Supporting Information

Interligand Communication in a Metal Mediated LL’CT System – A Case Study
Sara A. Dille,[a] Kyle J. Colston,[a] Stephen C. Ratvasky,[b] Jingzhi Pu,[a] and Partha Basu*[a]

[a] Dr. S. A. Dille, Mr. K. J. Colston, Dr. J. Pu, and Dr. P. Basu
Department of Chemistry and Chemical Biology
Indiana University – Purdue University Indianapolis
Indianapolis, IN, 46202, USA
E-mail: basup@iupui.edu

[b] Mr S. C. Ratvasky
Department of Chemistry and Biochemistry
Duquesne University
Pittsburgh, PA, 15282, USA
Experimental Section

Materials. All reagents and solvents were purchased from either Sigma Aldrich or Thermo Fisher Scientific and used as received without further purification. All work was carried out under an inert atmosphere either in a dry box or using Schlenk line techniques under argon. [MoOCl((Pr2Dt0)2][PF6], [MoOCl(Me2Dt0)2][PF6]12, N,N'-diisopropylpiperazine-2,3-dithione ((Pr2Dt0), and N,N'-dimethylpiperazine-2,3-dithione (Me2Dt0)60 were synthesized following literature procedures. Multivariate regression analyses for Kamlet-Taft models were performed using Minitab 18 software.

Physical Methods. 1H NMR spectra were recorded on either a Bruker 500 MHz Avance spectrometer or a Bruker 400 MHz Avance spectrometer in air-tight NMR tubes.31P NMR spectra were recorded on a Bruker 400 MHz Avance spectrometer in air-tight NMR tubes. Infrared spectroscopy (FTIR) was recorded using a Thermofisher Nicolet iS10 spectrometer at room temperature using a KBr pellet. Electronic absorbance spectra were collected on a Shimadzu UV-3600 Plus in an air tight quartz cuvette. Cyclic voltammetry was recorded on a Metrohm PGSTAT204 galvanostat/potentiostat. A Pt disk working electrode, Ag+/Ag reference electrode and Pt wire auxillary electrode and tetrabutylammonium hexafluorophosphate supporting electrolyte were used. All voltammograms were referenced to the Fe+/Fc couple as an internal standard. All potentials are presented versus the Fe+/Fc couple, Mass spectra were collected using an Agilent Technologies 6520 Accurate Mass-QTOF LC/MS.

Computational Methods. All computational work was performed using Gaussian 09 software package running on UNIX OS and visualized utilizing GaussView 5.0.9.
Calculations were done using the Lee-Yang-Parr nonlocal correlation functional (B3LYP) and a combination of the LANL2DZ effective core potential basis set for molybdenum and the 6-31G** basis set for all other atoms. The crystal structure geometry was optimized using DFT to afford the geometry used for subsequent calculations. Atomic composition for molecular orbitals was determined using C-squared population analysis from single-point calculations with the program QM-Forge. The lowest 60 transition energies were generated using non-equilibrium TDDFT calculations with the polarizable continuum model (PCM) algorithm. PCM-TDDFT calculations were performed using acetonitrile as the solvent to match experimental conditions. Electron density difference maps (EDDMs) were generated using the cubman package in Gaussian09.

X-Ray Crystallography. Single crystals were mounted using glass fiber and data collected using a Bruker SMART Apex II diffractometer with a graphite monochromator for Mo Kα radiation (0.71073 Å). The absorption correction was performed using SADABS routine. The structure solution and the refinement were done using SHELXS-97 and SHEXLX-2018 programs. The X-ray data were collected at room temperature (296 K). Hydrogen atoms were placed at calculated positions and refined as riding atoms with isotropic displacement parameters. The methyl group on the toluene-2,3-dithiolene moiety of MoO(tdt)(PrDt⁰) was refined as disordered over two moieties with different rotational orientations. The disorder extends to the other carbon and sulfur atoms of the toluene-2,3-dithiolene moiety, and they were included in the disorder modeling. The two moieties were restrained to have similar geometries (SAME command of Shelxl) and U_ij components of ADPs for disordered atoms closer to each other than 2.0 Angstrom were restrained to be
similar. Subject to these conditions, the occupancy ratio refined to 0.517(5) to 0.483(5).

Details of the structure determination are listed in Table S1.

Table S1. Crystallographic Data for MoO(tdt)(iPr2Dt)

| Property                              | Value                                      |
|---------------------------------------|---------------------------------------------|
| Formula                              | C_{17}H_{24}MoN_{2}OS_{4}                   |
| Formula weight                       | 496.56                                      |
| Temperature                          | 296 K                                       |
| Color/shape                          | Purple/Block                               |
| Crystal system                       | Orthorhombic                               |
| Space group                          | Pbca                                        |
| Unit cell dimensions                 | a=12.6981 (1) Å                            |
|                                       | b=15.8799 (2) Å                            |
|                                       | c=21.2754 (2) Å                            |
| Volume (Å³)                          | 4290.07(8)                                 |
| Z, Formula unit/unit cell            | 8                                           |
| density (calculated)                 | 1.534 Mg m⁻³                               |
| μ (mm⁻¹)                             | 1.01                                        |
| Diffractometer                       | Bruker Smart Apex II                       |
| Radiation, graphite monochrome       | Mo Kα (λ=0.71073 Å)                        |
| Crystal size                         | 0.20 x 0.11 x 0.08 mm                      |
| Reflections collected/unique         | 70727/6665                                  |
| Parameters/restraints                | 314/292                                     |
| R_{int}                              | 0.054                                       |
| Refinement method                    | Full-matrix least-squares on F²            |
| Goodness-of-fit on F²                | 1.03                                        |
| Final R indices [I>2σ(I)]            | R=0.041 wR2=0.100                          |
| Maximum residue peaks (eÅ⁻³)         | 0.53 and -0.44                             |
Syntheses of Complexes

**MoO(bdt)(Me₂Dt₀)** (1). [MoOCl(Me₂Dt₀)₂][PF₆] (200 mg, 0.312 mmol) was dissolved in 9 mL of acetonitrile generating a dark blue solution. A solution of benzenedithiol (63 mg, 0.34 mmol) and triethylamine (69 mg, 0.68 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple and the release of white vapor. The reaction mixture was stirred for one hour. The solution was filtered, and dark purple solid was collected. The crude product was washed with cold chloroform to obtain analytically pure product. Yield 56% (75 mg, 0.175 mmol). Calcd (Expt) for C₁₂H₁₄MoN₂O₄S₄: C, 33.80 (33.95): H, 3.31 (3.34); N, 6.33 (6.57). ¹H NMR (CD₃CN): δ 3.77 (s, CH₃, 6H), 3.89 (m, CH₂, 2H), 4.18 (m, CH₂, 2H), 7.10 (dd, J = 5.8 Hz, 3.2 Hz, aromatic, 2H), 7.65 (dd, J = 5.8 Hz, 3.2 Hz, aromatic, 2H). FTIR (KBr, cm⁻¹): 1531 (vs, C(-N)S), 1354 (vs, C=S), 940 (s, Mo=O). UV-Vis (MeCN): λₘₐₓ (ε, M⁻¹cm⁻¹) 380 nm (1610), 532 nm (4400).

