

## Supporting information

### **Double doping of a low-ionization energy polythiophene with a molybdenum dithiolene complex**

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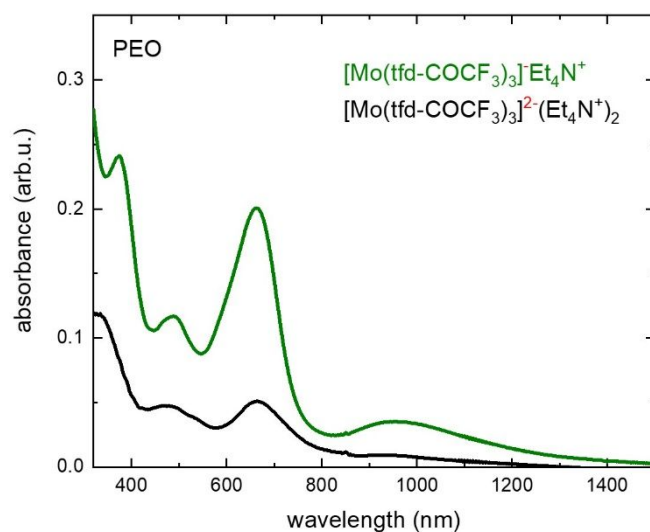
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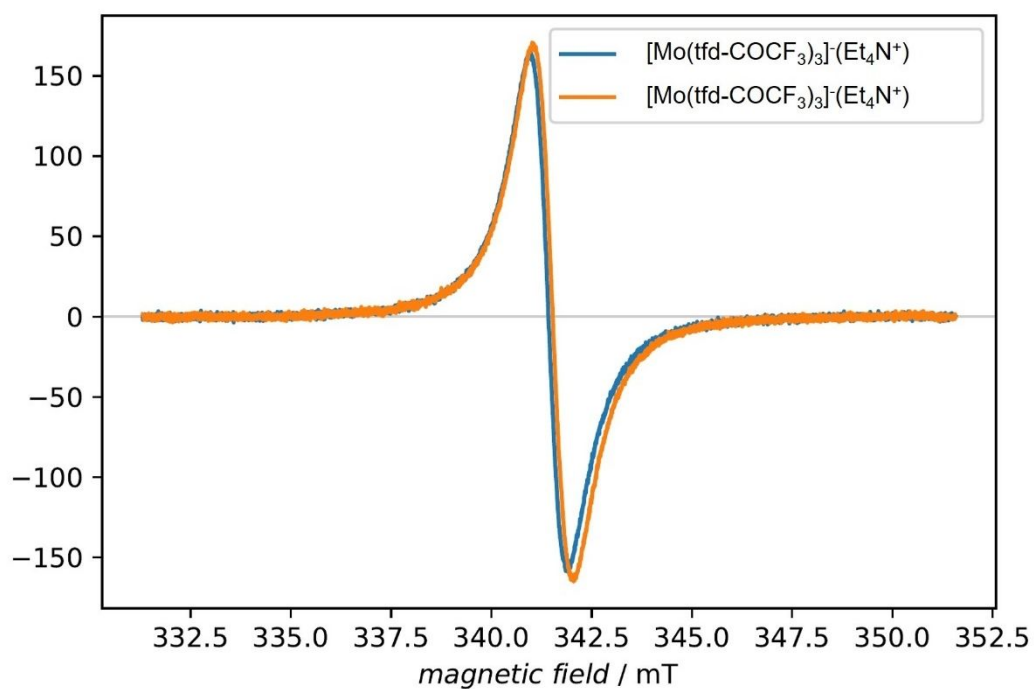
