Supporting information for

Redox Active Dysprosium Single Molecule Magnet: Spectroelectrochemistry and Theoretical Investigations

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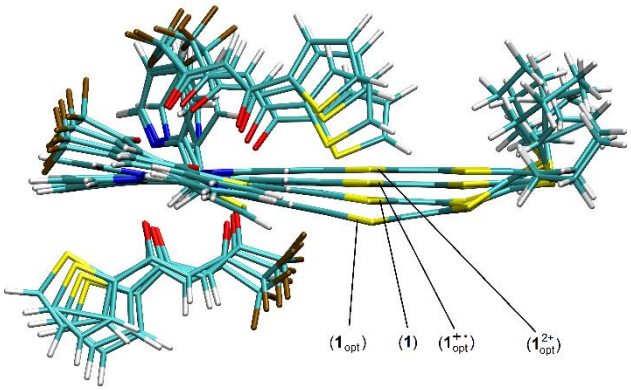

Figure S1. X-ray structure of **1** and optimized structures for the different oxidation states ($\mathbf{1}_{opt}$, $\mathbf{1}^{+\cdot}{}_{opt}$ and $\mathbf{1}^{2+}{}_{opt}$).

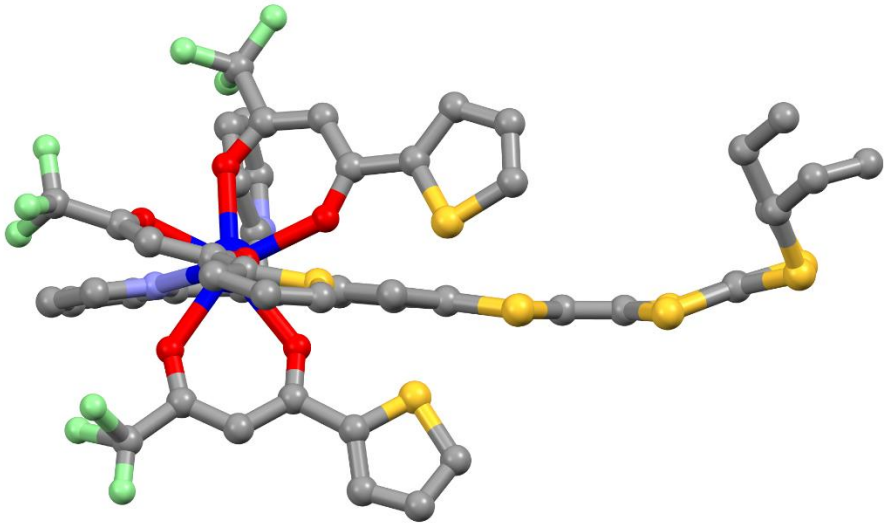

Figure S2. Experimental X-ray structure of **1**. *n*-hexane molecule of crystallization and hydrogen atoms are omitted for clarity. Color code: gray: carbon, red: oxygen, green: fluoride, blue: nitrogen and yellow: sulfur.

