: 2.606-A 4.4697 eV 277.39 nm f= 0.1518
$\langle S^2 \rangle$ = 1.448
90A → 122A 0.37515
91A → 122A 0.18457
92A → 122A -0.11898
90B → 121B -0.26197
90B → 122B -0.33766
91B → 121B -0.18825
91B → 122B 0.54558
92B → 122B 0.24676
94B → 122B -0.15098
98B → 122B 0.12915
118B → 125B -0.15518

119B → 125B -0.11224
120B → 124B -0.17477
Excited State 99: 2.982-A 4.4873 eV 276.30 nm *f* = 0.0012
<S^2> = 1.973
120A → 125A -0.10915
120B → 125B 0.97946
Excited State 100: 2.741-A 4.5151 eV 274.60 nm *f* = 0.1281
<S^2> = 1.628
90A → 122A 0.21114
91A → 122A -0.24858
92A → 122A -0.23415
98A → 122A 0.12324
117A → 123A -0.12225
117A → 124A 0.12760
120A → 123A 0.17470
121A → 126A -0.29999
90B → 121B -0.16358
90B → 122B 0.55281
91B → 121B 0.13174
91B → 122B 0.27701
92B → 122B 0.13100
118B → 123B 0.16605
118B → 124B -0.12368
118B → 125B 0.14391

Excitation energies and oscillator strengths (*f*) calculated for $[W(CN)_8]^{4-}$.

Excited State 1: Singlet-A" 2.9100 eV 426.06 nm *f* = 0.0003
<S^2> = 0.000
61 → 62 0.70509

Excited State 2: Singlet-A' 3.5020 eV 354.03 nm *f* = 0.0066
<S^2> = 0.000
61 → 63 0.70256

Excited State 3: Singlet-A" 3.5270 eV 351.53 nm *f* = 0.0076
<S^2> = 0.000
61 → 64 0.70161

Excited State 4: Singlet-A" 4.2306 eV 293.07 nm *f* = 0.0001
<S^2> = 0.000
61 → 65 0.67308
61 → 66 0.21264

Excited State 5: Singlet-A" 4.2677 eV 290.52 nm *f* = 0.0005
<S^2> = 0.000
61 → 65 -0.21357
61 → 66 0.67137

Excited State 6: Singlet-A' 5.1807 eV 239.32 nm *f* = 0.1105
<S^2> = 0.000
61 → 67 0.69309

Excited State 7: Singlet-A' 5.3492 eV 231.78 nm *f* = 0.1519
<S^2> = 0.000
61 → 68 0.68154

Excited State 8: Singlet-A" 5.3692 eV 230.92 nm *f* = 0.1498
<S^2> = 0.000
61 → 69 0.68278

Excited State 9: Singlet-A" 5.8437 eV 212.17 nm *f* = 0.0001
<S^2> = 0.000
61 → 71 0.16440
61 → 73 -0.44769
61 → 75 0.48395
61 → 89 0.16082

Excited State 10: Singlet-A" 5.9361 eV 208.86 nm *f* = 0.0009
<S^2> = 0.000
61 → 70 0.26755
61 → 73 0.45683
61 → 75 0.43924

Excited State 11: Singlet-A" 5.9726 eV 207.59 nm *f* = 0.0020
<S^2> = 0.000
61 → 70 -0.41323
61 → 71 0.12657
61 → 73 0.24020
61 → 75 0.10381
61 → 76 0.47455

Excited State 12: Singlet-A' 5.9916 eV 206.93 nm *f* = 0.0027
<S^2> = 0.000
61 → 72 -0.17052
61 → 74 0.60619
61 → 77 0.26527

Excited State 13: Singlet-A" 6.0550 eV 204.76 nm *f* = 0.0013
<S^2> = 0.000
61 → 70 0.49976
61 → 71 0.17600
61 → 75 -0.14457
61 → 76 0.42725

Excited State 14: Singlet-A" 6.1763 eV 200.74 nm *f* = 0.0340
<S^2> = 0.000
61 → 71 0.64531
61 → 73 0.11723
61 → 76 -0.22842

Excited State 15: Singlet-A' 6.1904 eV 200.28 nm *f* = 0.0342
<S^2> = 0.000
61 → 72 0.68156
61 → 74 0.16056

Excited State 16: Singlet-A' 6.3712 eV 194.60 nm *f* = 0.0062
<S^2> = 0.000
59 → 62 -0.18439
60 → 62 0.59209
61 → 78 0.28052
61 → 81 -0.10302

Excited State 17: Singlet-A' 6.4186 eV 193.16 nm *f* = 0.0081
<S^2> = 0.000
59 → 62 0.60968
60 → 62 0.23711
61 → 78 -0.13652
61 → 81 -0.15474

Excited State 18: Singlet-A" 6.4332 eV 192.73 nm *f* = 0.0000
<S^2> = 0.000
57 → 62 -0.16453
58 → 62 0.62556
61 → 79 -0.18531
61 → 80 0.11578

Excited State 19: Singlet-A" 6.4610 eV 191.90 nm *f* = 0.0000
<S^2> = 0.000
57 → 62 0.23775
58 → 62 -0.10721
61 → 79 -0.29986
61 → 80 0.54062

Excited State 20: Singlet-A' 6.4729 eV 191.54 nm *f* = 0.0012
<S^2> = 0.000
55 → 62 -0.11191
56 → 62 0.32742
59 → 62 0.14161
60 → 62 -0.19127
60 → 64 0.10521
61 → 78 0.52846

Excited State 21: Singlet-A" 6.4800 eV 191.33 nm *f* = 0.0012
<S^2> = 0.000

57 → 62 -0.32913
61 → 79 0.46177
61 → 80 0.38503

Excited State 22: Singlet-A' 6.5778 eV 188.49 nm f = 0.0112
$<S^2>$ = 0.000
55 → 62 0.39920
56 → 62 0.45280
58 → 63 0.12544
59 → 64 -0.11086
61 → 77 0.15546
61 → 78 -0.20186

Excited State 23: Singlet-A" 6.6141 eV 187.46 nm f = 0.0104
$<S^2>$ = 0.000
51 → 62 0.15090
52 → 62 0.10418
54 → 62 0.45621
57 → 62 0.30504
61 → 79 0.20389
61 → 82 0.27113

Excited State 24: Singlet-A' 6.6276 eV 187.07 nm f = 0.0011
$<S^2>$ = 0.000
53 → 62 0.17031
55 → 62 0.42115
56 → 62 -0.35584
59 → 62 0.16483
61 → 77 0.21976
61 → 78 0.21004

Excited State 25: Singlet-A" 6.6322 eV 186.94 nm f = 0.0004
$<S^2>$ = 0.000
54 → 62 -0.36466
57 → 62 0.39691
58 → 62 0.20524
60 → 63 0.14278
61 → 79 0.29251
61 → 82 -0.16168

Excited State 26: Singlet-A' 6.6705 eV 185.87 nm f = 0.0047
$<S^2>$ = 0.000
53 → 62 -0.19797
55 → 62 -0.19276
61 → 74 -0.23608
61 → 77 0.52801
61 → 83 -0.13840

Excited State 27: Singlet-A" 6.7109 eV 184.75 nm f = 0.0031
$<S^2>$ = 0.000
52 → 62 0.26789
54 → 62 -0.18869
59 → 63 0.39480
60 → 63 -0.35818
61 → 80 -0.11784
61 → 82 0.22860

Excited State 28: Singlet-A" 6.7511 eV 183.65 nm f = 0.0027
$<S^2>$ = 0.000
51 → 62 -0.13103
52 → 62 0.58598
58 → 64 0.13340
59 → 63 -0.14940
60 → 63 0.14948
61 → 82 -0.13407

Excited State 29: Singlet-A" 6.7584 eV 183.45 nm f = 0.0226
$<S^2>$ = 0.000
51 → 62 0.26656
54 → 62 -0.24583
55 → 63 -0.10019
57 → 62 -0.10472
59 → 63 -0.19688
60 → 63 0.29260
61 → 82 0.43416

Excited State 30: Singlet-A' 6.7876 eV 182.66 nm f = 0.0397
$<S^2>$ = 0.000
52 → 63 0.10573
53 → 62 0.56462
53 → 64 -0.10871
55 → 62 -0.11588
56 → 62 0.13936
58 → 63 -0.15448
61 → 77 0.11238

Excited State 31: Singlet-A" 6.8114 eV 182.03 nm f = 0.0150
$<S^2>$ = 0.000
51 → 62 0.53729
56 → 63 -0.11223
57 → 62 -0.10090
58 → 64 0.17561
61 → 82 -0.31713

Excited State 32: Singlet-A" 6.8496 eV 181.01 nm f = 0.0021
$<S^2>$ = 0.000
51 → 62 -0.14141
52 → 62 -0.10698
57 → 64 0.18280
58 → 64 0.60509
61 → 82 0.14902

Excited State 33: Singlet-A' 6.8635 eV 180.64 nm f = 0.0009
$<S^2>$ = 0.000
56 → 64 -0.22352
57 → 63 0.18256
58 → 63 -0.13565
59 → 64 -0.13649
60 → 62 0.11706
60 → 64 0.17582
61 → 81 0.54827

Excited State 34: Singlet-A' 6.9302 eV 178.90 nm f = 0.0075
$<S^2>$ = 0.000

54 → 63 0.11802
55 → 62 -0.10515
55 → 64 -0.15091
58 → 63 0.17879
59 → 64 0.15656
60 → 64 0.55191