**MoO(tdt)(Me₂Dt₀)** (2). [MoOCl(Me₂Dt₀)₂][PF₆] (200 mg, 0.312 mmol) was dissolved in 9 mL of acetonitrile generating a dark blue solution. A solution of toluene-3,4-dithiol (53 mg, 0.34 mmol) and triethylamine (69 mg, 0.68 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple and the release of white vapor. The reaction mixture was stirred for one hour. The solution was filtered and dark purple solid was collected. The crude product was washed with cold chloroform to obtain analytically pure product. (79 mg, 0.164 mmol). Calcd (Expt) for C₁₃H₁₆MoN₂O₄S₄: C, 35.45 (35.44): H, 3.66 (3.74); N, 6.36 (6.24). ¹H NMR (CD₃CN): δ 2.34 (s, CH₃, 3H), 3.76 (s, CH₃, 6H), 3.86 (m, CH₂, 2H), 4.14 (m,
CH₂, 2H), 6.92 (d, J= 7.87 Hz, aromatic 1H), 7.47 (s, aromatic, 1H), 7.51 (d, J= 7.9 Hz, aromatic, 1H). FTIR (KBr, cm⁻¹): 1534 (vs, C(-N)S), 1355 (vs, C=S), 929 (s, Mo=O).

**UV-Vis (MeCN):**  \( \lambda_{\text{max}} \) (\(\varepsilon, M^{-1} \text{cm}^{-1}\)) 380 nm (2320), 531 nm (6050).

MoO(qdt)(Me₂Dt⁰) (3). [MoOCl(Me₂Dt⁰)₂][PF₆] (200 mg, 0.312 mmol) was dissolved in 9 mL of acetonitrile generating a dark blue solution. A solution of quinioxalinedithiol (66 mg, 0.34 mmol) and triethylamine (69 mg, 0.68 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple and the release of white vapor. The reaction mixture was stirred for one hour. The solution was filtered and dark purple solid was collected. The crude product was washed with cold chloroform to obtain analytically pure product. (20 mg, 0.0401 mmol). 11% Calcd (Expt) for C₁₄H₁₄MoN₄O₄S₄ +H₂O: C, 33.87(34.21); H, 3.25(2.82); N, 11.28 11.84).

**¹H NMR (CD₃CN):**  δ 3.84 (s, CH₃, 6H), 4.00 (q, J= 8.2 Hz, 7.0 Hz, CH₂, 2H), 4.21(q, J= 7.0 Hz, 6.4 Hz, CH₂, 2H), 7.63 (dd, J= 6.4 Hz, 3.4 Hz, aromatic, 2H), 7.93 (dd, J= 6.4 Hz, 3.4 Hz, aromatic, 2H). FTIR (KBr, cm⁻¹): 1524 (vs, C(-N)S), 1350 (vs, C=S), 955 (s, Mo=O). UV-Vis (MeCN):  \( \lambda_{\text{max}} \) (\(\varepsilon, M^{-1} \text{cm}^{-1}\)) 410 nm (6110), 548 nm (7450).

MoO(bdtCl₂)(Me₂Dt⁰) (4). [MoOCl(Me₂Dt⁰)₂][PF₆] (200 mg, 0.312 mmol) was dissolved in 9 mL of acetonitrile generating a dark blue solution. A solution of 3,6-dichloro-1,2-benzenedithiol (72 mg, 0.34 mmol) and triethylamine (69 mg, 0.68 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple. The reaction mixture was stirred for thirty minutes. The solution was filtered and dark purple solid was collected. The crude product was washed with cold chloroform to obtain analytically pure product. Yield 42% (65 mg,
0.132 mmol). Calcd (Expt) for C₁₂H₁₂Cl₂MoN₂OS₄: C, 29.10 (29.35): H, 2.44 (2.50); N, 5.66 (5.69). ¹H NMR (CD₃CN): δ 3.81 (s, CH₃, 6H), 4.10 (m, CH₂, 2H), 4.20 (m, CH₂, 2H), 7.12 (s, aromatic 2H). FTIR (KBr, cm⁻¹): 1527 (vs, C(-N)S), 1353 (vs, C=S), 926 (s, Mo=O). Electronic spectrum, UV-Vis (MeCN): λ<sub>max</sub> (ε, M⁻¹cm⁻¹) 385 nm (2050), 531 nm (4460).

**MoO(bdt)(Pr₂Dt⁰) (5).** [MoOCl(Pr₂Dt⁰)]²[PΦ₆] (200 mg, 0.26 mmol) was dissolved in 10.5 mL of acetonitrile generating a dark blue solution. A solution of benzenedithiol (49 mg, 0.34 mmol) and triethylamine (59 mg, 0.58 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple. The reaction mixture was stirred for one hour. The solution was filtered, and dark purple solid was collected. Free Pr₂Dt⁰ ligand was present in the crude product. To remove the free ligand, the crude product (80 mmol) was dissolved in 7.5 mL of CH₃Cl. MoCl₅ (10 mg, 0.037 mmol) was dissolved in 3 mL of MeOH generating HCl gas. The MoCl₅ was stirred until the cessation of the HCl gas was observed resulting in a green solution. The methanolic solution was added dropwise to the chloroform solution of the crude MoO(bdt)(Pr₂Dt⁰). The mixture was stirred for 1.5 hours and filtered. A dark purple filtrate of analytically pure complex was collected. Yield 41% (52 mg, 0.107 mmol) Calcd (Expt) for C₁₆H₂₂MoN₂OS₄: C, 39.82 (40.24): H, 4.60 (4.51); N, 5.81 (5.54). ¹H NMR (CD₃CN): δ 1.28 (d, J= 6.7 Hz, CH₃, 6H), 1.43 (d, J= 6.7 Hz, CH₃, 6H), 3.74 (m, CH₂, 2H), 4.02 (m, CH₂, 2H), 5.23 (h, J= 6.7 Hz, CH, 2H), 7.10 (dd, J= 5.8 Hz, 3.2 Hz, aromatic, 2H), 7.65 (dd, J= 5.8 Hz, 3.3 Hz, aromatic, 2H) FTIR (KBr, cm⁻¹): 1501 (vs, C(-N)S), 1350 (vs, C=S), 931 (s, Mo=O). Electronic spectrum, UV-Vis (MeCN): λ<sub>max</sub> (ε, M⁻¹cm⁻¹) 380 (2880), 529 nm (6900).
MoO(tdt)(Pr2Dt) (6). [MoOCl(Pr2Dt)][PF6] (200 mg, 0.26 mmol) was dissolved in 9 mL of acetonitrile generating a dark blue solution. A solution of toluene-3,4-dithiol (0.45 g, 0.00029 moles) and triethylamine (59 mg, 0.58 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple. The reaction mixture was stirred for one hour. The solution was filtered and dark purple solid was collected. The crude product was washed with cold chloroform resulting in analytically pure product. Yield 58% (77 mg, 0.155 mmol). Calcd (Expt) for C17H24MoN2O5S4: C, 41.12 (41.04); H, 4.87 (4.76); N, 5.64 (5.52). 1H NMR (CD2Cl2): δ 1.24 (d, CH3, J = 6.9 Hz, 6H), 1.43 (d, J = 6.8 Hz, CH3, 6H), 2.34 (s, CH3, 3H), 3.62 (m, CH2, 2H), 3.88 (m, CH2, 2H), 5.22 (h, J = 6.7 Hz, CH, 2H), 6.93 (d, J = 8.1 Hz, aromatic H), 7.47 (s, aromatic, H), 7.51 (d, J = 7.9 Hz, aromatic, H). FTIR (KBr, cm⁻¹): 1501 (vs, C(-N)S), 1350 (vs, C=S), 931 (s, Mo=O). UV-Vis (MeCN): λmax (ε, M⁻¹cm⁻¹) 380 (2880) 533 nm (7500).