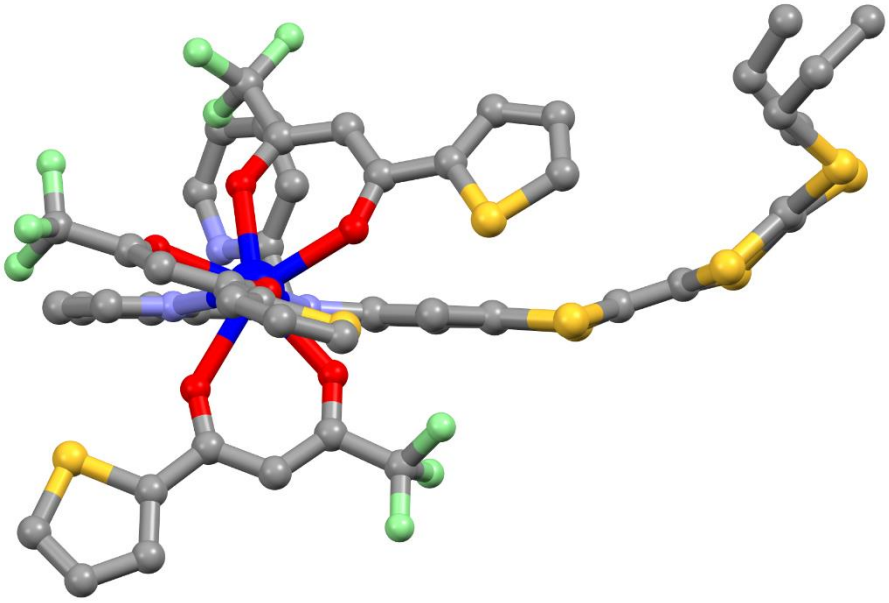

Figure S3. Optimized structure of ($\mathbf{1}_{opt}$). Color code: gray: carbon, red: oxygen, green: fluoride, blue: nitrogen and yellow: sulfur.

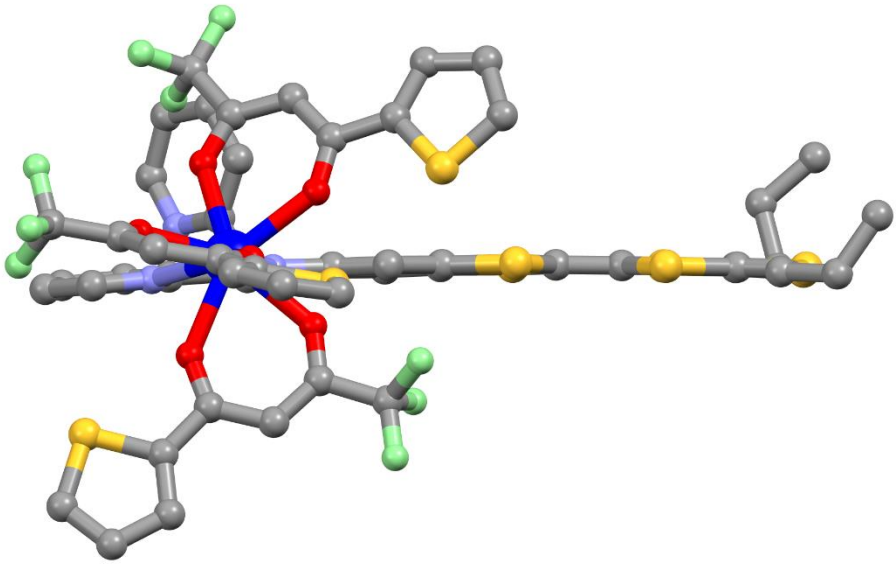

Figure S4. Optimized structure of $\mathbf{1}^{+\bullet}$ ($\mathbf{1}^{+\bullet}_{opt}$). Color code: gray: carbon, red: oxygen, green: fluoride, blue: nitrogen and yellow: sulfur.).