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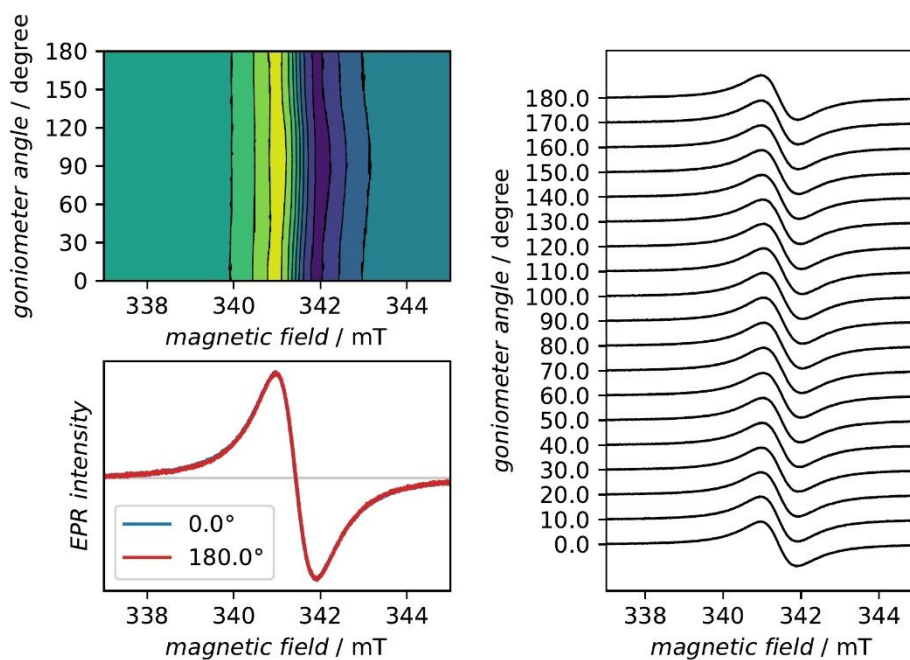
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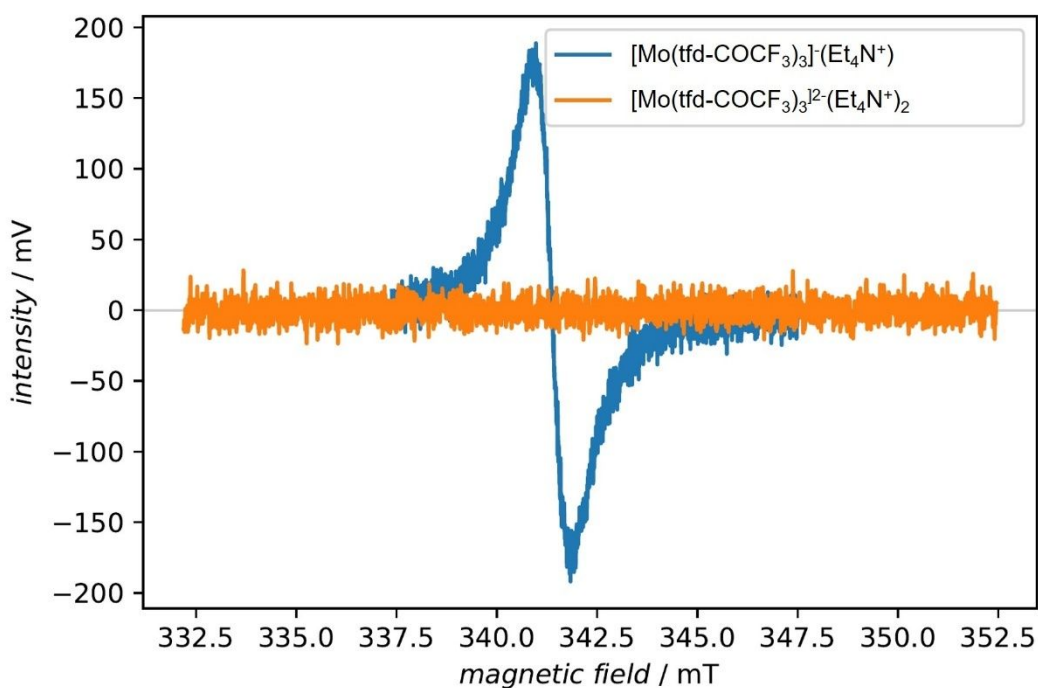
**Figure S1.** UV-vis spectra of  $[\text{Mo}(\text{tfd-COCF}_3)_3]\cdot\text{Et}_4\text{N}^+$  and  $[\text{Mo}(\text{tfd-COCF}_3)_3]_2\cdot(\text{Et}_4\text{N}^+)_2$  dispersed in a PEO matrix.