MoO(qdt)(Pr2Dt) (7). [MoOCl(Pr2Dt)][PF6] (200 mg, 0.26 mmol) was dissolved in 13.5 mL of acetonitrile generating a dark blue solution. A solution of quinoxalinedithiol (56 mg, 0.29 mmol) and triethylamine (59 mg, 0.58 mmol) in 4.5 mL of methanol was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple. The reaction mixture was stirred for one hour. The solution was filtered and green solid was collected. The green solid was washed with CH3Cl to collect a dark purple filtrate. The solvent was dried en vacuo to obtain analytically pure complex. Yield 34% (49 mg, 0.091 mmol). Calcd (Expt) for C18H22MoN4O5S4 + CH3Cl: C, 40.27 (39.77): H, 4.63 (4.52); N, 9.89 (9.16). 1H NMR (CD3CN): δ 1.27 (d, J = 6.7 Hz, CH3, 6H), 1.41 (d, J = 6.7 Hz, CH3, 6H), 3.63 (m, CH2, 2H), 3.87 (m, CH2, 2H), 5.25 (h, J = 6.7 Hz, CH,
2H), 7.54 (dd, J= 6.4 Hz, 3.4 Hz, aromatic, 2H), 7.98 (dd, J= 6.3 Hz, 3.4 Hz, aromatic, 2H). FTIR (KBr, cm⁻¹): 1491 (vs, C(-N)S), 1351 (vs, C=S), 950 (s, Mo=O). UV-Vis (MeCN): λmax (ε, M⁻¹cm⁻¹) 400 nm (8240), 543 nm (7070).

**MoO(bdtCl₂)(iPr₂D₅) (8).** [MoOCl(iPr₂D₅)₂][PF₆] (200 mg, 0.26 mmol) was dissolved in 13.5 mL of acetonitrile generating a dark blue solution. A solution of 3,6-dichloro-1,2-benzenedithiol (61 mg, 0.29 mmol) and triethylamine (59 mg, 0.58 mmol) in 1.5 mL of acetonitrile was added dropwise to the dark blue solution initiating an instantaneous color change to dark purple. The reaction mixture was stirred for 15 minutes. The solution was filtered and dark purple solid was collected. The crude product was washed with cold chloroform to obtain analytically pure complex. Yield: 68% (100 mg, 0.181 mmol).

Calcd (Expt) for C₁₆H₂₀Cl₂MoN₂O₄S₄: C, 34.85 (34.65); H, 3.66 (3.67); N, 5.08 (5.08). ¹H NMR (CD₃CN): δ1.31 (d, J= 6.7 Hz, CH₃, 6H), 1.45 (d, J= 6.7 Hz, CH₃, 6H), 3.71 (m, CH₂, 2H), 4.01 (m, CH₂, 2H), 5.28 (h, J= 6.7 Hz, CH, 2H), 7.23 (s, aromatic 2H). FTIR (KBr, cm⁻¹): 1507 (vs, C(-N)S), 1356 (vs, C=S), 939 (s, Mo=O). UV-Vis (MeCN): λmax (ε, M⁻¹cm⁻¹) 390 nm (4070), 530 nm (9400).
Figure S1. A plot showing the linearity of the peak current ($i_p$) versus the square root of the scan rate ($v$) for complex 6 suggesting diffusion-controlled processes. Fit of the equations are given below.

**Oxidation**
- $i_p = 1.40E-6 (+/- 6.36E-8)n^{3/2}A^{1/2}D^{1/2}C^b - 8.71E-7 (+/- 1.27E-6)$; $r^2 = 0.99$
- $i_p = 5.63E-7 (+/- 7.84E-8) n^{3/2}A^{1/2}D^{1/2}C^b - 8.56E-7 (+/- 1.57E-6)$; $r^2 = 0.93$

**Reduction**
- $i_p = -1.44E-6 (+/- 1.91E-8) n^{3/2}A^{1/2}D^{1/2}C^b - 2.12E-6 (+/- 3.82E-7)$; $r^2 = 0.99$
- $i_p = -1.60E-6 (+/- 3.78E-8) n^{3/2}A^{1/2}D^{1/2}C^b - 3.44E-6 (+/- 7.56E-7)$; $r^2 = 0.99$
Figure S2 Cyclic voltammogram of $^{1}\text{Pr}_2\text{D}t^0$. Scan rate, 100mV s$^{-1}$; solvent, acetonitrile; temperature, 25°C; Glassy carbon working electrode, Ag/Ag$^{+}$ reference electrode, and a Pt-wire auxiliary electrode; supporting electrolyte, $^{1}$BuNPF$_6$. Potentials referenced internally to Fc$^{+}$/Fc couple.
Figure S3. Cyclic voltammogram of Zn(mnt)(iPr_2Dt_0) at varying scan rates; solvent, acetonitrile; temperature, 25°C; Platinum disk working electrode, Ag/Ag^+ reference electrode, and a Pt-wire auxiliary electrode; supporting electrolyte, Bu_4NPF_6. Potentials referenced internally to Fc^+/Fc couple. The two poorly defined couples are due to potential dissociation of the iPr_2Dt_0 ligand. Support for this suggestion comes from multiple scan rate experiment. As scan rate increases, the couple at E_{1/2} = -1556 mV (ΔE_p = 90 mV) is no longer observed. The peak height of the two reversible couples attributed to the coordinated ligand increase with increasing scan rate whereas the peak heights of the couple at E_{1/2} = -1556 mV (ΔE_p = 90 mV) does not increase. At a scan rate of 100 mV/sec the peak heights -1923 mV is ~50% and at a scan rate of 900 mV/sec the peak height at -1923 mV is ~8% of the peak height at -1601 mV. Dissociation of the ligand is a slow electron process and the concentration of free ligand can be decreased by increasing the scan rate.
Electronic Spectra

Figure S4. Absorbance spectra for complexes possessing the Me₂Dt⁰ dithione ligand in acetonitrile.