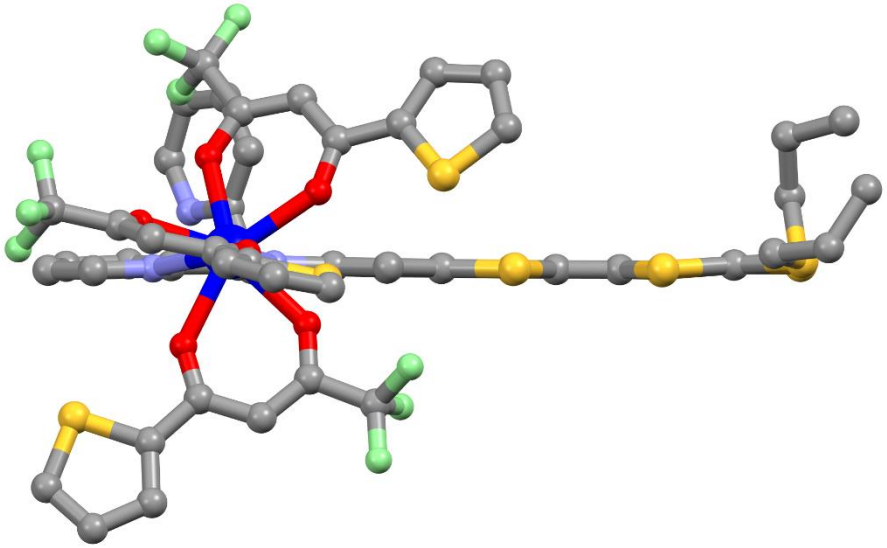

Figure S5. Optimized structure of $\mathbf{1}^{2+}$ ($\mathbf{1}^{2+}_{opt}$). Color code: gray: carbon, red: oxygen, green: fluoride, blue: nitrogen and yellow: sulfur.

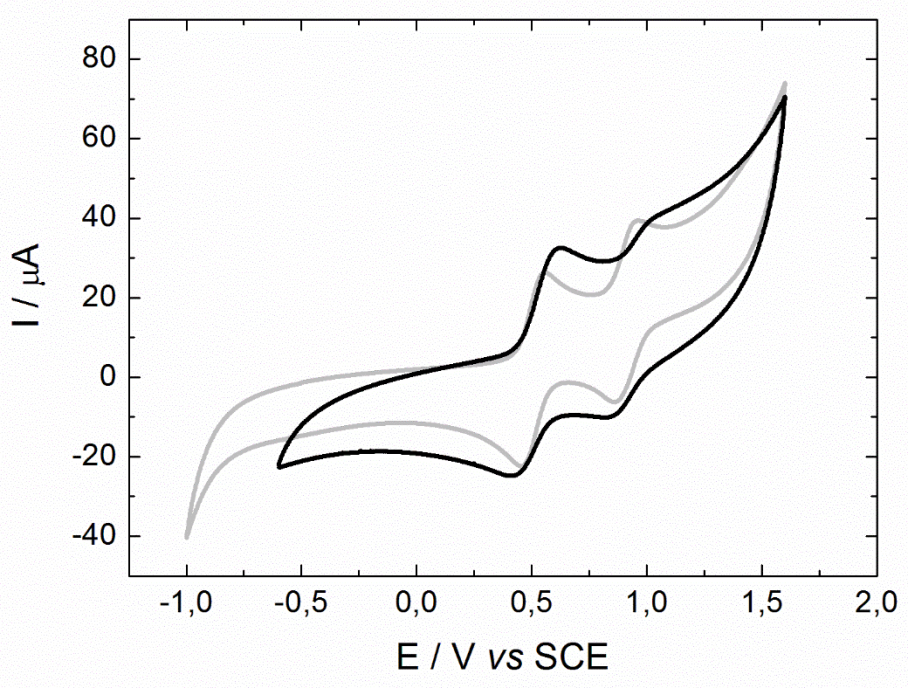

Figure S6. Cyclic voltammograms of **L** (gray line) and **1** (black line) in CH_2Cl_2 at a scan rate of 100 $mV \cdot s^{-1}$. The potentials were measured *vs*. a saturated calomel electrode (SCE) with Pt wires as working and counter electrodes.