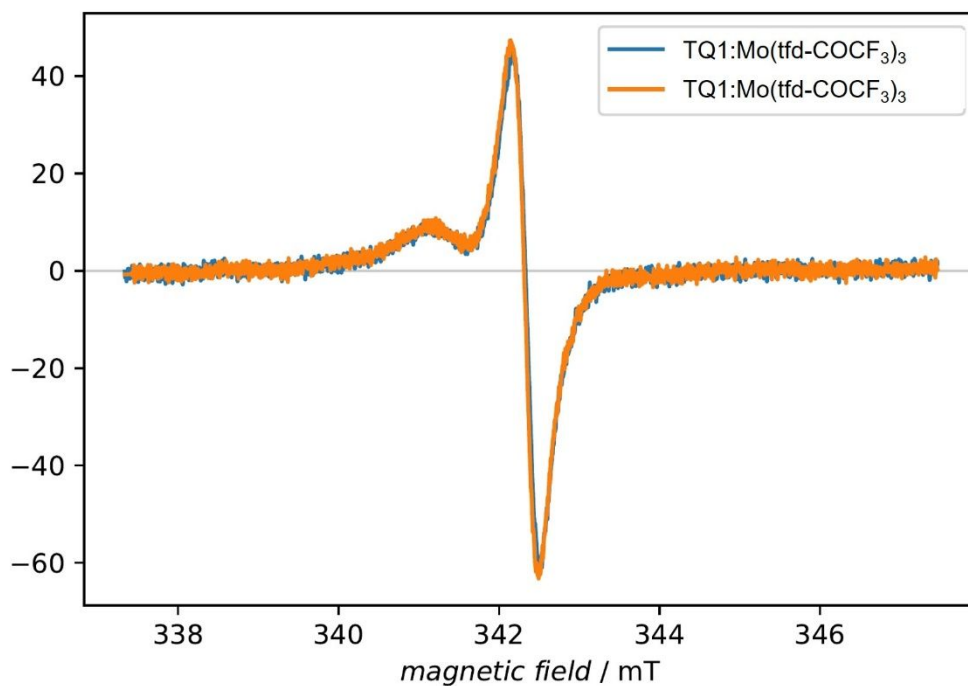
**Figure S2.** EPR spectra of a thin film of  $[\text{Mo}(\text{tfd-COCF}_3)_3]\cdot\text{Et}_4\text{N}^+$  dispersed in a PEO matrix recorded at  $0^\circ$  and  $90^\circ$ .



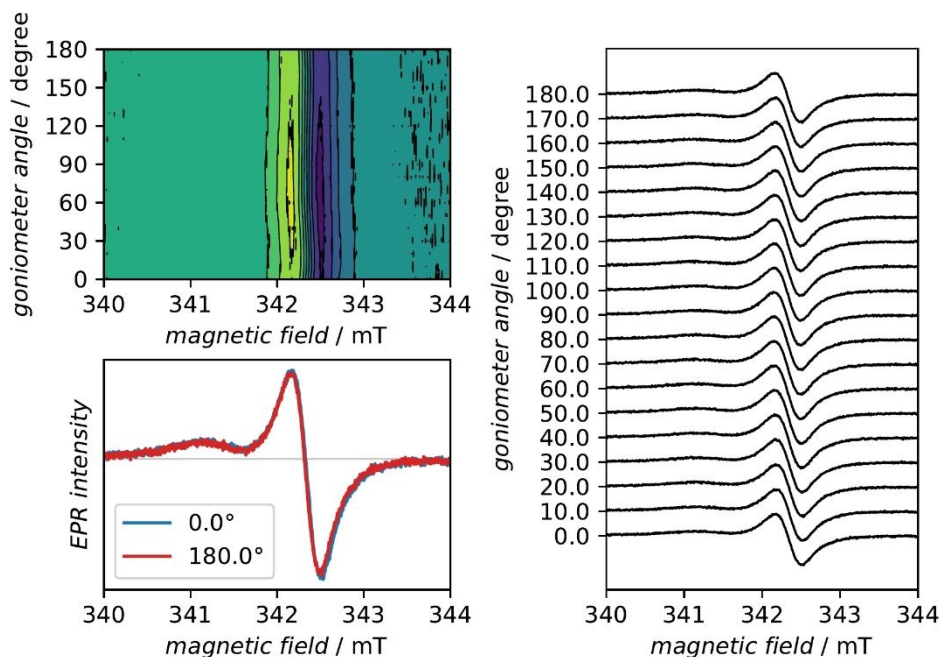
**Figure S3.** Angular-dependent EPR spectra of a thin film of  $[\text{Mo}(\text{tfd-COCF}_3)_3] \cdot \text{Et}_4\text{N}^+$  dispersed in a PEO matrix. Spectra have been recorded using a motorized goniometer with an accuracy of  $0.125^\circ$ . The sample has been manually positioned within the spectrometer initially with the substrate plane parallel to the magnetic field (with an accuracy of  $\pm 5^\circ$ ).