Figure S5. Absorbance spectra for complexes possessing the Pr₂Dt⁰ dithione ligand in acetonitrile.
Figure S6. Absorbance spectra of 3 and 7 in acetonitrile.
Solvatochromic Effect
Figure S7. Linear correlation between dipole moment ($\mu$) and the energy of the LL’CT of MoO(bdt)(Me$_2$Dt$^0$). Equation of fit: $E = -518.86\mu + 20432 \ R^2 = 0.97$

Figure S8. Linear correlation between dipole moment ($\mu$) and the energy of the LL’CT of MoO(tdt)(Me$_2$Dt$^0$). Equation of fit: $E = -739.58\mu + 21146 \ R^2 = 0.93$
Figure S9. Linear correlation between dipole moment ($\mu$) and the energy of the LL’CT of MoO(bdtCl$_2$)(Me$_2$Dt$_0$). Equation of fit: $E = -397.27 \mu + 20038$ $R^2 = 0.98$

Figure S10. Linear correlation between dipole moment ($\mu$) and the energy of the LL’CT of MoO(bdt)(iPr$_2$Dt$^0$). Equation of fit: $E = -399.03 \mu + 20108$ $R^2 = 0.92$
Figure S11. Linear correlation between dipole moment ($\mu$) and the energy of the LL’CT of MoO(tdt)(iPr$_2$Dt$^0$). Equation of fit: $E = -610.17\mu + 20884$ $R^2 = 0.90$

Figure S12. Linear correlation between dipole moment ($\mu$) and the energy of the LL’CT of MoO(bdtCl$_2$)(iPr$_2$Dt$^0$). Equation of fit: $E = -338.3\mu + 19833$ $R^2 = 0.93$
Table S2. Composition of the molecular orbitals in 6.

| Orbital | E, eV | Mo | Mo(d) | O | tdt | S<sub>tdt</sub> | iPr<sub>2</sub>Dt<sup>0</sup> | S<sup>Dt</sup> |
|---------|-------|----|-------|---|-----|------------|----------------|-------------|
| L+2     | -1.05 | 48.47 | 39.81 | 10.24 | 17.89 | 16.34 | 23.40 | 17.21 |
| L+1     | -1.41 | 42.55 | 35.40 | 9.71 | 9.32 | 7.67 | 38.42 | 14.78 |
| LUMO    | -2.81 | 25.82 | 25.37 | 1.70 | 3.08 | 1.56 | 69.40 | 22.97 |
| HOMO    | -4.91 | 7.46 | 4.16 | 5.63 | 82.26 | 39.79 | 4.66 | 1.60 |
| H-1     | -5.33 | 1.36 | 1.05 | 1.99 | 87.81 | 49.26 | 8.84 | 6.62 |
| H-2     | -5.64 | 46.93 | 44.00 | 0.11 | 13.71 | 8.27 | 39.25 | 6.25 |

Table S3. Dt<sup>0</sup> fold angles (°) of 2, 5, 6, 7, and 8 calculated from gas phase optimizations. Compounds without crystallographic data were generated by editing 6 in GaussView6.0 to change Dt<sup>2-</sup> and Dt<sup>0</sup> substituents.

| Complex                                         | Dt<sup>0</sup> Fold Angle (°) |
|------------------------------------------------|-----------------------------|
| MoO(tdt)(Me<sub>2</sub>Dt<sup>0</sup>) (2)      | 65.93                       |
| MoO(bdt)(iPr<sub>2</sub>Dt<sup>0</sup>) (5)      | 70.53                       |
| MoO(tdt)(iPr<sub>2</sub>Dt<sup>0</sup>) (6)      | 70.47                       |
| MoO(bdtCl<sub>2</sub>)(iPr<sub>2</sub>Dt<sup>0</sup>) (7) | 69.85                       |
| MoO(qdt)(iPr<sub>2</sub>Dt<sup>0</sup>) (8)      | 69.58                       |

Table S4. Equation of the fit for the multivariate regression using the Kamlet-Taft model.

| Complex                                         | Equation of Fit               |
|------------------------------------------------|------------------------------|
| MoO(bdt)(Me<sub>2</sub>Dt<sup>0</sup>) (1)      | 23726+1514 $\pi$– 1675 $\alpha$ - 9347 $\beta$ |
| MoO(tdt)(Me<sub>2</sub>Dt<sup>0</sup>) (2)      | 26752+1785 $\pi$– 27025 $\alpha$ - 1406 $\beta$ |
| MoO(bdtCl<sub>2</sub>)( Me<sub>2</sub>Dt<sup>0</sup>)(4) | 20777-3031 $\pi$+ 425.9 $\alpha$ + 606 $\beta$ |
| MoO(bdt)(iPr<sub>2</sub>Dt<sup>0</sup>) (5)      | 23041+366 $\pi$– 12720 $\alpha$ - 6794 $\beta$ |
| MoO(tdt)(iPr<sub>2</sub>Dt<sup>0</sup>) (6)      | 25891+1336 $\pi$– 23442 $\alpha$ - 11962 $\beta$ |
| MoO(bdtCl<sub>2</sub>)(iPr<sub>2</sub>Dt<sup>0</sup>) (8) | 21829-104 $\pi$– 7887 $\alpha$ - 4563 $\beta$ |
Table S5. Correlation coefficients of peak maxima fit to the Kamlet-Taft model.

| Complex                          | Dipole Moment $R^2$ | Kamlet-Taft $R^2$ |
|---------------------------------|---------------------|-------------------|
| MoO(bdt)(Me$_2$Dt$^0$) (1)      | 0.97                | 0.98              |
| MoO(tdt)(Me$_2$Dt$^0$) (2)      | 0.93                | 0.94              |
| MoO(bdtCl$_2$(Me$_2$Dt$^0$) (4) | 0.98                | 0.98              |
| MoO(bdt)(iPr$_2$Dt$^0$) (5)     | 0.92                | 0.96              |
| MoO(tdt)(iPr$_2$Dt$^0$) (6)     | 0.90                | 0.97              |
| MoO(bdtCl$_2$(iPr$_2$Dt$^0$) (8)| 0.93                | 0.99              |