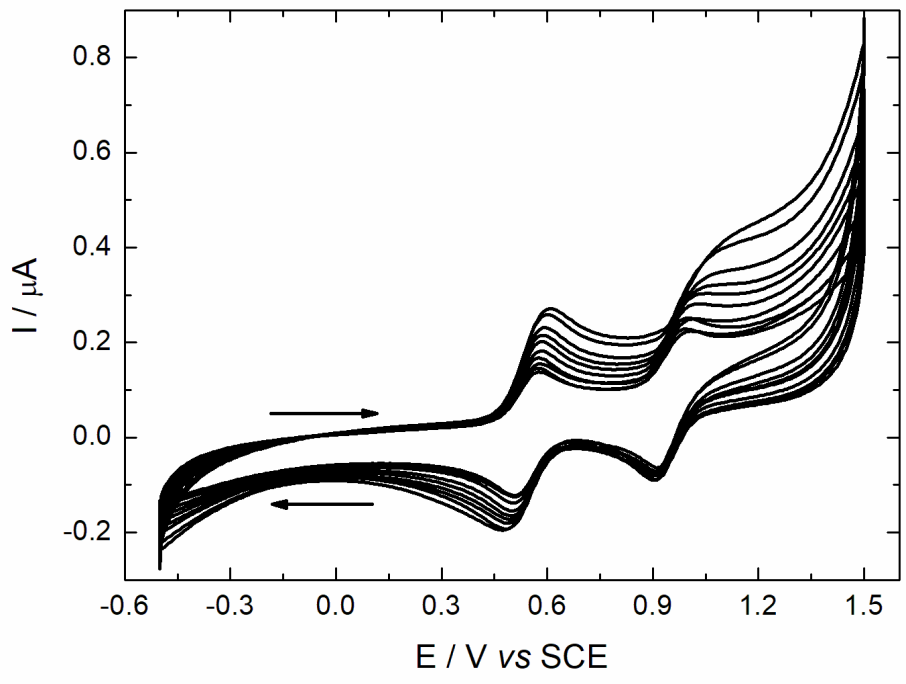

Figure S7. Cyclic voltammograms of **1** (black line, ten first cycles) in CH_2Cl_2 at a scan rate of 100 $mV \cdot s^{-1}$. The potentials were measured *vs*. a saturated calomel electrode (SCE) with Pt wires as working and counter electrodes.

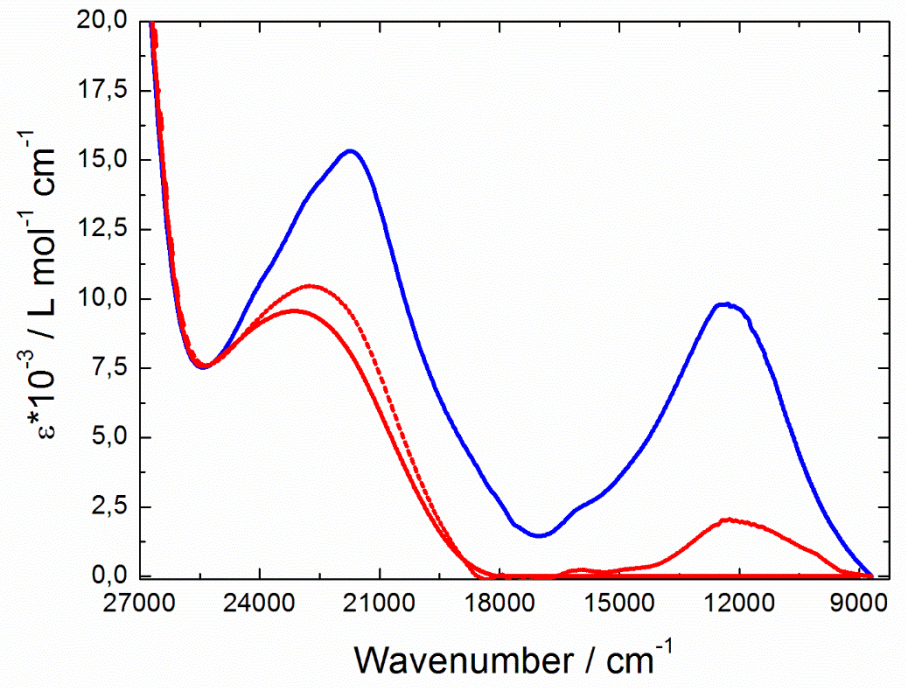

Figure S8. Initial absorption spectra of **1** (red line), after oxidation under a 0.7 V applied currant $\mathbf{1}^{+\bullet}$ (blue line) and after switch off the applied currant for 20 minutes (dashed red line). The full red line is recovered after 35 minutes.