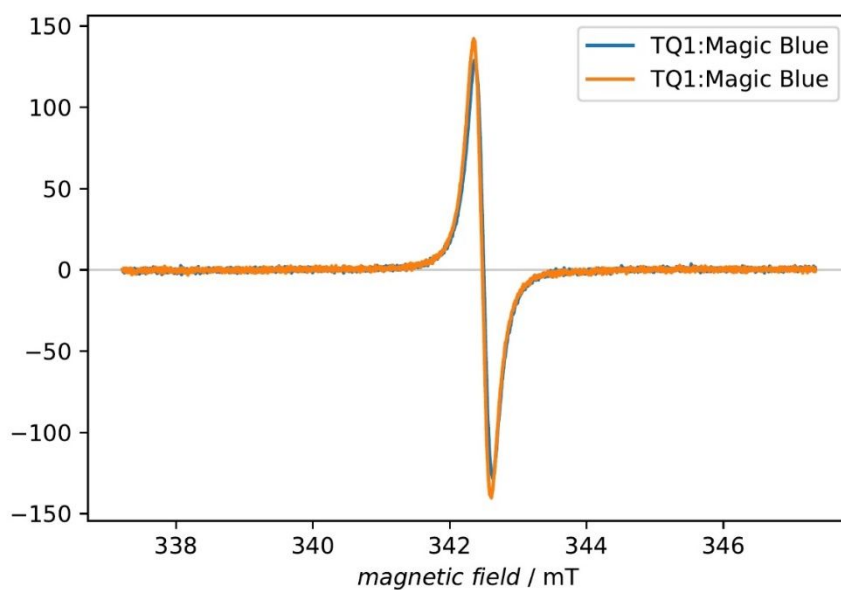
**Figure S4.** Non-smoothed EPR spectra of  $[\text{Mo}(\text{tfd-COCF}_3)_3] \cdot \text{Et}_4\text{N}^+$  and  $[\text{Mo}(\text{tfd-COCF}_3)_3]_2 \cdot (\text{Et}_4\text{N}^+)_2$  dispersed in a PEO matrix.



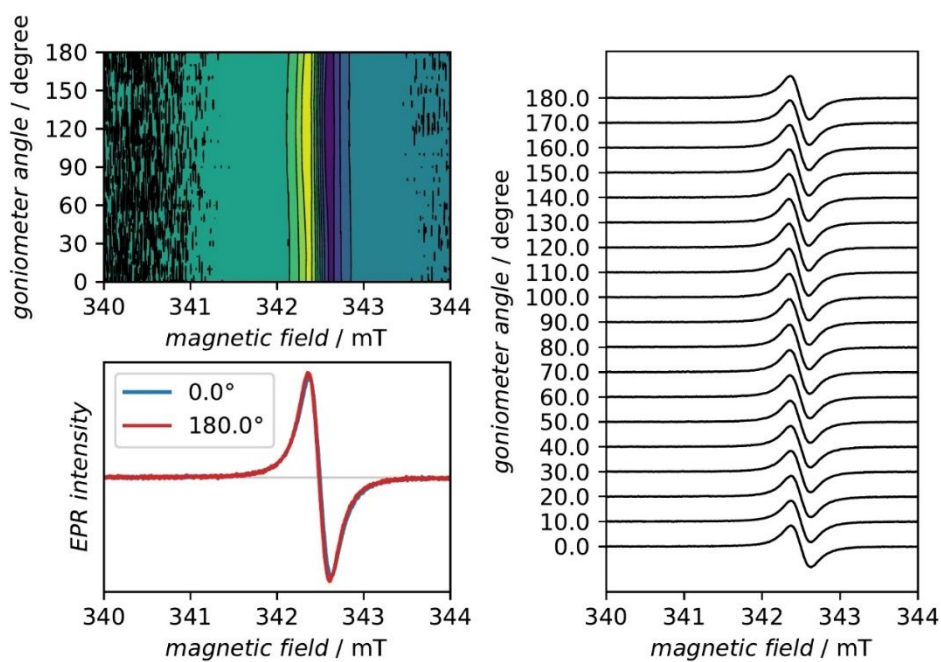
**Figure S5.** EPR spectra of TQ1 sequentially doped with Mo(tfd-COCF<sub>3</sub>)<sub>3</sub> recorded at 0° and 90°.