Excited State 35: Singlet-A" 6.9476 eV 178.46 nm f = 0.0098
$\langle S^2 \rangle$ = 0.000
55 → 63 0.24260
56 → 63 0.12804
59 → 63 0.43672
60 → 63 0.40554

Excited State 36: Singlet-A' 6.9483 eV 178.44 nm f = 0.0007
$\langle S^2 \rangle$ = 0.000
50 → 62 0.25584
53 → 62 -0.10148
55 → 62 0.12193
55 → 64 -0.10598
57 → 63 0.14859
58 → 63 -0.24972
59 → 64 0.33275
60 → 64 0.10323
61 → 83 0.41046

Excited State 37: Singlet-A' 7.0232 eV 176.53 nm f=0.0019
$\langle S^2 \rangle$ = 0.000
50 → 62 0.53706
57 → 63 -0.21279
59 → 64 -0.30450
60 → 64 0.19154

Excited State 38: Singlet-A' 7.0338 eV 176.27 nm f = 0.0033
$\langle S^2 \rangle$ = 0.000
49 → 62 -0.23494
50 → 62 -0.28588
52 → 63 -0.12330
56 → 64 0.36381
58 → 63 -0.19012
59 → 64 -0.23657
60 → 64 0.21438
61 → 83 0.18660

Excited State 39: Singlet-A' 7.0609 eV 175.59 nm f = 0.0229
$\langle S^2 \rangle$ = 0.000
49 → 62 0.31612
52 → 63 0.21833
54 → 63 0.14622
56 → 64 0.14044
57 → 63 0.27397
58 → 63 0.19310
59 → 64 -0.28228
60 → 66 -0.12253
61 → 83 0.21185

Excited State 40: Singlet-A' 7.0812 eV 175.09 nm f = 0.0103
$\langle S^2 \rangle$ = 0.000
49 → 62 -0.28201
50 → 62 0.11494
52 → 63 -0.18154
53 → 62 0.10847
54 → 63 -0.19409
57 → 63 0.31640
58 → 63 0.39502

Excited State 41: Singlet-A" 7.0892 eV 174.89 nm f = 0.0032
$\langle S^2 \rangle$ = 0.000
46 → 62 -0.11703
48 → 62 0.41708
51 → 64 -0.13010
52 → 64 0.22799
54 → 64 -0.22317
55 → 63 0.26467
57 → 64 0.16507
58 → 66 -0.10361
59 → 63 -0.15054

Excited State 42: Singlet-A" 7.1012 eV 174.60 nm f = 0.0001
$\langle S^2 \rangle$ = 0.000
48 → 62 -0.10171
56 → 63 0.40384
57 → 64 0.50069
58 → 64 -0.10887

Excited State 43: Singlet-A" 7.1200 eV 174.14 nm f = 0.0000
$\langle S^2 \rangle$ = 0.000
48 → 62 0.12487
51 → 62 0.11990
56 → 63 0.50384
57 → 64 -0.35873
58 → 64 0.17070
60 → 63 -0.11118

Excited State 44: Singlet-A" 7.1437 eV 173.56 nm f = 0.0086
$\langle S^2 \rangle$ = 0.000
46 → 62 -0.21168
48 → 62 -0.23565
51 → 64 0.31350
53 → 63 0.24876
55 → 63 0.36908
59 → 63 -0.14870

Excited State 45: Singlet-A' 7.2052 eV 172.08 nm f = 0.0027
$\langle S^2 \rangle$ = 0.000
53 → 62 -0.13799
53 → 64 -0.12915
54 → 63 0.14055
55 → 64 0.28417
56 → 64 0.34471
58 → 63 0.21608

59 → 64 0.26154
61 → 81 0.25081
Excited State 46: Singlet-A" 7.2203 eV 171.72 nm *f* = 0.0160
<S^2> = 0.000
46 → 62 0.14353
52 → 62 0.11707
52 → 64 -0.16065
53 → 63 -0.31517
54 → 64 0.32984
55 → 63 0.36722
57 → 64 0.10063
Excited State 47: Singlet-A' 7.2414 eV 171.22 nm *f* = 0.0053
<S^2> = 0.000
53 → 62 -0.13542
55 → 64 0.37277
57 → 63 0.30685
58 → 63 -0.18550
60 → 64 0.12054
61 → 81 -0.24655
61 → 83 -0.24974
Excited State 48: Singlet-A" 7.2920 eV 170.03 nm *f* = 0.0015
<S^2> = 0.000
51 → 62 -0.10295
51 → 64 -0.32000
53 → 63 0.46541
54 → 64 0.30749
Excited State 49: Singlet-A' 7.3138 eV 169.52 nm *f* = 0.0005
<S^2> = 0.000
49 → 62 0.22183
50 → 64 0.10819
51 → 63 -0.10631
52 → 63 -0.23441
53 → 64 0.14061
55 → 64 -0.19607
56 → 64 0.29764
57 → 63 0.19959
60 → 66 0.19711
61 → 83 -0.28706
Excited State 50: Singlet-A" 7.3648 eV 168.35 nm *f* = 0.0000
<S^2> = 0.000
46 → 62 0.56251
49 → 63 -0.10617
51 → 64 0.18349
53 → 63 0.20284
54 → 64 -0.19443
54 → 66 -0.12084
Excited State 51: Singlet-A' 7.3674 eV 168.29 nm *f* = 0.0132
<S^2> = 0.000
47 → 62 -0.16064
49 → 62 -0.26412
54 → 63 0.46042
55 → 64 -0.20165
57 → 63 0.12073
59 → 65 -0.11700
60 → 65 -0.22910
Excited State 52: Singlet-A' 7.3707 eV 168.21 nm *f* = 0.0011
<S^2> = 0.000
49 → 62 -0.16940
52 → 63 0.14533
53 → 64 0.14051
55 → 64 -0.10333
60 → 65 0.60057
Excited State 53: Singlet-A' 7.4008 eV 167.53 nm *f* = 0.0053
<S^2> = 0.000
49 → 62 -0.17145
50 → 64 -0.17795
51 → 63 -0.24633
52 → 63 0.42509
54 → 63 -0.21091
55 → 64 -0.15493
59 → 66 0.13731
60 → 65 -0.19112
Excited State 54: Singlet-A" 7.4017 eV 167.51 nm *f* = 0.0001
<S^2> = 0.000
47 → 63 -0.10727
48 → 62 -0.26957
50 → 63 0.25489
52 → 64 0.49380
54 → 64 0.10598
58 → 66 -0.18970
Excited State 55: Singlet-A" 7.4367 eV 166.72 nm *f* = 0.0089
<S^2> = 0.000
48 → 62 0.30525
51 → 64 0.38626
52 → 64 0.21317
54 → 64 0.33050
55 → 63 -0.10152
57 → 66 0.15289
58 → 65 0.11108
Excited State 56: Singlet-A' 7.4481 eV 166.46 nm *f* = 0.0066
<S^2> = 0.000
47 → 62 0.47245
49 → 62 -0.10076
51 → 63 0.24000
53 → 64 0.36633
Excited State 57: Singlet-A' 7.4711 eV 165.95 nm *f* = 0.0025
<S^2> = 0.000
50 → 64 -0.13296
53 → 64 0.17368
59 → 65 0.63648
60 → 65 -0.10920