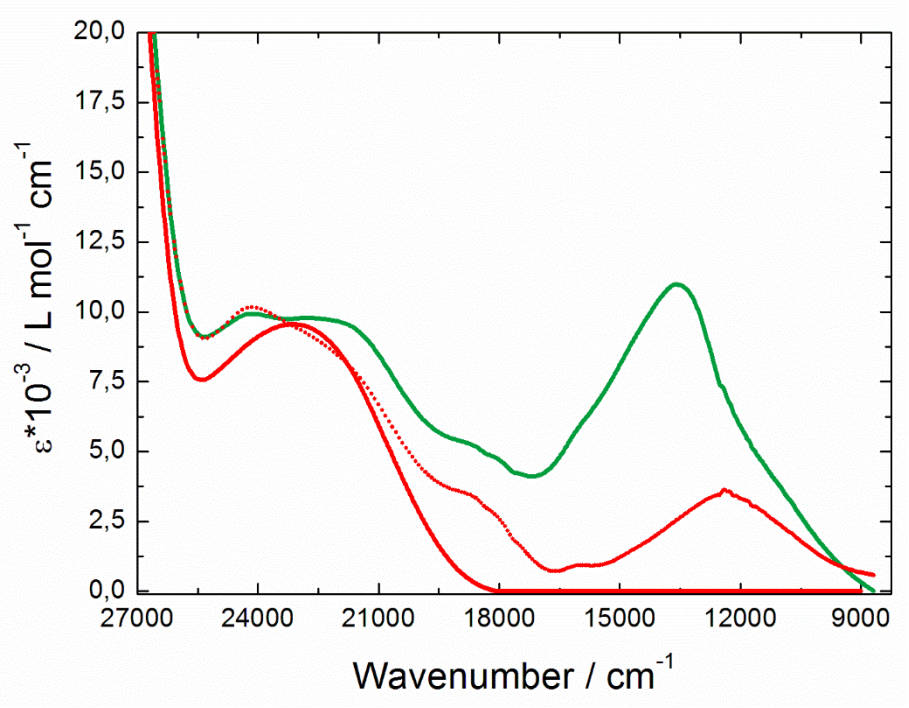

Figure S9. Initial absorption spectra of **1** (red line), after oxidation under a 1.1 V applied currant $\mathbf{1}^{2+}$ (green line) and after switch off the applied currant for 20 minutes (dashed red line).