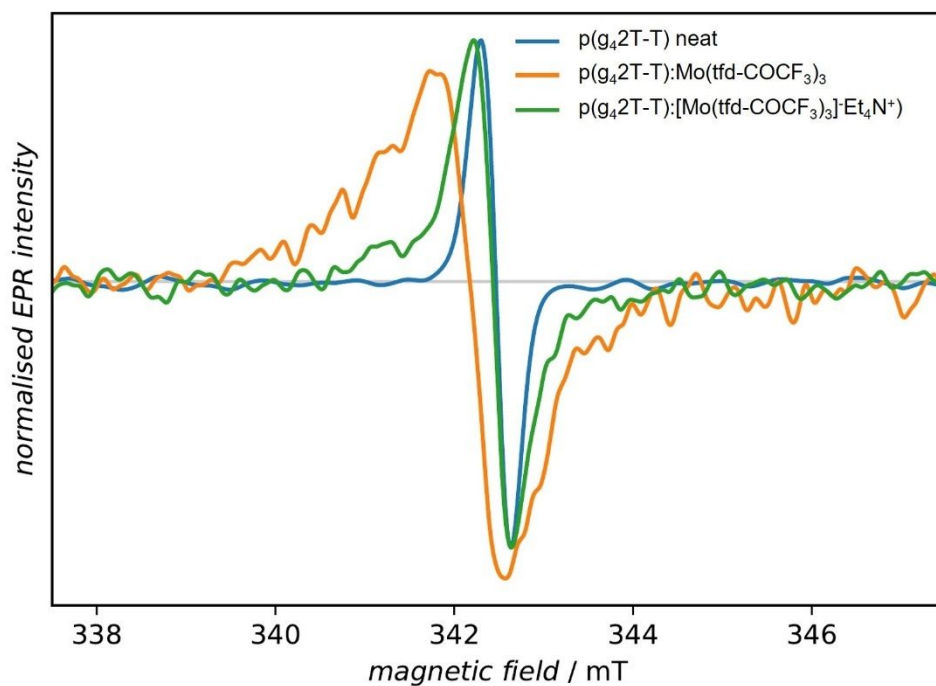
**Figure S6.** Angular-dependent EPR spectra of a TQ1 sequentially doped with Mo(tfd-COCF<sub>3</sub>)<sub>3</sub>. Spectra have been recorded using a motorized goniometer with an accuracy of 0.125°. The sample has been manually positioned within the spectrometer initially with the substrate plane parallel to the magnetic field (with an accuracy of ±5°).