Excited State 58: Singlet-A' 7.4998 eV 165.32 nm f = 0.0120

$<S^2>$ = 0.000

51 → 63 0.36985

52 → 63 0.10905

53 → 64 -0.31185

55 → 64 -0.20581

56 → 65 -0.14231

59 → 65 0.11315

60 → 66 0.23688

Excited State 59: Singlet-A" 7.5048 eV 165.21 nm f = 0.0039

$<S^2>$ = 0.000

50 → 63 0.27211

52 → 64 -0.11291

58 → 65 0.62132

Excited State 60: Singlet-A' 7.5549 eV 164.11 nm f = 0.0607

$<S^2>$ = 0.000

47 → 62 -0.32026

49 → 62 -0.11102

50 → 64 0.34227

52 → 63 0.16786

53 → 64 0.21188

55 → 66 0.13770

56 → 65 -0.25087

59 → 66 -0.17051

60 → 65 -0.11197

Excited State 61: Singlet-A" 7.5631 eV 163.93 nm f = 0.0397

$<S^2>$ = 0.000

48 → 62 -0.14440

50 → 63 -0.25276

52 → 64 0.20169

57 → 65 0.48552

58 → 65 0.19628

58 → 66 0.16835

Excited State 62: Singlet-A" 7.5933 eV 163.28 nm f = 0.0456

$<S^2>$ = 0.000

48 → 62 0.10075

50 → 63 0.21555

52 → 64 -0.16913

57 → 65 0.48453

57 → 66 0.13810

58 → 65 -0.13622

58 → 66 -0.25779

Excited State 63: Singlet-A' 7.6040 eV 163.05 nm f = 0.0737

$<S^2>$ = 0.000

47 → 62 -0.28040

50 → 64 -0.12274

51 → 63 0.36744

53 → 64 0.13038

55 → 66 0.10666

56 → 65 0.39443

59 → 65 -0.13592

Excited State 64: Singlet-A' 7.6189 eV 162.73 nm f = 0.0398

$<S^2>$ = 0.000

46 → 63 -0.15040

50 → 64 0.14606

51 → 63 -0.14753

53 → 66 0.10571

56 → 65 0.45951

56 → 66 -0.12629

59 → 65 0.11265

59 → 66 -0.26415

60 → 66 0.10939

61 → 87 0.17415

Excited State 65: Singlet-A" 7.6194 eV 162.72 nm f = 0.0000

$<S^2>$ = 0.000

48 → 64 -0.12596

49 → 63 0.60092

52 → 66 0.11038

53 → 63 -0.12993

Excited State 66: Singlet-A' 7.6482 eV 162.11 nm f = 0.0659

$<S^2>$ = 0.000

46 → 63 -0.14906

50 → 64 -0.35326

53 → 64 0.14189

54 → 63 0.11958

55 → 65 -0.10095

56 → 65 -0.12828

59 → 65 -0.10311

59 → 66 -0.10594

60 → 66 0.41982

Excited State 67: Singlet-A" 7.6634 eV 161.79 nm f = 0.0465

$<S^2>$ = 0.000

48 → 64 0.28686

49 → 63 0.11196

50 → 63 0.34885

57 → 66 -0.18771

58 → 65 -0.13576

58 → 66 0.25874

61 → 85 0.25028

61 → 88 -0.12905

Excited State 68: Singlet-A" 7.6695 eV 161.66 nm f = 0.0382

$<S^2>$ = 0.000

48 → 64 0.53579

49 → 63 0.12499

50 → 63 -0.21664

58 → 66 -0.12709

61 → 84 0.11681

61 → 85 -0.12020

Excited State 69: Singlet-A' 7.6759 eV 161.52 nm f = 0.0265

$<S^2>$ = 0.000

50 → 64 -0.22606
54 → 63 -0.12298
55 → 65 0.10175
56 → 65 -0.10222
60 → 66 -0.25733
61 → 86 -0.34429
61 → 87 0.40966

Excited State 70: Singlet-A' 7.6966 eV 161.09 nm f = 0.0096

$\langle S^2 \rangle$ = 0.000

45 → 62 0.10631
50 → 64 0.19696
52 → 63 -0.13418
55 → 65 -0.14255
56 → 66 0.19413
59 → 66 0.38941
60 → 66 0.16881
61 → 86 -0.18745
61 → 87 0.30767

Excited State 71: Singlet-A" 7.7125 eV 160.76 nm f = 0.0071

$\langle S^2 \rangle$ = 0.000

50 → 63 -0.13003
52 → 64 -0.10477
57 → 66 0.13223
58 → 66 -0.26802
61 → 84 -0.12222
61 → 85 0.47577
61 → 88 -0.31581

Excited State 72: Singlet-A" 7.7661 eV 159.65 nm f = 0.0000

$\langle S^2 \rangle$ = 0.000

44 → 62 0.22846
46 → 64 -0.25856
56 → 67 0.10133
57 → 66 0.47101
58 → 66 0.17933
61 → 84 0.16654

Excited State 73: Singlet-A' 7.7669 eV 159.63 nm f = 0.0048

$\langle S^2 \rangle$ = 0.000

45 → 62 0.15892
46 → 63 -0.11126
55 → 65 0.57875
56 → 66 0.25879

Excited State 74: Singlet-A' 7.7883 eV 159.19 nm f = 0.0038

$\langle S^2 \rangle$ = 0.000

45 → 62 0.21086
46 → 63 -0.14445
49 → 64 -0.13660
53 → 64 -0.11120
55 → 65 -0.29820
55 → 66 -0.10143
56 → 66 0.35966
59 → 66 -0.21802
60 → 66 -0.14133
61 → 86 0.14934
61 → 87 0.11707

Excited State 75: Singlet-A" 7.8033 eV 158.89 nm f = 0.0000

$\langle S^2 \rangle$ = 0.000

48 → 64 -0.11287
54 → 65 -0.34881
57 → 66 -0.11167
61 → 84 0.50884
61 → 85 0.10517
61 → 89 -0.19889

Excited State 76: Singlet-A" 7.8146 eV 158.66 nm f = 0.0000

$\langle S^2 \rangle$ = 0.000

52 → 65 -0.25091
54 → 65 -0.24739
61 → 75 -0.13397
61 → 89 0.56490

Excited State 77: Singlet-A' 7.8338 eV 158.27 nm f = 0.0019

$\langle S^2 \rangle$ = 0.000

48 → 63 0.15094
49 → 64 0.62265
61 → 86 0.16270
61 → 87 0.11313

Excited State 78: Singlet-A" 7.8380 eV 158.18 nm f = 0.0056

$\langle S^2 \rangle$ = 0.000

54 → 65 0.51855
61 → 84 0.36990
61 → 88 -0.20954
61 → 89 0.13768

Excited State 79: Singlet-A' 7.8489 eV 157.96 nm f = 0.0084

$\langle S^2 \rangle$ = 0.000

48 → 63 -0.10560
49 → 64 -0.12197
53 → 65 -0.25004
55 → 65 0.10595
56 → 66 -0.15562
61 → 86 0.45239
61 → 87 0.34915

Excited State 80: Singlet-A' 7.8612 eV 157.72 nm f = 0.0003

$\langle S^2 \rangle$ = 0.000

48 → 63 0.61976
49 → 64 -0.19325

Excited State 81: Singlet-A' 7.8810 eV 157.32 nm f = 0.0073

$\langle S^2 \rangle$ = 0.000

45 → 62 -0.17023
47 → 64 0.37526
53 → 65 0.10703
53 → 66 0.21856
55 → 66 0.27934
56 → 66 0.22659
57 → 67 -0.11542

58 → 67 -0.11054
59 → 66 0.11872
61 → 86 0.15749
Excited State 82: Singlet-A" 7.8847 eV 157.25 nm f = 0.0164
$<S^2>$ = 0.000
44 → 62 -0.25507
47 → 63 0.44179
51 → 66 -0.11469
54 → 66 0.19551
57 → 66 0.16872
58 → 66 -0.19940
59 → 67 0.12377
60 → 67 -0.16540
Excited State 83: Singlet-A" 7.8972 eV 157.00 nm f = 0.0010
$<S^2>$ = 0.000
51 → 65 -0.10750
52 → 65 0.59522
54 → 66 0.12913
61 → 85 -0.11487
61 → 89 0.22244
Excited State 84: Singlet-A' 7.8980 eV 156.98 nm f = 0.0078
$<S^2>$ = 0.000
45 → 62 0.20585
48 → 63 -0.14349
53 → 65 0.56197
56 → 66 -0.20829
61 → 86 0.17132
Excited State 85: Singlet-A" 7.9087 eV 156.77 nm f = 0.0004
$<S^2>$ = 0.000
51 → 65 0.10328
52 → 65 0.17668
54 → 65 0.14114
60 → 67 0.10322
61 → 85 0.34520
61 → 88 0.48768
61 → 89 0.12707
Excited State 86: Singlet-A" 7.9236 eV 156.47 nm f = 0.0011
$<S^2>$ = 0.000
38 → 62 0.21521
46 → 66 0.11671
47 → 63 -0.10108
48 → 64 0.10482
49 → 63 -0.10583
51 → 65 -0.15569
52 → 66 0.42034
56 → 68 0.10631
57 → 69 -0.10951
59 → 67 -0.13298
60 → 67 -0.16180
61 → 85 0.13515
61 → 88 0.12245
Excited State 87: Singlet-A" 7.9397 eV 156.16 nm f = 0.0058
$<S^2>$ = 0.000
44 → 62 0.26124
46 → 62 0.11412
51 → 65 0.48740
52 → 66 0.13273
54 → 66 0.31239
57 → 66 -0.10955
Excited State 88: Singlet-A' 7.9465 eV 156.02 nm f = 0.0206
$<S^2>$ = 0.000
43 → 62 0.31477
46 → 63 0.11981
47 → 64 -0.29677
48 → 63 0.10994
53 → 65 0.16309
53 → 66 -0.13744
55 → 66 0.37091
59 → 66 -0.12737
Excited State 89: Singlet-A" 7.9620 eV 155.72 nm f = 0.0178
$<S^2>$ = 0.000
46 → 62 0.15364
47 → 63 -0.13173
51 → 65 -0.29894
51 → 66 0.10573
52 → 65 -0.11495
54 → 66 0.48818
60 → 67 0.18614
Excited State 90: Singlet-A" 7.9669 eV 155.62 nm f = 0.0027
$<S^2>$ = 0.000
44 → 62 0.36014
51 → 65 -0.26742
52 → 66 -0.20440
56 → 68 0.16219
57 → 69 -0.12900
60 → 67 -0.21638
60 → 68 0.24055
61 → 88 0.13375
Excited State 91: Singlet-A' 7.9673 eV 155.62 nm f = 0.0003
$<S^2>$ = 0.000
43 → 62 0.10741
45 → 62 0.50068
53 → 65 -0.24146
53 → 66 0.18608
55 → 66 0.21232
56 → 66 -0.15722
61 → 87 -0.10944
Excited State 92: Singlet-A" 8.0068 eV 154.85 nm f = 0.0092
$<S^2>$ = 0.000
44 → 62 0.31608

47 → 63 0.23725
51 → 65 -0.15777
51 → 66 -0.15669
52 → 66 0.20244
54 → 66 -0.12110
56 → 68 -0.20631
57 → 66 -0.17662
60 → 67 0.14556
60 → 68 -0.19009