Theoretical Calculations

Charge = 0 Multiplicity = 1

| Element | Mo     | C      | N      | S      | O      | S      | C      | C      | H      | H      | S      | C      | C      | H      | H      | H      | H      | H      | H      |
|---------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| Mo      | -0.432 | 0.003  | 0.128  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | -3.633 | -1.017 | 0.206  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| N       | 3.519  | -1.183 | -0.197 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| S       | -2.028 | -1.772 | 0.311  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| O       | -0.02  | 0.464  | 1.72   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| S       | -2.327 | 1.303  | -0.534 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | -3.764 | 0.33   | -0.151 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| N       | 2.922  | 1.551  | 0.052  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | -5.04  | 0.908  | -0.239 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| H       | -5.126 | 1.955  | -0.517 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| S       | 0.967  | -1.799 | -0.81  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | -6.191 | 0.164  | 0.024  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| S       | 0.729  | 1.37   | -1.528 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | -6.045 | -1.188 | 0.377  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | 3.845  | -2.633 | -0.168 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| H       | 3.062  | -3.121 | -0.75  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | -4.786 | -1.772 | 0.468  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| H       | -4.687 | -2.819 | 0.742  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| C       | 3.953  | 3.772  | -0.375 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| H       | 3.752  | 4.848  | -0.362 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| H       | 4.174  | 3.479  | -1.405 |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
| H       | 4.849  | 3.604  | 0.234  |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |
Excitation energies and oscillator strengths:

Excited State  1:  Singlet-A  1.9465 eV  636.95 nm  f=0.0115  <S**2>=0.000
113 ->114  0.70049
This state for optimization and/or second-order correction.
Total Energy, E(TD-HF/TD-KS) = -2507.39909459
Copying the excited state density for this state as the 1-particle RhoCI density.

Excited State  2:  Singlet-A  2.3850 eV  519.84 nm  f=0.0091  <S**2>=0.000
111 ->114  0.62758
112 ->114  -0.30273

Excited State  3:  Singlet-A  2.4944 eV  497.06 nm  f=0.1752  <S**2>=0.000
111 ->114  0.30744
112 ->114  0.56780
112 ->115  -0.14893
112 ->116  -0.17355
| Excited State | Singlet-A | E (eV) | λ (nm) | f | <S**2> |
|---------------|-----------|--------|--------|---|--------|
| 4             | 2.8406    | 436.47 | 0.0146 | 0.000 |
| 112 -> 114    | 0.10439   |
| 112 -> 115    | 0.56066   |
| 112 -> 116    | -0.15536  |
| 113 -> 115    | -0.33777  |
| 5             | 3.0308    | 409.08 | 0.0034 | 0.000 |
| 112 -> 115    | 0.33978   |
| 113 -> 115    | 0.59747   |
| 6             | 3.0657    | 404.43 | 0.0046 | 0.000 |
| 109 -> 114    | 0.17121   |
| 110 -> 114    | 0.64063   |
| 112 -> 116    | 0.18533   |
| 7             | 3.2085    | 386.43 | 0.1255 | 0.000 |
| 110 -> 114    | -0.18069  |
| 112 -> 116    | 0.22461   |
| 112 -> 115    | 0.11939   |
| 113 -> 116    | -0.25749  |
| 8             | 3.3153    | 373.98 | 0.0232 | 0.000 |
| 111 -> 115    | 0.15594   |
| 112 -> 116    | 0.28653   |
| 113 -> 116    | 0.61029   |
| 9             | 3.4525    | 359.11 | 0.0056 | 0.000 |
| 109 -> 114    | 0.12833   |
| 112 -> 117    | 0.55843   |
| 112 -> 118    | 0.24788   |
| 113 -> 117    | -0.26725  |
| 10            | 3.4706    | 357.25 | 0.0032 | 0.000 |
| 109 -> 114    | 0.66222   |
| 110 -> 114    | -0.17874  |
| 11           | 3.5612    | 348.15 | 0.0651 | 0.000 |
| 111 -> 115    | 0.64750   |
| 113 -> 116    | -0.10787  |
| 113 -> 117    | 0.14787   |
| 12            | 3.6969    | 335.37 | 0.0319 | 0.000 |
| 111 -> 115    | -0.11913  |
| 111 -> 116    | -0.18849  |
| 112 -> 117    | 0.24848   |

22
112 -> 118  0.11893
113 -> 117  0.56256
113 -> 118  0.17253

Excited State 13:  Singlet-A  3.8104 eV  325.39 nm  f=0.0194  <S**2>=0.000
105 -> 114  -0.17815
107 -> 114  0.53624
111 -> 116  0.37740

Excited State 14:  Singlet-A  3.8181 eV  324.73 nm  f=0.0181  <S**2>=0.000
105 -> 114  0.12181
107 -> 114  -0.36016
111 -> 116  0.53083
113 -> 117  0.14213
113 -> 118  0.16120

Excited State 15:  Singlet-A  3.9125 eV  316.89 nm  f=0.0544  <S**2>=0.000
108 -> 114  -0.10789
112 -> 117  -0.22713
112 -> 118  0.50035
113 -> 117  0.11114
113 -> 118  -0.32321

Excited State 16:  Singlet-A  3.9883 eV  310.87 nm  f=0.0213  <S**2>=0.000
106 -> 114  -0.31306
108 -> 114  0.55191
110 -> 115  -0.16152
112 -> 118  0.18408

Excited State 17:  Singlet-A  4.0079 eV  309.35 nm  f=0.0178  <S**2>=0.000
106 -> 114  0.51169
108 -> 114  0.39698
110 -> 115  0.17722
112 -> 118  -0.10505

Excited State 18:  Singlet-A  4.0864 eV  303.40 nm  f=0.0936  <S**2>=0.000
106 -> 114  0.13289
110 -> 115  0.11047
111 -> 117  0.11717
112 -> 117  -0.12638
112 -> 118  0.27889
113 -> 117  -0.16113
113 -> 118  0.52994

Excited State 19:  Singlet-A  4.1768 eV  296.84 nm  f=0.0248  <S**2>=0.000
111 -> 117  0.65572
Excited State 20: Singlet-A 4.2372 eV 292.61 nm f=0.0116 \ <S^2>=0.000
104 ->114 0.21900
106 ->114 -0.20315
110 ->115 0.59258

Excited State 21: Singlet-A 4.3018 eV 288.21 nm f=0.0041 \ <S^2>=0.000
103 ->114 -0.11423
104 ->114 -0.12329
105 ->114 0.61661
106 ->114 -0.11980
107 ->114 0.24420