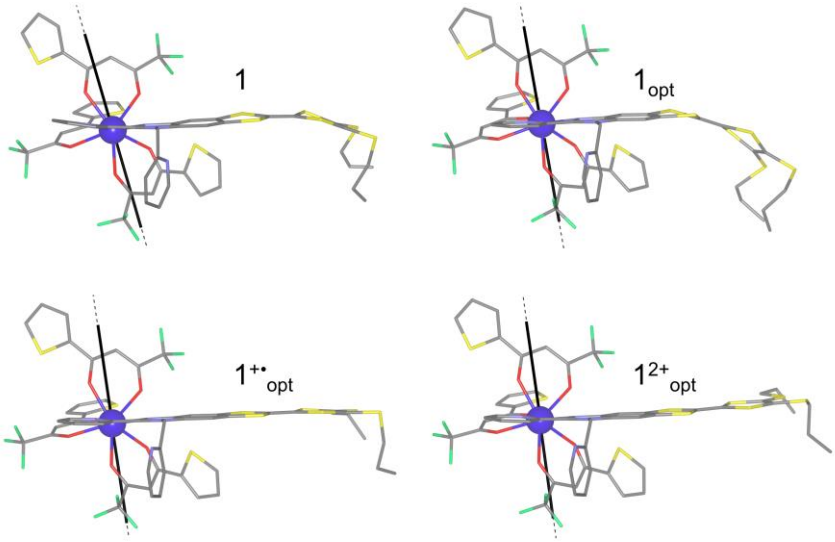

Figure S10. Representation of the calculated local ground state magnetic anisotropy axes for the complexes **1**, $\mathbf{1}_{opt}$, $\mathbf{1}^{+\bullet}{}_{opt}$ and $\mathbf{1}^{2+}{}_{opt}$.

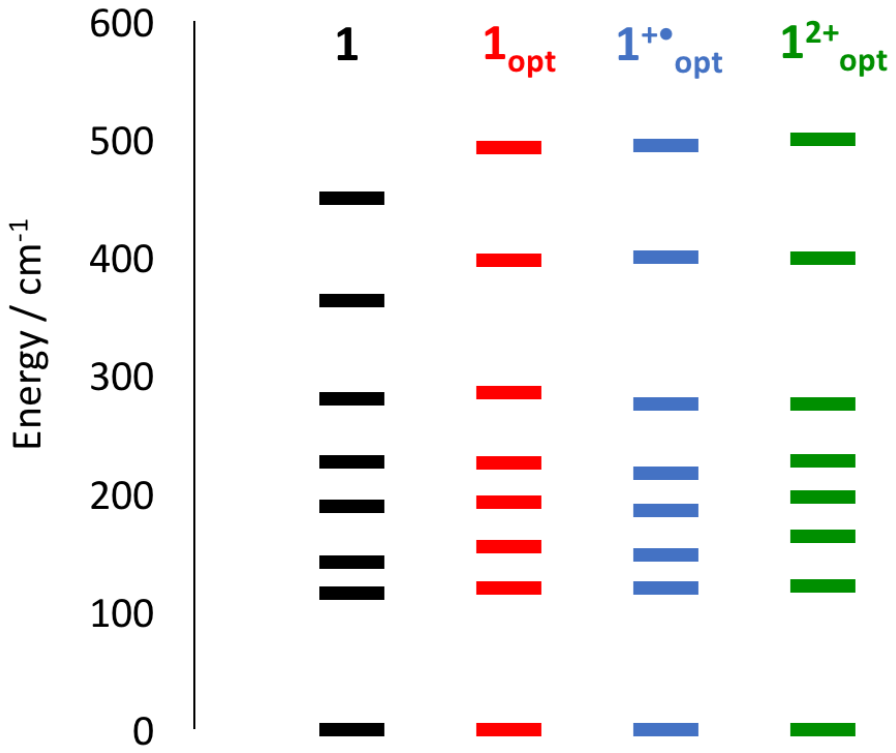

Figure S11. Energy splitting of the $^6H_{15/2}$ ground state for the complexes **1**, **1**$_{opt}$, **1**$^{+\bullet}_{opt}$ and **1**$^{2+}_{opt}$.

Table S1. Main components of the magnetic anisotropy g-tensor (g_{XX}, g_{YY} and g_{ZZ}) and wavefunction composition of the ground state of **1**, **1**$_{opt}$, **1**$^{+\bullet}_{opt}$ and **1**$^{2+}_{opt}$.

Complexe	g_{XX}	g_{YY}	g_{ZZ}	wavefunction composition
1	0.02	0.03	19.52	0.93 |±15/2> + 0.06 |±11/2>
1$_{opt}$	0.02	0.03	19.56	0.93 |±15/2> + 0.06 |±11/2>
1$^{+\bullet}_{opt}$	0.02	0.04	19.57	0.94 |±15/2> + 0.06 |±11/2>
1$^{2+}_{opt}$	0.01	0.01	19.45	0.93 |±15/2> + 0.06 |±11/2>