Excited State 93: Singlet-A' 8.0225 eV 154.55 nm f = 0.0087
$<S^2>$ = 0.000
50 → 65 0.47310
50 → 69 -0.13092
53 → 66 0.14142
56 → 69 -0.13819
57 → 67 0.17676
59 → 69 0.16402
60 → 69 0.27706

Excited State 94: Singlet-A" 8.0392 eV 154.23 nm f = 0.0015
$<S^2>$ = 0.000
50 → 68 0.13803
51 → 66 -0.18881
56 → 67 -0.19378
57 → 69 0.36200
59 → 67 0.12534
59 → 68 0.11725
60 → 67 0.16402
60 → 68 0.31061

Excited State 95: Singlet-A' 8.0481 eV 154.05 nm f = 0.0600
$<S^2>$ = 0.000
43 → 62 -0.13543
45 → 62 -0.12480
46 → 63 -0.10606
47 → 64 -0.25518
50 → 65 -0.35043
53 → 66 0.40123
58 → 67 0.12156

Excited State 96: Singlet-A' 8.0625 eV 153.78 nm f = 0.0254
$<S^2>$ = 0.000
43 → 62 -0.28300
47 → 64 -0.22131
50 → 65 0.33896
50 → 66 0.11384
50 → 69 0.11784
53 → 66 0.14959
56 → 69 0.10392
57 → 67 -0.13423
59 → 69 -0.18163
60 → 69 -0.25669

Excited State 97: Singlet-A" 8.0721 eV 153.60 nm f = 0.0353
$<S^2>$ = 0.000
47 → 63 0.16087
51 → 66 0.54464
52 → 66 0.20178
59 → 67 0.14948

Excited State 98: Singlet-A' 8.0856 eV 153.34 nm f = 0.0515
$<S^2>$ = 0.000
43 → 62 -0.31330
45 → 62 0.17159
46 → 63 0.45654
49 → 66 -0.11433
57 → 67 0.10432
59 → 66 -0.13168
60 → 69 0.14671

Excited State 99: Singlet-A" 8.1161 eV 152.76 nm f = 0.0054
$<S^2>$ = 0.000
46 → 64 0.45256
48 → 66 0.12542
50 → 67 0.10130
56 → 68 -0.21490
57 → 66 0.12466
57 → 69 -0.18841
58 → 66 0.16559
59 → 67 0.16923

Excited State 100: Singlet-A" 8.1284 eV 152.53 nm f = 0.0053
$<S^2>$ = 0.000
44 → 62 0.10206
46 → 64 0.34494
50 → 67 -0.14188
50 → 68 -0.12381
53 → 68 0.14353
55 → 67 -0.10192
55 → 68 -0.13223
56 → 67 0.11285
56 → 68 0.29950
57 → 69 0.21789
59 → 68 -0.12451
60 → 68 -0.15273

Excitation energies and oscillator strengths (*f*) calculated for $[Mo(CN)_8]^{4-}$.

Excited State 1: Singlet-A" 3.1296 eV 396.17 nm *f* = 0.0002
<S^2> = 0.000
61 → 62 0.70341

Excited State 2: Singlet-A' 3.6926 eV 335.76 nm *f* = 0.0038
<S^2> = 0.000
61 → 63 0.69770

Excited State 3: Singlet-A" 3.7062 eV 334.54 nm *f* = 0.0044
<S^2> = 0.000
61 → 64 0.69376

Excited State 4: Singlet-A" 4.3309 eV 286.28 nm *f* = 0.0001
<S^2> = 0.000
61 → 65 0.70274

Excited State 5: Singlet-A" 4.6172 eV 268.52 nm *f* = 0.0002
<S^2> = 0.000
61 → 66 0.53116
61 → 67 -0.17474
61 → 69 -0.39739
61 → 76 0.16166

Excited State 6: Singlet-A" 4.9720 eV 249.37 nm *f* = 0.0000
<S^2> = 0.000
61 → 66 0.30080
61 → 67 0.62714
61 → 69 0.12162

Excited State 7: Singlet-A' 4.9863 eV 248.65 nm *f* = 0.0005
<S^2> = 0.000
61 → 68 0.70563

Excited State 8: Singlet-A" 5.0358 eV 246.20 nm *f* = 0.0006
<S^2> = 0.000
61 → 66 0.34207
61 → 67 -0.27061
61 → 69 0.54989

Excited State 9: Singlet-A" 5.4250 eV 228.54 nm *f* = 0.0001
<S^2> = 0.000
61 → 69 0.14590
61 → 73 -0.17672
61 → 76 0.65409

Excited State 10: Singlet-A' 5.5070 eV 225.14 nm *f* = 0.0337
<S^2> = 0.000
61 → 70 0.46147
61 → 71 0.34495
61 → 74 0.36510
61 → 77 -0.11530

Excited State 11: Singlet-A" 5.5725 eV 222.49 nm *f* = 0.0312
<S^2> = 0.000
61 → 72 0.49952
61 → 73 0.21110
61 → 75 -0.41344
61 → 76 0.10302

Excited State 12: Singlet-A" 5.6004 eV 221.39 nm *f* = 0.0033
<S^2> = 0.000
61 → 72 -0.11840
61 → 73 0.63998
61 → 75 0.20774
61 → 76 0.14614

Excited State 13: Singlet-A' 5.6562 eV 219.20 nm *f* = 0.0695
<S^2> = 0.000
61 → 70 0.50364
61 → 71 -0.41386
61 → 74 -0.21690

Excited State 14: Singlet-A' 5.9029 eV 210.04 nm *f* = 0.0685
<S^2> = 0.000
61 → 70 -0.10771
61 → 71 -0.41067
61 → 74 0.51307
61 → 77 -0.12721

Excited State 15: Singlet-A" 5.9143 eV 209.63 nm *f* = 0.0706
<S^2> = 0.000
61 → 72 0.43924
61 → 75 0.51001

Excited State 16: Singlet-A' 6.4471 eV 192.31 nm *f* = 0.0034
<S^2> = 0.000
59 → 62 -0.27798
60 → 62 0.59134
61 → 74 0.10706
61 → 77 0.12685

Excited State 17: Singlet-A' 6.4700 eV 191.63 nm *f* = 0.0030
<S^2> = 0.000
55 → 62 0.16634
59 → 62 0.44591
60 → 62 0.32531
61 → 77 -0.34856

Excited State 18: Singlet-A" 6.4874 eV 191.12 nm *f* = 0.0006
<S^2> = 0.000
56 → 62 0.13210
58 → 62 0.66360
60 → 63 -0.11126

Excited State 19: Singlet-A' 6.4937 eV 190.93 nm *f* = 0.0011
<S^2> = 0.000
55 → 62 -0.11173
57 → 62 -0.19182
59 → 62 0.42144
61 → 74 0.10668
61 → 77 0.46505

Excited State 20: Singlet-A' 6.6477 eV 186.51 nm *f* = 0.0096
<S^2> = 0.000
55 → 62 0.28814
55 → 64 -0.13045

57 → 62 0.50693
58 → 63 0.13289
59 → 64 -0.10613
61 → 77 0.22271

Excited State 21: Singlet-A" 6.6765 eV 185.70 nm f = 0.0078
$<S^2>$ = 0.000
56 → 62 0.64733
58 → 62 -0.11194
61 → 79 -0.15442

Excited State 22: Singlet-A' 6.6922 eV 185.27 nm f = 0.0016
$<S^2>$ = 0.000
53 → 62 -0.22207
55 → 62 0.50446
57 → 62 -0.35702
61 → 78 -0.15302

Excited State 23: Singlet-A" 6.7419 eV 183.90 nm f = 0.0150
$<S^2>$ = 0.000
54 → 62 0.64439
55 → 63 -0.12826
57 → 63 -0.10739
59 → 63 -0.11570

Excited State 24: Singlet-A" 6.7878 eV 182.66 nm f = 0.0001
$<S^2>$ = 0.000
52 → 62 0.29629
53 → 63 -0.10257
54 → 62 -0.11796
59 → 63 -0.36022
60 → 63 0.36546
61 → 79 0.15154
61 → 80 -0.25359

Excited State 25: Singlet-A' 6.8169 eV 181.88 nm f = 0.0223
$<S^2>$ = 0.000
53 → 62 0.56404
53 → 64 0.13967
55 → 62 0.14759
57 → 64 0.10244
58 → 63 0.14556
61 → 78 -0.20904

Excited State 26: Singlet-A" 6.8368 eV 181.35 nm f = 0.0099
$<S^2>$ = 0.000
51 → 62 0.44141
52 → 62 -0.41489
55 → 63 0.10390
60 → 63 0.14560
61 → 79 0.18883

Excited State 27: Singlet-A" 6.8385 eV 181.30 nm f = 0.0008
$<S^2>$ = 0.000
51 → 62 -0.23755
52 → 62 -0.28127
58 → 64 0.30192
59 → 63 -0.26615
60 → 63 0.29822
61 → 80 0.27822

Excited State 28: Singlet-A' 6.8929 eV 179.87 nm f = 0.0028
$<S^2>$ = 0.000
53 → 62 0.11376
57 → 62 -0.16462
59 → 64 0.10501
60 → 64 0.35732
61 → 78 0.52822