Excited State 22: Singlet-A 4.3662 eV 283.97 nm f=0.0326 \ <S^2>=0.000
104 ->114 0.62972
105 ->114 0.15770
106 ->114 0.13554
110 ->115 -0.16634

Excited State 23: Singlet-A 4.4504 eV 278.59 nm f=0.0073 \ <S^2>=0.000
109 ->115 0.13758
110 ->116 0.67425

Excited State 24: Singlet-A 4.4871 eV 276.31 nm f=0.0033 \ <S^2>=0.000
111 ->117 -0.11582
111 ->118 0.64608
113 ->119 -0.12271

Excited State 25: Singlet-A 4.5981 eV 269.64 nm f=0.0558 \ <S^2>=0.000
103 ->114 0.56617
105 ->114 0.14260
107 ->115 0.11061
109 ->115 0.24862
113 ->119 0.14037

Excited State 26: Singlet-A 4.6295 eV 267.81 nm f=0.0029 \ <S^2>=0.000
103 ->114 -0.31111
109 ->115 0.55523
113 ->119 0.13118

Excited State 27: Singlet-A 4.6418 eV 267.10 nm f=0.0615 \ <S^2>=0.000
109 ->115 -0.14256
111 ->118 0.10484
111 ->121 0.11979
112 ->119 -0.10786
| Transition | Excited State | Energy (eV) | Wavelength (nm) | Oscillator Strength (f) | Squared dipole moment ($<S^2>$) |
|------------|---------------|-------------|-----------------|-------------------------|-------------------------------|
| 113 -> 119 | Excited State 28: Singlet-A | 4.7577 | 260.59 | 0.0236 | 0.000 |
| 109 -> 116 | 0.27602 |
| 111 -> 121 | 0.16228 |
| 112 -> 119 | 0.46782 |
| 113 -> 120 | -0.34471 |
| 109 -> 116 | Excited State 29: Singlet-A | 4.7731 | 259.76 | 0.0089 | 0.000 |
| 107 -> 115 | 0.21167 |
| 109 -> 116 | 0.51861 |
| 112 -> 119 | -0.31514 |
| 113 -> 119 | -0.10042 |
| 109 -> 116 | Excited State 30: Singlet-A | 4.8107 | 257.73 | 0.0291 | 0.000 |
| 111 -> 121 | 0.01900 |
| 111 -> 119 | -0.20458 |
| 113 -> 119 | 0.14494 |
| 113 -> 120 | 0.47616 |
| 113 -> 121 | -0.12391 |
| 113 -> 121 | 0.11561 |
| 110 -> 117 | Excited State 31: Singlet-A | 4.8633 | 254.94 | 0.0077 | 0.000 |
| 112 -> 119 | 0.13687 |
| 113 -> 121 | 0.11108 |
| 110 -> 117 | 0.67121 |
| 112 -> 119 | Excited State 32: Singlet-A | 4.8841 | 253.85 | 0.0383 | 0.000 |
| 110 -> 118 | 0.15455 |
| 109 -> 115 | 0.50334 |
| 107 -> 115 | -0.22939 |
| 108 -> 115 | -0.30955 |
| 110 -> 118 | 0.11500 |
| 111 -> 119 | 0.34877 |
| 107 -> 115 | 0.49262 |
| 107 -> 116 | -0.14406 |
| 112 -> 119 | 0.13687 |
| 113 -> 121 | 0.11108 |
| 110 -> 118 | 0.15455 |
| 106 -> 115 | 0.50334 |
| 107 -> 115 | -0.22939 |
| 108 -> 115 | -0.30955 |
| 110 -> 118 | 0.15050 |
| 111 -> 119 | 0.52546 |
| 113 -> 121 | -0.34854 |
| Excited State | Configuration | Energy (eV) | Wavelength (nm) | Oscillator Strength (f) | Square of Dipole Moment (S**2) |
|---------------|---------------|------------|----------------|------------------------|-------------------------------|
| 35: Singlet-A  | 106 \rightarrow 115 | 5.1479     | 240.85         | 0.0589                 | 0.000                         |
|               | 107 \rightarrow 115 | -0.14084   |                |                        |                               |
|               | 108 \rightarrow 115 | 0.45922    |                |                        |                               |
|               | 110 \rightarrow 118 | -0.22321   |                |                        |                               |
|               | 111 \rightarrow 119 | 0.10786    |                |                        |                               |
|               | 113 \rightarrow 121 | 0.27042    |                |                        |                               |
| 36: Singlet-A  | 106 \rightarrow 115 | 5.1506     | 240.72         | 0.0061                 | 0.000                         |
|               | 106 \rightarrow 116 | -0.17265   |                |                        |                               |
|               | 108 \rightarrow 115 | 0.23890    |                |                        |                               |
| 37: Singlet-A  | 106 \rightarrow 116 | 5.2094     | 238.00         | 0.0348                 | 0.000                         |
|               | 107 \rightarrow 116 | 0.31467    |                |                        |                               |
|               | 108 \rightarrow 115 | 0.23890    |                |                        |                               |
|               | 109 \rightarrow 118 | -0.12356   |                |                        |                               |
|               | 110 \rightarrow 118 | 0.56891    |                |                        |                               |
| 38: Singlet-A  | 106 \rightarrow 116 | 5.2194     | 237.54         | 0.0081                 | 0.000                         |
|               | 107 \rightarrow 116 | -0.15554   |                |                        |                               |
|               | 108 \rightarrow 115 | 0.010114   |                |                        |                               |
|               | 109 \rightarrow 117 | 0.20857    |                |                        |                               |
|               | 111 \rightarrow 119 | -0.12246   |                |                        |                               |
|               | 112 \rightarrow 120 | 0.30144    |                |                        |                               |
| 39: Singlet-A  | 107 \rightarrow 116 | 5.2623     | 235.61         | 0.0072                 | 0.000                         |
|               | 108 \rightarrow 116 | 0.11285    |                |                        |                               |
|               | 109 \rightarrow 117 | 0.13057    |                |                        |                               |
|               | 111 \rightarrow 120 | 0.10582    |                |                        |                               |
|               | 112 \rightarrow 119 | -0.12338   |                |                        |                               |
|               | 112 \rightarrow 120 | 0.60004    |                |                        |                               |
|               | 113 \rightarrow 120 | -0.10003   |                |                        |                               |
| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 40            | 5.2988    | 233.99      | 0.0160         | 0.000 |
| 104 -&gt; 115 | 0.15956   |             |                |     |
| 105 -&gt; 116 | 0.15174   |             |                |     |
| 106 -&gt; 116 | 0.42719   |             |                |     |
| 107 -&gt; 116 | -0.27396  |             |                |     |
| 108 -&gt; 116 | -0.29159  |             |                |     |
| 109 -&gt; 117 | -0.13034  |             |                |     |
| 110 -&gt; 118 | 0.11539   |             |                |     |
| 112 -&gt; 120 | 0.18514   |             |                |     |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 41            | 5.3569    | 231.45      | 0.0043         | 0.000 |
| 106 -&gt; 116 | 0.23066   |             |                |     |
| 108 -&gt; 116 | 0.32084   |             |                |     |
| 111 -&gt; 120 | 0.20483   |             |                |     |
| 112 -&gt; 120 | -0.10581  |             |                |     |
| 112 -&gt; 121 | 0.48865   |             |                |     |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 42            | 5.3827    | 230.34      | 0.0046         | 0.000 |
| 106 -&gt; 116 | 0.19008   |             |                |     |
| 107 -&gt; 117 | -0.11211  |             |                |     |
| 108 -&gt; 116 | 0.47604   |             |                |     |
| 112 -&gt; 121 | -0.39969  |             |                |     |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 43            | 5.4083    | 229.25      | 0.1751         | 0.000 |
| 104 -&gt; 115 | 0.38268   |             |                |     |
| 105 -&gt; 115 | 0.10464   |             |                |     |
| 106 -&gt; 116 | -0.16401  |             |                |     |
| 107 -&gt; 116 | -0.17642  |             |                |     |
| 107 -&gt; 117 | -0.11803  |             |                |     |
| 109 -&gt; 117 | 0.31810   |             |                |     |
| 109 -&gt; 118 | -0.28836  |             |                |     |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 44            | 5.4326    | 228.22      | 0.0101         | 0.000 |
| 102 -&gt; 114 | 0.16363   |             |                |     |
| 105 -&gt; 115 | 0.57934   |             |                |     |
| 107 -&gt; 115 | 0.20014   |             |                |     |
| 109 -&gt; 117 | -0.11005  |             |                |     |
| 111 -&gt; 120 | -0.11958  |             |                |     |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 45            | 5.4489    | 227.54      | 0.0006         | 0.000 |
| 102 -&gt; 114 | 0.64540   |             |                |     |
| 105 -&gt; 115 | -0.12692  |             |                |     |
| 111 -&gt; 120 | 0.16194   |             |                |     |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | f | <S^2> |
|---------------|-----------|-------------|----------------|---|-------|
| 46            | 5.4807    | 226.22      | 0.0124         | 0.000 |
| 102 -&gt; 114 | 0.64540   |             |                |     |
| 105 -&gt; 115 | -0.12692  |             |                |     |
| 111 -&gt; 120 | 0.16194   |             |                |     |
| Transition | E (eV) | f | \(<S^2>\) |
|------------|--------|---|-----------|
| 100 -> 114 | -0.23961 | 0.0621 | 0.000 |
| 101 -> 114 | 0.19987 | | |
| 102 -> 114 | 0.13808 | | |
| 103 -> 115 | -0.10328 | | |
| 104 -> 115 | -0.19897 | | |
| 109 -> 118 | -0.18857 | | |
| 111 -> 120 | -0.10507 | | |
| 111 -> 121 | 0.16781 | | |
| 113 -> 122 | 0.43012 | | |