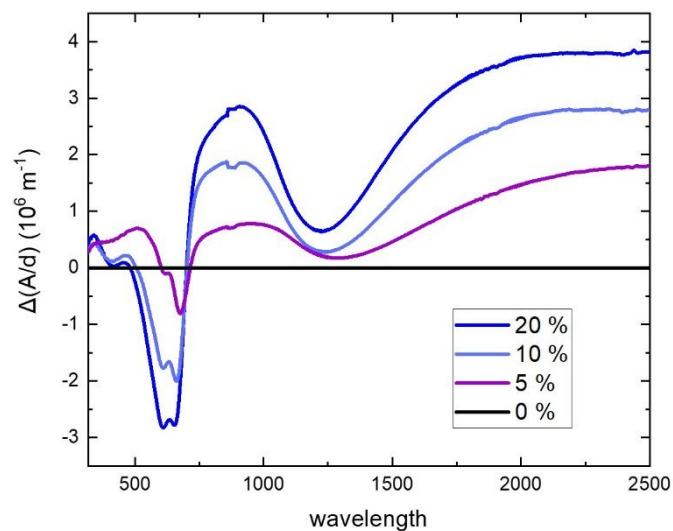
**Figure S7.** EPR spectra of TQ1 sequentially doped with Magic Blue recorded at 0° and 90°.



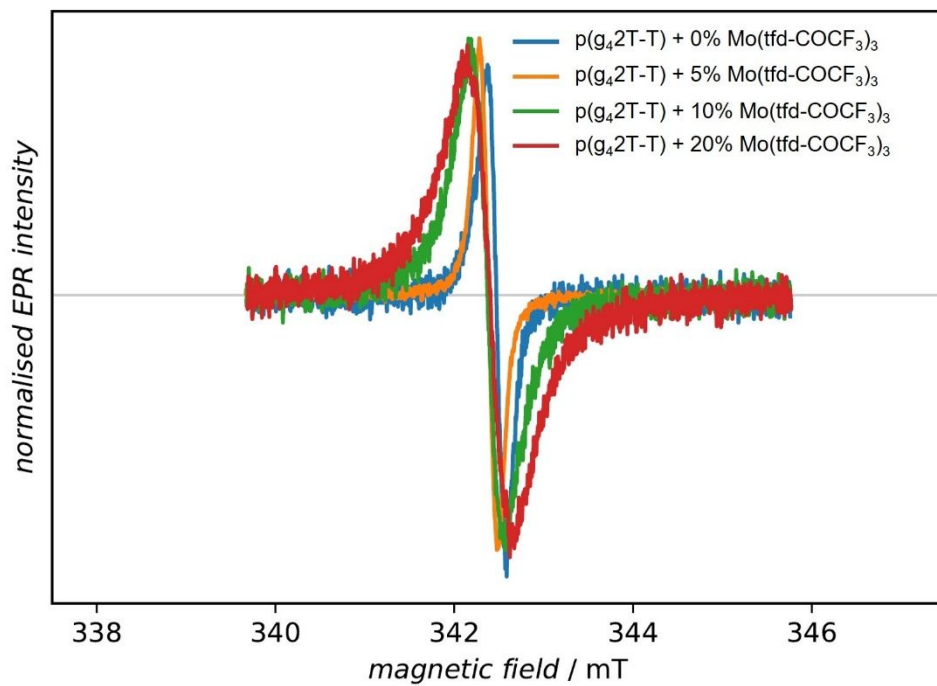
**Figure S8.** Angular-dependent EPR spectra of a TQ1 sequentially doped with Magic Blue. Spectra have been recorded using a motorized goniometer with an accuracy of 0.125°. The sample has been manually positioned within the spectrometer initially with the substrate plane parallel to the magnetic field (with an accuracy of  $\pm 5^\circ$ ).



**Figure S9.** Non-smoothed EPR spectra of neat p(g<sub>4</sub>2T-T) and p(g<sub>4</sub>2T-T) sequentially doped with Mo(tfd-COCF<sub>3</sub>)<sub>3</sub> and [Mo(tfd-COCF<sub>3</sub>)<sub>3</sub>]<sup>-</sup>Et<sub>4</sub>N<sup>+</sup>.



**Figure S10.** UV-vis-NIR absorbance spectra displaying the difference in thickness-normalized absorbance Δ(A/d) between the spectra of neat p(g<sub>4</sub>2T-T) and p(g<sub>4</sub>2T-T) co-processed with 5 mol%, 10 mol% and 20 mol% of Mo(tfd-COCF<sub>3</sub>)<sub>3</sub>.



**Figure S11.** Non-smoothed EPR spectra of co-processed p(g<sub>4</sub>2T-T):Mo(tfd-COCF<sub>3</sub>)<sub>3</sub> films.