Excited State 29: Singlet-A" 6.9007 eV 179.67 nm f = 0.0002
$<S^2>$ = 0.000
51 → 62 0.37862
52 → 62 0.22638
56 → 64 -0.10592
58 → 64 0.41493
60 → 63 -0.11293
61 → 79 -0.22458
61 → 80 0.10473

Excited State 30: Singlet-A" 6.9292 eV 178.93 nm f = 0.0011
$<S^2>$ = 0.000
51 → 62 0.10617
52 → 62 0.22488
55 → 63 -0.10783
56 → 62 0.10516
59 → 63 0.29137
60 → 63 0.22530
61 → 79 0.33070
61 → 80 0.38628

Excited State 31: Singlet-A" 6.9406 eV 178.64 nm f = 0.0019
$<S^2>$ = 0.000
51 → 62 -0.15517
58 → 64 0.42242
59 → 63 0.14818
60 → 63 -0.10468
61 → 79 0.35748
61 → 80 -0.31916

Excited State 32: Singlet-A' 7.0201 eV 176.61 nm f = 0.0062
$<S^2>$ = 0.000
55 → 62 -0.13365
55 → 64 0.12769
59 → 64 -0.10706
60 → 64 0.52748
61 → 78 -0.32191

Excited State 33: Singlet-A' 7.0448 eV 175.99 nm f = 0.0090
$<S^2>$ = 0.000
55 → 64 0.12878
57 → 64 0.13353
59 → 64 0.60816
61 → 78 -0.14539
61 → 82 0.10091

Excited State 34: Singlet-A" 7.0508 eV 175.84 nm f = 0.0113
$<S^2>$ = 0.000
55 → 63 -0.20418
57 → 63 -0.25589
59 → 63 0.30496
60 → 63 0.30619
61 → 79 -0.28295
61 → 80 -0.24085
Excited State 35: Singlet-A' 7.0565 eV 175.70 nm f = 0.0006
$<S^2>$ = 0.000
50 → 62 0.66053
56 → 63 0.15234
57 → 64 0.12140
Excited State 36: Singlet-A' 7.0739 eV 175.27 nm f = 0.0005
$<S^2>$ = 0.000
53 → 62 -0.11108
55 → 62 -0.12434
58 → 63 0.62225
61 → 82 0.17960
Excited State 37: Singlet-A' 7.1462 eV 173.50 nm f = 0.0032
$<S^2>$ = 0.000
49 → 62 0.33265
52 → 63 0.14666
53 → 64 0.10771
56 → 63 -0.31511
57 → 64 0.37038
59 → 64 -0.10247
60 → 64 -0.14761
61 → 82 -0.20313
Excited State 38: Singlet-A" 7.1655 eV 173.03 nm f = 0.0001
$<S^2>$ = 0.000
48 → 62 -0.29091
53 → 63 -0.18156
54 → 62 0.11849
54 → 64 -0.16375
55 → 63 0.31583
56 → 64 0.23257
57 → 63 0.10470
59 → 63 0.19377
60 → 63 0.13188
61 → 81 -0.22660
Excited State 39: Singlet-A' 7.1757 eV 172.78 nm f = 0.0065
$<S^2>$ = 0.000
49 → 62 0.42655
52 → 63 0.21594
54 → 63 -0.20631
55 → 64 0.15359
56 → 63 0.23252
57 → 64 -0.21906
60 → 65 -0.13314
61 → 82 0.15425
Excited State 40: Singlet-A" 7.1811 eV 172.65 nm f = 0.0004
$<S^2>$ = 0.000
48 → 62 0.23433
51 → 62 0.14624
54 → 64 0.11432
55 → 63 -0.23428
56 → 64 0.48909
61 → 81 -0.27267
Excited State 41: Singlet-A" 7.1899 eV 172.44 nm f = 0.0002
$<S^2>$ = 0.000
54 → 64 0.18632
55 → 63 -0.20399
56 → 64 -0.22206
57 → 63 0.45699
58 → 64 -0.11509
60 → 63 0.11732
61 → 81 -0.26698
Excited State 42: Singlet-A" 7.2087 eV 171.99 nm f = 0.0000
$<S^2>$ = 0.000
53 → 63 0.12768
55 → 63 -0.11129
56 → 64 0.30517
57 → 63 0.32912
61 → 81 0.47031
Excited State 43: Singlet-A' 7.2177 eV 171.78 nm f = 0.0015
$<S^2>$ = 0.000
55 → 64 0.15409
56 → 63 0.32183
57 → 64 0.20347
59 → 64 -0.10512
60 → 65 0.47484
61 → 83 0.14879
Excited State 44: Singlet-A" 7.2322 eV 171.43 nm f = 0.0027
$<S^2>$ = 0.000
48 → 62 0.37757
51 → 64 0.34187
53 → 63 -0.29425
55 → 63 0.12943
56 → 64 -0.10055
57 → 63 0.20540
61 → 79 -0.14157
Excited State 45: Singlet-A' 7.2399 eV 171.25 nm f = 0.0100
$<S^2>$ = 0.000
56 → 63 -0.28307
57 → 64 -0.28945
59 → 65 0.21052
60 → 65 0.43677
61 → 83 -0.17222

Excited State 46: Singlet-A" 7.2826 eV 170.25 nm f = 0.0081

$<S^2>$ = 0.000

52 → 64	0.10133
53 → 63	0.27961
54 → 64	0.35629
55 → 63	0.29511
58 → 65	0.35184

Excited State 47: Singlet-A' 7.2888 eV 170.10 nm f = 0.0002

$<S^2>$ = 0.000

52 → 63	0.12789
55 → 64	-0.30824
56 → 63	0.18932
57 → 64	0.13762
59 → 65	0.50137
61 → 83	0.15098

Excited State 48: Singlet-A" 7.3393 eV 168.93 nm f = 0.0030

$<S^2>$ = 0.000

52 → 64	0.14185
53 → 63	-0.27265
54 → 64	0.40737
55 → 63	0.16971
58 → 65	-0.37693
61 → 81	0.16165

Excited State 49: Singlet-A' 7.3482 eV 168.73 nm f = 0.0158

$<S^2>$ = 0.000

52 → 63	-0.26206
53 → 62	-0.12579
53 → 64	0.14380
54 → 63	0.14475
55 → 64	0.32894
57 → 65	0.10830
59 → 64	-0.10416
59 → 65	0.37873
60 → 65	-0.12231
61 → 82	0.14211
61 → 83	-0.10245

Excited State 50: Singlet-A" 7.3797 eV 168.01 nm f = 0.0167

$<S^2>$ = 0.000

51 → 64	-0.27858
53 → 63	-0.37600
54 → 64	0.11272
55 → 63	-0.11913
58 → 65	0.41114
61 → 81	0.11627

Excited State 51: Singlet-A" 7.4367 eV 166.72 nm f = 0.0002

$<S^2>$ = 0.000

46 → 62	0.43078
51 → 64	-0.13018
56 → 65	0.45774
60 → 68	0.11486

Excited State 52: Singlet-A' 7.4404 eV 166.64 nm f = 0.0075

$<S^2>$ = 0.000

47 → 62	0.25804
49 → 62	0.29750
52 → 63	-0.22497
54 → 63	0.34537
57 → 64	-0.14018
57 → 65	-0.12148
61 → 82	-0.19801

Excited State 53: Singlet-A" 7.4470 eV 166.49 nm f = 0.0003

$<S^2>$ = 0.000

46 → 62	-0.45650
56 → 65	0.45771
60 → 68	0.10742

Excited State 54: Singlet-A' 7.4481 eV 166.46 nm f = 0.0007

$<S^2>$ = 0.000

57 → 64	-0.10211
57 → 65	0.59435
60 → 67	0.13845
61 → 82	-0.17003

Excited State 55: Singlet-A' 7.4781 eV 165.80 nm f = 0.0023

$<S^2>$ = 0.000

47 → 62	0.27437
49 → 62	-0.10726
50 → 64	0.17658
51 → 63	0.29833
52 → 63	0.32961
53 → 64	0.12861
54 → 63	0.28338
61 → 82	0.13900

Excited State 56: Singlet-A' 7.4914 eV 165.50 nm f = 0.0127

$<S^2>$ = 0.000

49 → 62	0.15406
54 → 63	0.14860
55 → 64	-0.25356
57 → 64	0.10395
57 → 65	0.16746
61 → 82	0.45426

Excited State 57: Singlet-A" 7.4914 eV 165.50 nm f = 0.0009

$<S^2>$ = 0.000

48 → 62	-0.20785
50 → 63	-0.25566
51 → 64	0.27195
52 → 64	0.47245
58 → 66	0.11165
58 → 69	-0.10617