**Excited State 47:** Singlet-A 5.5015 eV 225.37 nm f=0.0621 \(<S^2>\)=0.000

| Transition | E (eV) | f | \(<S^2>\) |
|------------|--------|---|-----------|
| 100 -> 114 | 0.23975 | | |
| 101 -> 114 | -0.17767 | | |
| 103 -> 115 | 0.12739 | | |
| 106 -> 117 | -0.10970 | | |
| 107 -> 116 | 0.16829 | | |
| 109 -> 118 | 0.13177 | | |
| 111 -> 120 | 0.21300 | | |
| 111 -> 121 | 0.10324 | | |
| 112 -> 122 | -0.10107 | | |
| 113 -> 122 | 0.42764 | | |

**Excited State 48:** Singlet-A 5.5346 eV 224.02 nm f=0.0222 \(<S^2>\)=0.000

| Transition | E (eV) | f | \(<S^2>\) |
|------------|--------|---|-----------|
| 104 -> 115 | -0.26849 | | |
| 105 -> 115 | 0.16586 | | |
| 107 -> 117 | -0.22299 | | |
| 108 -> 116 | -0.11954 | | |
| 111 -> 120 | 0.41425 | | |
| 112 -> 121 | -0.14051 | | |
| 113 -> 121 | -0.13108 | | |
| 113 -> 122 | -0.11303 | | |
| 113 -> 123 | 0.10605 | | |

**Excited State 49:** Singlet-A 5.5548 eV 223.20 nm f=0.0278 \(<S^2>\)=0.000

| Transition | E (eV) | f | \(<S^2>\) |
|------------|--------|---|-----------|
| 100 -> 114 | -0.24516 | | |
| 101 -> 114 | 0.29052 | | |
| 104 -> 115 | 0.34898 | | |
| 107 -> 116 | 0.10953 | | |
| 107 -> 117 | 0.12770 | | |
| 110 -> 119 | -0.14344 | | |
| 111 -> 120 | 0.24071 | | |
| 113 -> 121 | -0.10523 | | |

**Excited State 50:** Singlet-A 5.5971 eV 221.52 nm f=0.3068 \(<S^2>\)=0.000

| Transition | E (eV) | f | \(<S^2>\) |
|------------|--------|---|-----------|
| 105 -> 116 | -0.23973 | | |
| 107 -> 117 | 0.16790 | | |
| Transition | 110 -> 119 | 111 -> 121 | 113 -> 120 | 113 -> 122 | 113 -> 123 |
|------------|------------|------------|------------|------------|------------|
| 110 -> 119 | 0.15418    |            |            |            |            |
| 111 -> 121 | 0.34851    |            |            |            |            |
| 113 -> 120 | 0.15887    |            |            |            |            |
| 113 -> 122 | -0.13031   |            |            |            |            |
| 113 -> 123 | 0.38273    |            |            |            |            |

Excited State 51: Singlet-A 5.6245 eV 220.44 nm $f=0.0377$ $<S^2>=0.000$

| Transition | 105 -> 116 | 107 -> 117 | 110 -> 119 | 111 -> 121 | 112 -> 122 | 113 -> 120 | 113 -> 122 | 113 -> 123 | 113 -> 124 |
|------------|------------|------------|------------|------------|------------|------------|------------|------------|------------|
| 105 -> 116 | 0.27856    |            |            |            |            |            |            |            |            |
| 107 -> 117 | 0.25760    |            |            |            |            |            |            |            |            |
| 110 -> 119 | 0.25134    |            |            |            |            |            |            |            |            |
| 111 -> 121 | -0.19794   |            |            |            |            |            |            |            |            |
| 112 -> 122 | -0.16703   |            |            |            |            |            |            |            |            |
| 113 -> 120 | -0.10374   |            |            |            |            |            |            |            |            |
| 113 -> 122 | 0.11229    |            |            |            |            |            |            |            |            |
| 113 -> 123 | 0.29587    |            |            |            |            |            |            |            |            |
| 113 -> 124 | 0.21572    |            |            |            |            |            |            |            |            |

Excited State 52: Singlet-A 5.6343 eV 220.05 nm $f=0.0832$ $<S^2>=0.000$

| Transition | 103 -> 115 | 105 -> 116 | 107 -> 117 | 110 -> 119 | 111 -> 120 | 113 -> 123 | 113 -> 124 |
|------------|------------|------------|------------|------------|------------|------------|------------|
| 103 -> 115 | -0.13358   |            |            |            |            |            |            |
| 105 -> 116 | -0.10829   |            |            |            |            |            |            |
| 107 -> 117 | 0.19326    |            |            |            |            |            |            |
| 110 -> 119 | 0.43097    |            |            |            |            |            |            |
| 111 -> 120 | 0.10085    |            |            |            |            |            |            |
| 113 -> 123 | -0.37643   |            |            |            |            |            |            |
| 113 -> 124 | -0.15653   |            |            |            |            |            |            |

Excited State 53: Singlet-A 5.6411 eV 219.79 nm $f=0.3031$ $<S^2>=0.000$

| Transition | 103 -> 115 | 105 -> 116 | 109 -> 117 | 109 -> 118 | 111 -> 121 | 112 -> 122 | 112 -> 125 | 113 -> 122 | 113 -> 124 |
|------------|------------|------------|------------|------------|------------|------------|------------|------------|------------|
| 103 -> 115 | -0.13789   |            |            |            |            |            |            |            |            |
| 105 -> 116 | 0.22921    |            |            |            |            |            |            |            |            |
| 109 -> 117 | 0.16348    |            |            |            |            |            |            |            |            |
| 109 -> 118 | 0.37368    |            |            |            |            |            |            |            |            |
| 111 -> 121 | 0.13076    |            |            |            |            |            |            |            |            |
| 112 -> 122 | 0.28176    |            |            |            |            |            |            |            |            |
| 112 -> 125 | -0.10729   |            |            |            |            |            |            |            |            |
| 113 -> 122 | 0.10099    |            |            |            |            |            |            |            |            |
| 113 -> 124 | 0.21434    |            |            |            |            |            |            |            |            |

Excited State 54: Singlet-A 5.6804 eV 218.27 nm $f=0.0302$ $<S^2>=0.000$

| Transition | 104 -> 116 | 105 -> 116 | 107 -> 116 | 107 -> 117 | 109 -> 118 | 112 -> 122 | 113 -> 124 |
|------------|------------|------------|------------|------------|------------|------------|------------|
| 104 -> 116 | 0.19742    |            |            |            |            |            |            |
| 105 -> 116 | 0.27118    |            |            |            |            |            |            |
| 107 -> 116 | 0.11384    |            |            |            |            |            |            |
| 107 -> 117 | 0.16546    |            |            |            |            |            |            |
| 109 -> 118 | -0.20397   |            |            |            |            |            |            |
| 112 -> 122 | 0.35766    |            |            |            |            |            |            |
| 113 -> 124 | -0.32231   |            |            |            |            |            |            |
| Excited State | Singlet-A | E (eV) | λ (nm) | f | S^2 |
|---------------|-----------|--------|--------|---|-----|
| 55            | 5.6932    | 217.77 | 0.1248 | 0.000 |
| 101 -> 114    | 0.13860   |
| 103 -> 115    | -0.12258  |
| 104 -> 116    | 0.21958   |
| 105 -> 116    | 0.14729   |
| 106 -> 117    | -0.19196  |
| 106 -> 118    | 0.11019   |
| 107 -> 117    | -0.14419  |
| 107 -> 118    | 0.13816   |
| 108 -> 117    | 0.24140   |
| 109 -> 118    | 0.17675   |
| 111 -> 121    | 0.13873   |
| 112 -> 122    | -0.15132  |
| 113 -> 122    | -0.11436  |
| 113 -> 123    | 0.13431   |
| 113 -> 124    | -0.20188  |
| 56            | 5.7106    | 217.11 | 0.0709 | 0.000 |
| 104 -> 116    | -0.18794  |
| 105 -> 116    | 0.23858   |
| 106 -> 117    | 0.39136   |
| 107 -> 117    | -0.14292  |
| 108 -> 117    | -0.16357  |
| 110 -> 119    | 0.13909   |
| 112 -> 122    | -0.21471  |
| 113 -> 124    | -0.22602  |
| 57            | 5.7214    | 216.70 | 0.0515 | 0.000 |
| 101 -> 114    | 0.19353   |
| 103 -> 115    | 0.19094   |
| 104 -> 116    | 0.43783   |
| 106 -> 117    | 0.17131   |
| 108 -> 117    | -0.22790  |
| 110 -> 119    | 0.15879   |
| 113 -> 123    | -0.10885  |
| 113 -> 124    | 0.19923   |
| 58            | 5.7396    | 216.01 | 0.0513 | 0.000 |
| 99 -> 114     | 0.13143   |
| 100 -> 114    | 0.24059   |
| 101 -> 114    | 0.29713   |
| 103 -> 115    | 0.23650   |
| 104 -> 116    | -0.24899  |
| 107 -> 117    | -0.12148  |
| 110 -> 119    | 0.16992   |
| 112 -> 122    | 0.23811   |
| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | Oscillator Strength (f) | <S**2>> |
|---------------|-----------|-------------|-----------------|-------------------------|---------|
| 59:           | 5.7444    | 215.84      | 0.1251          | 0.000                   |
| 100           | 0.20373   |             |                 |                         |
| 101           | 0.37849   |             |                 |                         |
| 103           | 0.23903   |             |                 |                         |
| 106           | -0.25745  |             |                 |                         |
| 107           | -0.12772  |             |                 |                         |
| 110           | -0.14531  |             |                 |                         |
| 111           | 0.14409   |             |                 |                         |
| 112           | -0.10573  |             |                 |                         |
| 113           | -0.11229  |             |                 |                         |

| Excited State | Singlet-A | Energy (eV) | Wavelength (nm) | Oscillator Strength (f) | <S**2>> |
|---------------|-----------|-------------|-----------------|-------------------------|---------|
| 60:           | 5.7640    | 215.10      | 0.0240          | 0.000                   |
| 103           | 0.29635   |             |                 |                         |
| 106           | 0.16461   |             |                 |                         |
| 108           | 0.46809   |             |                 |                         |
| 108           | 0.13178   |             |                 |                         |
| 111           | 0.15247   |             |                 |                         |
| 113           | -0.12717  |             |                 |                         |
| 113           | 0.17370   |             |                 |                         |