Excited State 58: Singlet-A' 7.5095 eV 165.10 nm f = 0.0020

$<S^2>$ = 0.000

47 → 62	0.41136
51 → 63	-0.39382
53 → 64	0.25731

54 → 63 -0.17708
61 → 82 0.13380
Excited State 59: Singlet-A" 7.5169 eV 164.94 nm f = 0.0066
<S^2> = 0.000
46 → 62 -0.10251
48 → 62 0.27089
51 → 64 -0.32634
52 → 64 0.38098
54 → 64 -0.24538
55 → 63 0.11034
Excited State 60: Singlet-A' 7.5308 eV 164.64 nm f = 0.0016
<S^2> = 0.000
51 → 63 0.12520
53 → 64 0.22314
55 → 65 -0.30927
56 → 63 -0.10723
57 → 64 -0.10719
59 → 66 -0.11978
60 → 66 -0.12864
61 → 83 0.47670
Excited State 61: Singlet-A' 7.5917 eV 163.32 nm f = 0.0214
<S^2> = 0.000
47 → 62 0.16627
53 → 64 -0.29749
55 → 64 0.16466
55 → 65 0.33048
56 → 63 -0.14503
59 → 69 0.11215
60 → 66 -0.22048
60 → 69 0.13980
61 → 83 0.24193
Excited State 62: Singlet-A' 7.6125 eV 162.87 nm f = 0.0415
<S^2> = 0.000
47 → 62 -0.24996
50 → 64 -0.15257
52 → 63 0.13605
53 → 64 0.32024
55 → 64 -0.11845
55 → 65 0.37863
57 → 64 -0.10237
59 → 66 0.15072
60 → 66 -0.11980
Excited State 63: Singlet-A' 7.6236 eV 162.63 nm f = 0.0299
<S^2> = 0.000
47 → 62 0.13331
50 → 64 0.21808
51 → 63 0.30680
52 → 63 -0.24395
53 → 64 0.11765
54 → 63 -0.29372
55 → 64 -0.13404
55 → 65 0.21541
57 → 65 0.11974
Excited State 64: Singlet-A" 7.6466 eV 162.14 nm f = 0.0569
<S^2> = 0.000
48 → 62 -0.15509
49 → 63 -0.11184
50 → 63 0.43693
52 → 64 0.23973
54 → 65 -0.19344
58 → 66 -0.24363
Excited State 65: Singlet-A' 7.6808 eV 161.42 nm f = 0.0804
<S^2> = 0.000
47 → 62 -0.15870
50 → 64 0.41737
51 → 63 -0.31303
53 → 65 0.15614
54 → 63 0.10277
59 → 65 -0.10094
59 → 66 -0.19190
59 → 69 0.10516
Excited State 66: Singlet-A" 7.6978 eV 161.06 nm f = 0.0404
<S^2> = 0.000
50 → 63 0.23917
52 → 65 -0.16996
54 → 65 0.55199
58 → 66 0.18850
Excited State 67: Singlet-A" 7.7023 eV 160.97 nm f = 0.0001
<S^2> = 0.000
46 → 62 -0.10989
49 → 63 0.63847
Excited State 68: Singlet-A' 7.7055 eV 160.90 nm f = 0.0527
<S^2> = 0.000
50 → 64 -0.30269
53 → 65 0.48514
59 → 66 -0.23727
60 → 66 0.13635
60 → 67 -0.12729
Excited State 69: Singlet-A" 7.7317 eV 160.36 nm f = 0.0057
<S^2> = 0.000
48 → 64 0.37497
50 → 63 0.11642
51 → 65 0.19902
52 → 65 0.44462
58 → 67 -0.14913
59 → 68 0.13721
Excited State 70: Singlet-A" 7.7478 eV 160.03 nm f = 0.0281
<S^2> = 0.000
47 → 63 -0.10788
48 → 64 0.20595

50 → 63 -0.26291
51 → 65 0.34878
52 → 65 -0.21217
54 → 65 0.17332
58 → 66 -0.26988
60 → 68 -0.10188

Excited State 71: Singlet-A" 7.7484 eV 160.01 nm f = 0.0025

$\langle S^2 \rangle$ = 0.000

48 → 64 0.49104
51 → 65 -0.33952
52 → 65 -0.21655
59 → 68 -0.10696

Excited State 72: Singlet-A' 7.7913 eV 159.13 nm f = 0.0002

$\langle S^2 \rangle$ = 0.000

46 → 63 0.20010
47 → 64 -0.14028
50 → 64 0.15535
53 → 65 0.38704
55 → 65 -0.13935
59 → 66 0.25280
59 → 69 -0.16211
60 → 66 -0.17073
60 → 69 0.12182

Excited State 73: Singlet-A" 7.8165 eV 158.62 nm f = 0.0020

$\langle S^2 \rangle$ = 0.000

46 → 64 -0.13133
47 → 63 0.16622
50 → 63 0.10476
51 → 65 0.35826
52 → 65 -0.28075
54 → 65 -0.19839
58 → 66 0.19849
58 → 69 -0.22660

Excited State 74: Singlet-A' 7.8212 eV 158.52 nm f = 0.0002

$\langle S^2 \rangle$ = 0.000

50 → 65 0.55625
56 → 68 0.19703
57 → 67 -0.15732
59 → 69 -0.10128
60 → 69 -0.23207

Excited State 75: Singlet-A' 7.8720 eV 157.50 nm f = 0.0001

$\langle S^2 \rangle$ = 0.000

45 → 62 -0.18796
47 → 64 -0.11869
48 → 63 -0.15045
49 → 64 -0.11940
57 → 66 -0.14597
57 → 69 0.10130
59 → 66 0.18985
59 → 69 -0.12307
60 → 66 0.35090
60 → 67 -0.17723
61 → 83 0.18907

Excited State 76: Singlet-A' 7.8913 eV 157.11 nm f = 0.0008

$\langle S^2 \rangle$ = 0.000

48 → 63 0.13991
49 → 64 0.62559
60 → 66 0.16914

Excited State 77: Singlet-A' 7.9193 eV 156.56 nm f = 0.0004

$\langle S^2 \rangle$ = 0.000

48 → 63 0.13549
49 → 64 -0.16507
50 → 65 0.12097

57 → 65 -0.17829
60 → 66 0.33120
60 → 67 0.46837
60 → 69 0.13542

Excited State 78: Singlet-A" 7.9194 eV 156.56 nm f = 0.0002
$\langle S^2 \rangle$ = 0.000
44 → 62 -0.30897
46 → 64 0.25538
56 → 66 0.38630
56 → 67 -0.10732
56 → 69 -0.20937
60 → 68 -0.23388

Excited State 79: Singlet-A' 7.9225 eV 156.50 nm f = 0.0006
$\langle S^2 \rangle$ = 0.000
45 → 62 0.14221
48 → 63 0.49794
49 → 64 -0.16691
57 → 66 0.10896
58 → 68 0.10705
59 → 66 0.15809
60 → 67 -0.21424

Excited State 80: Singlet-A' 7.9326 eV 156.30 nm f = 0.0009
$\langle S^2 \rangle$ = 0.000
45 → 62 -0.18395
47 → 64 -0.24719
48 → 63 0.31893
49 → 65 0.15992
55 → 66 0.12559
57 → 66 -0.14422
59 → 66 -0.28992
59 → 67 -0.15542
60 → 67 -0.12208
60 → 69 0.13205

Excited State 81: Singlet-A" 7.9400 eV 156.15 nm f = 0.0000
$\langle S^2 \rangle$ = 0.000
44 → 62 -0.18892
46 → 64 0.15460
47 → 63 -0.10023
48 → 65 0.11513
56 → 65 -0.15222
60 → 68 0.59125

Excited State 82: Singlet-A" 7.9449 eV 156.06 nm f = 0.0095
$\langle S^2 \rangle$ = 0.000
44 → 62 -0.17343
47 → 63 0.47112
54 → 65 0.10382
58 → 66 -0.33695
59 → 68 -0.10655

Excited State 83: Singlet-A' 7.9504 eV 155.95 nm f = 0.0036
$\langle S^2 \rangle$ = 0.000
45 → 62 0.29749
46 → 63 0.26367
47 → 64 -0.23871
48 → 63 -0.19189
49 → 65 0.13899
53 → 65 -0.14080
57 → 66 0.19413
57 → 69 -0.11746
59 → 69 -0.11252
60 → 66 0.10528
60 → 67 -0.18477

Excited State 84: Singlet-A' 7.9673 eV 155.62 nm f = 0.0090
$\langle S^2 \rangle$ = 0.000
45 → 62 0.17218
47 → 64 0.35428
49 → 65 0.14662
57 → 66 -0.34314
59 → 67 -0.22105
59 → 69 -0.10930
60 → 69 0.25226

Excited State 85: Singlet-A" 8.0019 eV 154.94 nm f = 0.0030

$<S^2>$ = 0.000

44 → 62	-0.21568
47 → 63	0.17445
48 → 65	0.16648
56 → 66	-0.24208
58 → 66	0.15422
58 → 67	0.28696
58 → 69	0.16226
59 → 68	0.40387
60 → 68	-0.10581

Excited State 86: Singlet-A' 8.0099 eV 154.79 nm f = 0.0000

$<S^2>$ = 0.000

43 → 62	0.11527
50 → 65	0.12988
58 → 68	-0.12609
59 → 66	0.15940
59 → 67	0.38078
59 → 69	0.18419
60 → 67	-0.14479
60 → 69	0.37532

Excited State 87: Singlet-A" 8.0271 eV 154.46 nm f = 0.0005

$<S^2>$ = 0.000

44 → 62	0.16721
46 → 64	-0.10353
48 → 65	-0.13926
56 → 65	-0.12659
56 → 66	0.17832
58 → 66	-0.20092
58 → 69	-0.24466
59 → 68	0.47276

Excited State 88: Singlet-A' 8.0377 eV 154.25 nm f = 0.0024

$<S^2>$ = 0.000

43 → 62	-0.23399
45 → 62	-0.10258
55 → 66	0.13174
55 → 69	-0.10204
57 → 66	0.17172
57 → 67	-0.10375
58 → 68	0.25462
59 → 66	0.16679
59 → 67	-0.24879
59 → 69	0.27044
60 → 69	0.21372

Excited State 89: Singlet-A" 8.0443 eV 154.13 nm f = 0.0005

$<S^2>$ = 0.000

38 → 62	-0.19661
44 → 62	0.11738
51 → 65	0.11015
52 → 65	0.12824
52 → 66	-0.17318
52 → 69	0.13117
58 → 67	0.46713
59 → 68	-0.18188

Excited State 90: Singlet-A" 8.0625 eV 153.78 nm f = 0.0002

$<S^2>$ = 0.000

38 → 62	0.29619
51 → 66	0.14611
51 → 69	-0.12220
52 → 65	0.11977
52 → 66	0.21412
52 → 67	-0.10072
52 → 69	-0.18817
58 → 67	0.32933
60 → 70	-0.12131

Excited State 91: Singlet-A' 8.0629 eV 153.77 nm f = 0.0006

$<S^2>$ = 0.000

45 → 62	0.21563
53 → 66	-0.15436
53 → 67	0.10078
55 → 66	0.16139
55 → 69	-0.11572
57 → 66	-0.10055
58 → 68	0.38918
59 → 67	0.27259
61 → 84	-0.19144

Excited State 92: Singlet-A' 8.0689 eV 153.66 nm f = 0.0105

$<S^2>$ = 0.000

42 → 62	-0.10615
43 → 62	-0.18255
45 → 62	0.16484
46 → 63	-0.10437
47 → 64	0.14136
47 → 65	0.11934
49 → 65	0.10817
55 → 66	0.37711
55 → 69	-0.13694
58 → 68	-0.34314

Excited State 93: Singlet-A" 8.0894 eV 153.27 nm f = 0.0017

$<S^2>$ = 0.000

44 → 62	-0.27650
46 → 62	0.10927
46 → 64	-0.13999
48 → 65	-0.15854
54 → 66	0.34395
54 → 67	-0.15252
54 → 69	-0.22735
56 → 69	0.11227
58 → 67	0.11264
58 → 69	-0.23911
61 → 85	0.16005

Excited State 94: Singlet-A' 8.0901 eV 153.26 nm f= 0.0023

$<S^2>$ = 0.000

43 → 62	0.14735
45 → 62	0.23746
49 → 65	-0.14926
53 → 66	-0.16148
55 → 65	-0.10075
57 → 67	0.13291
57 → 69	0.10510
58 → 68	-0.16499
59 → 66	0.10753
59 → 67	-0.22687
59 → 69	0.35046
61 → 84	-0.15973

Excited State 95: Singlet-A" 8.1178 eV 152.73 nm f= 0.0196

$<S^2>$ = 0.000

44 → 62	0.32565
46 → 64	0.22477
47 → 63	0.20992
54 → 66	0.29060
54 → 69	-0.15081
56 → 67	-0.16943
56 → 69	-0.17495
57 → 68	-0.10389

Excited State 96: Singlet-A' 8.1180 eV 152.73 nm f= 0.0211

$<S^2>$ = 0.000

45 → 62	0.27265
46 → 63	-0.29030
47 → 64	-0.22741
49 → 65	0.30144
52 → 68	-0.12606
53 → 66	0.14711
59 → 66	0.12693
60 → 67	0.14457
61 → 84	0.16822

Excited State 97: Singlet-A" 8.1417 eV 152.28 nm f= 0.0016

$<S^2>$ = 0.000

48 → 65	-0.18774
56 → 66	0.20490
56 → 67	0.33158
56 → 69	0.13128
56 → 72	0.13993
57 → 68	-0.29693
57 → 71	-0.10720
58 → 66	0.11250
58 → 69	0.20475
60 → 70	0.14302

Excited State 98: Singlet-A' 8.1436 eV 152.25 nm f= 0.0141

$<S^2>$ = 0.000

43 → 62	0.42338
49 → 65	0.18914
56 → 68	-0.23695
57 → 67	-0.31050
57 → 69	-0.10413
58 → 68	0.13277

Excited State 99: Singlet-A' 8.1574 eV 151.99 nm f= 0.0072

$<S^2>$ = 0.000

43 → 62	-0.12196
45 → 62	-0.17981
46 → 63	0.19727
49 → 65	0.35184
56 → 68	0.10798
57 → 67	0.18337
57 → 69	-0.14148
59 → 69	0.19946
60 → 69	-0.14000
61 → 84	-0.26859

Excited State 100: Singlet-A" 8.1605 eV 151.93 nm f= 0.0035

$<S^2>$ = 0.000

44 → 62	0.11465
51 → 66	0.11605
52 → 66	-0.12169
52 → 69	0.10366
56 → 66	0.13768
56 → 67	0.34372
56 → 72	0.11880
57 → 68	0.36279
57 → 71	0.16274
60 → 71	-0.12170
60 → 77	-0.10169

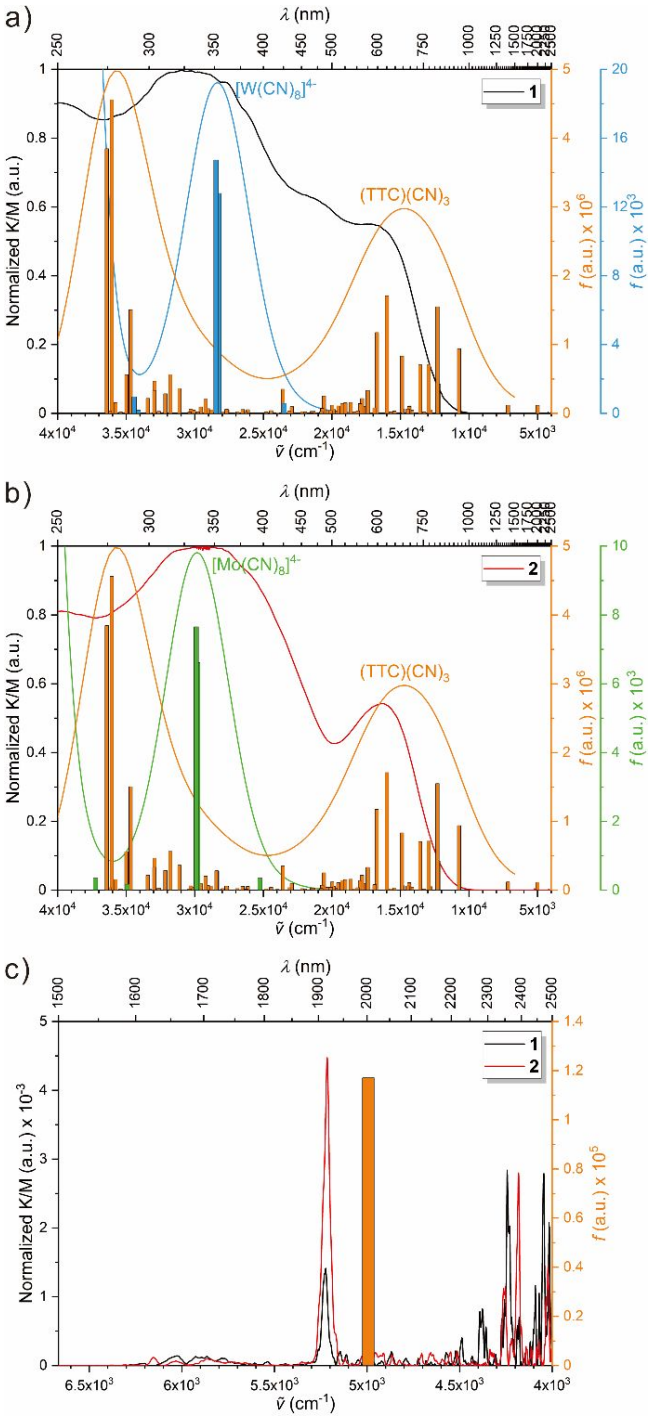

Figure S13. (a, b) Comparison of the measured solid-state UV–Vis–NIR absorption spectra for **1** (black) and **2** (red) and calculated for **[(TTC)(CN)$_3$]** (orange), **[W(CN)$_8$]$^{4-}$** (blue), and **[Mo(CN)$_8$]$^{4-}$** models (green). Bars correspond to the oscillator strength values. (c) Enlarged

spectra of **1** and **2** with absorption bands corresponding to the theoretically predicted HOMO–LUMO transition for β spins (orange bar).

5. Powder X-ray diffraction (PXRD) studies

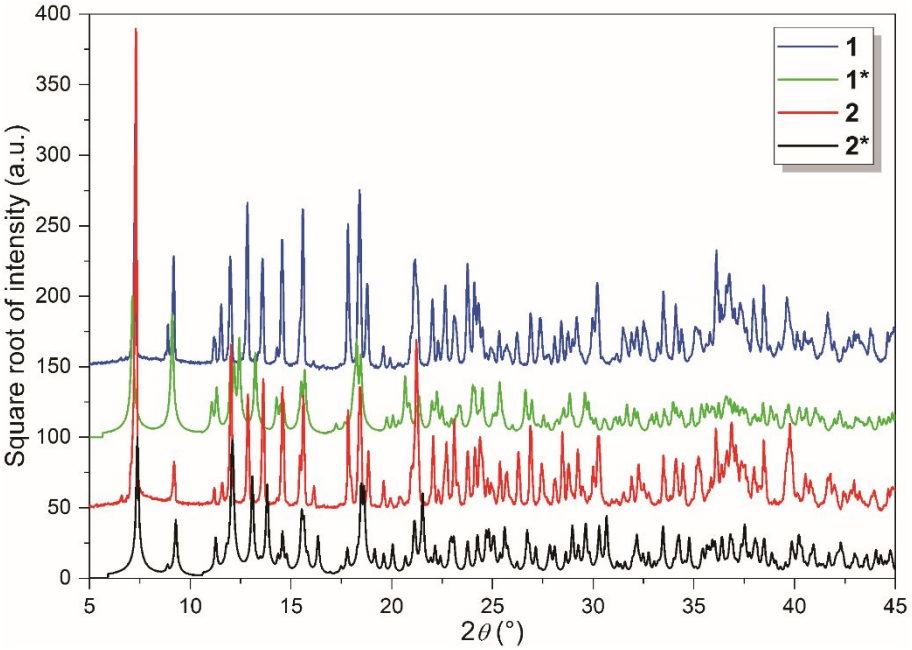

Figure S14. Experimental and simulated PXRD patterns based on the SXRD data for **1** and **2**.

6. Thermogravimetric analysis

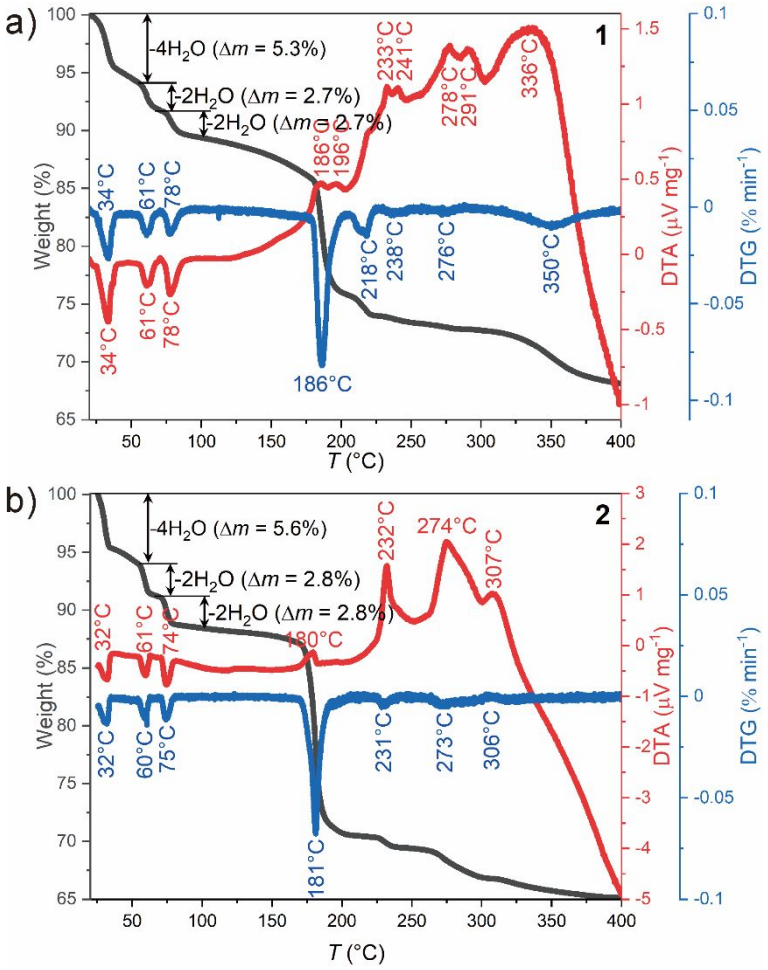

Figure S15. (a, b) Thermogravimetric plots for powdered samples of **1** and **2** with the weight losses due to the removal of solvent indicated.

7. References

S1. *Infrared and Raman Characteristic Group Frequencies: Tables and Charts, 3rd Edition*; Socrates, G., Ed.; John Wiley and Sons, Ltd, Chichester, 2001.

S2. Stefańczyk, O.; Majcher, A. M.; Rams, M.; Nitek, W.; Mathonière, C.; Sieklucka, B. Photo-induced magnetic properties of the $[Cu^{II}(bapa)]_2[Mo^{IV}(CN)_8]\cdot 7H_2O$ molecular ribbon. *J. Mater. Chem. C* 2015, *3*, 8712–8719.

S3. Korzeniak, T.; Sasmal, S.; Pinkowicz, D.; Nitek, W.; Pełka, R.; Czernia, D.; Stefańczyk, O.; Sieklucka, B. Chiral Photomagnets Based on Copper(II) complexes of 1,2-Diaminocyclohexane and Octacyanidomolybdate(IV) Ions. *Inorg. Chem.* 2020, *59*, 5872–5882.

S4. Zhang, W.; Sun, H.–L.; Sato, O. Synthesis, characterization, and photoresponsive properties of a series of Mo(IV)–Cu(II) complexes. *Dalton Trans.* 2011, *40*, 2735–2743.

S5. Zhang, W.; Sun, H.–L.; Sato, O. A one-dimensional homochiral Mo(IV)-Cu(II) coordination polymer: spontaneous resolution and photoresponsive properties. *CrystEngComm* 2010, *12*, 4045–4047.

S6. Korzeniak, T.; Sasmal, S.; Pinkowicz, D.; Sieklucka B. The photomagnetic effect in 2-D cyanido-bridged coordination polymer $[Cu(aepa)]_{10}[Mo(CN)_8]_5 \cdot 30H_2O$. *New J. Chem.* **2018**, *42*, 17009–17015.

S7. Zhang, W.; Sun, H.–L.; Sato, O. Synthesis, characterization, and photoresponsive properties of a series of Mo(IV)–Cu(II) complexes. Dalton *Trans.* **2011**, *40*, 2735–2743.

S8. Stefańczyk, O.; Ohkoshi, S. Synthesis of Two-Dimensional Photomagnetic $K_4\{[Cu^{II}(ida)]_2[M^{IV}(CN)_8]\} \cdot 4H_2O$ (M^{IV} = Mo, W) Materials. *Inorg. Chem.* **2020**, *59*, 4292–4299.

S9. Catala, L.; Mathonière, C.; Gloter, A.; Stephan, O.; Gacoin, T.; Boilot, J.–P.; Mallah, T. Photomagnetic nanorods of the $Mo(CN)_8Cu_2$ coordination network. *Chem. Commun.* **2005**, 746–748.

S10. Umeta, Y.; Chorazy, S.; Nakabayashi, K.; Ohkoshi, S. Synthesis of the Single–Crystalline Form and First–Principles Calculations of Photomagnetic Copper(II) Octacyanidomolybdate(IV). *Eur. J. Inorg. Chem.* **2016**, 1980–1988.

S11. Ohkoshi, S.; Machida, N.; Zhong, Z. J.; Hashimoto, K. Photo-induced magnetization in copper(II) octacyanomolybdate(IV). *Synth. Met.* **2001**, *122*, 523–527.

S12. Hozumi, T.; Hashimoto, K.; Ohkoshi, S. Electrochemical Synthesis, Crystal Structure, and Photomagnetic Properties of a Three-Dimensional Cyano-Bridged Copper−Molybdenum Complex. *J. Am. Chem. Soc.* **2005**, *127*, 3864–3869.

S13. Llunell, M.; Casanova, D.; Cirera, J.; Alemany, M. P.; Alvarez, S. *SHAPE,* v.2.0, University of Barcelona, Barcelona, Spain, **2010**.

S14. Alvarez, S.; Alemany, P.; Casanova, D.; Cirera, J.; Llunell, M.; Avnir, D. Shape maps and polyhedral interconversion paths in transition metal chemistry. *Coord. Chem. Rev.* **2005**, *249*, 1693–1708.

S15. Ginsberg, A. P. Magnetic Exchange in Transition Metal Complexes. 12. Calculation of Cluster Exchange Coupling Constants with the Xα-Scattered Wave Method. *J. Am. Chem. Soc.* **1980**, *102*, 111–117.

S16. Noodleman, L. Valence Bond Description of Antiferromagnetic Coupling in Transition Metal Dimers. *J. Chem. Phys.* **1981**, *74*, 5737–5743.

S17. Noodleman, L.; Davidson, E. R. Ligand Spin Polarization and Antiferromagnetic Coupling in Transition Metal Dimers. *Chem. Phys.* **1986**, *109*, 131–143.

S18. Bencini, A.; Gatteschi, D. X. Xα-SW calculations of the electronic structure and magnetic properties of weakly coupled transition–metal clusters. The$[Cu_2Cl_6]^{2-}$ dimers. *J. Am. Chem. Soc.* **1986**, *108*, 5763–5771.

S19. Yamaguchi, K.; Takahara, Y.; Fueno, T. *Ab-Initio Molecular Orbital Studies of Structure and Reactivity of Transition Metal-OXO Compounds*; Springer: 1986; pp 155–184.

S20. Soda, T.; Kitagawa, Y.; Onishi, T.; Takano, Y.; Shigeta, Y.; Nagao, H.; Yoshioka, Y.; Yamaguchi, K. Ab initio computations of effective exchange integrals for H–H, H–He–H and Mn_2O_2 complex: comparison of broken-symmetry approaches. *Chem. Phys. Lett.* **2000**, *319*, 223–230.