

## Electronic Supplementary Information

# A biferrocenium salt containing paramagnetic tetracyanoquinodimethane hexamers: Charge disproportionation via donor–acceptor interactions

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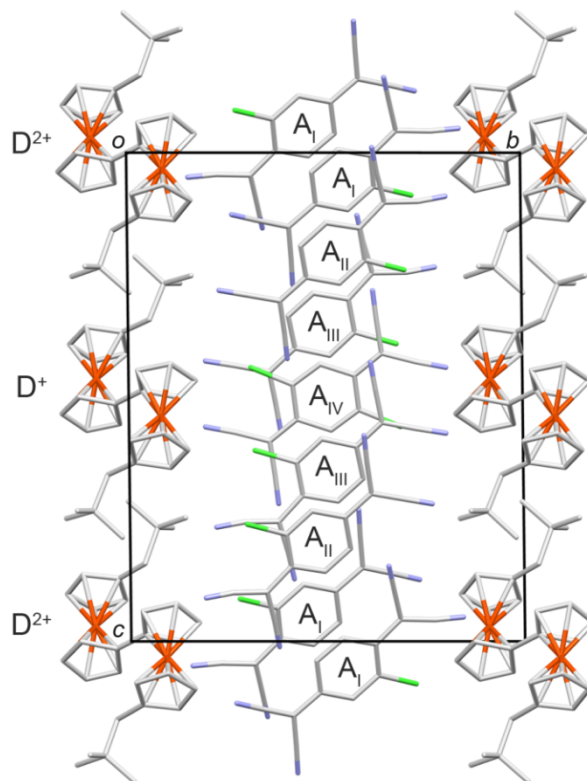
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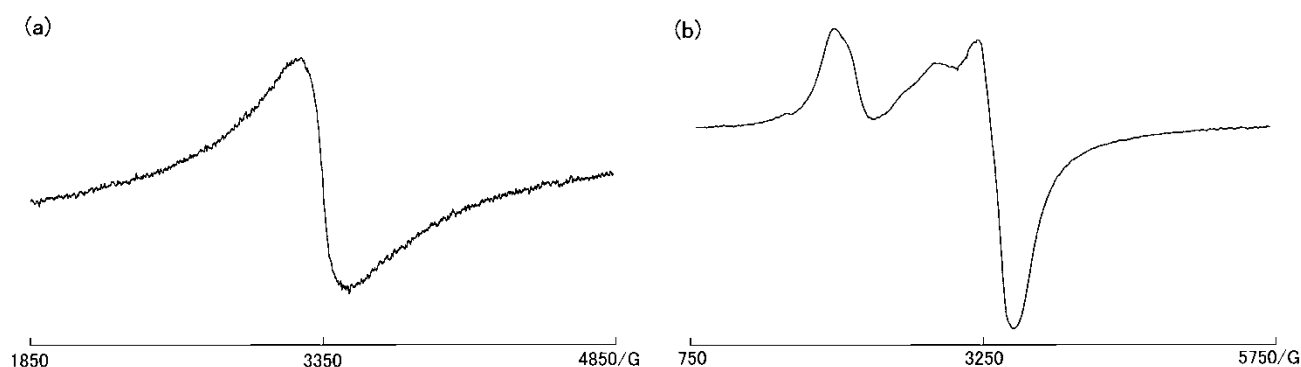
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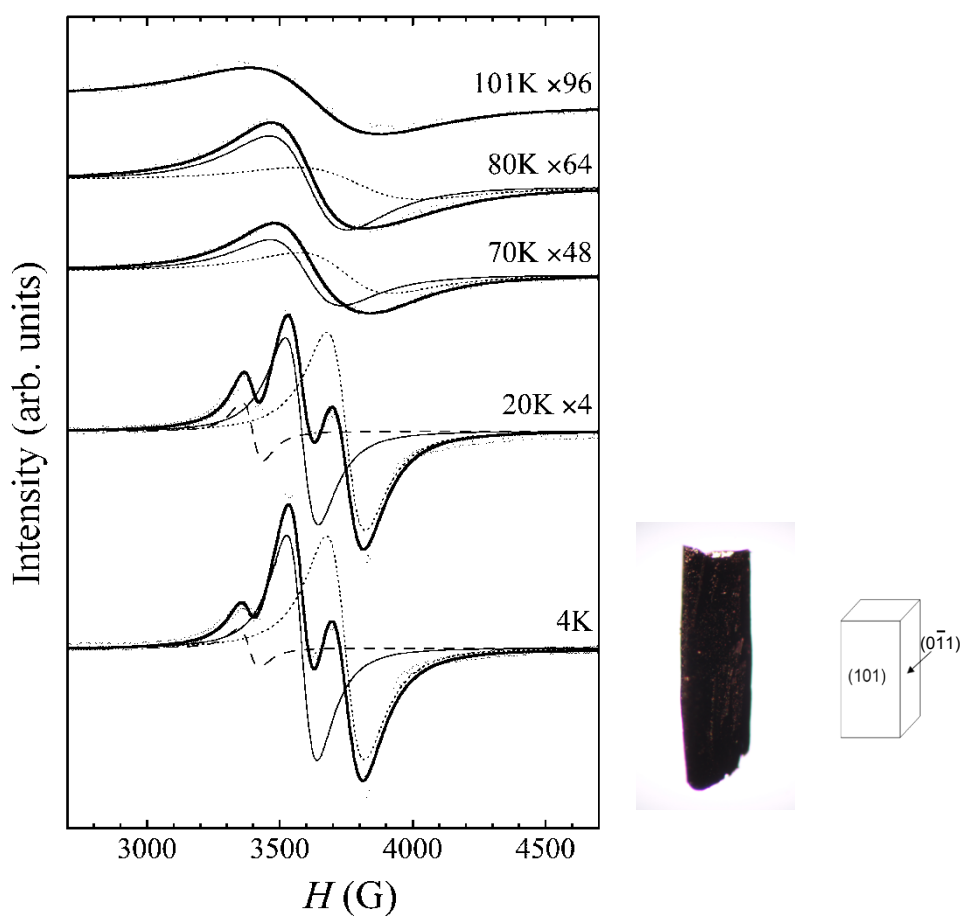
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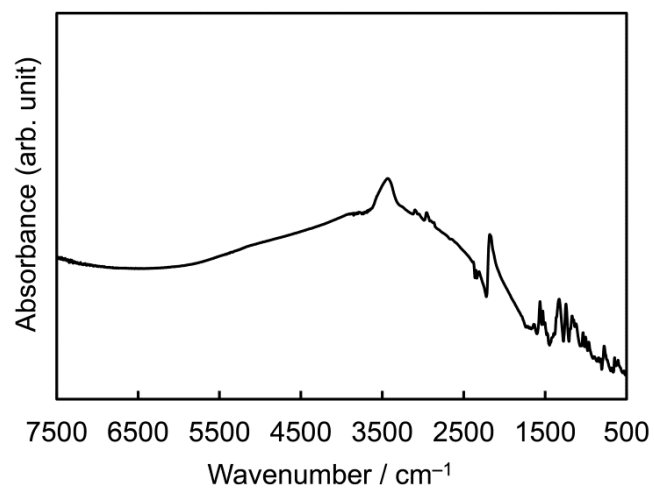
**Fig. S1.** Packing diagram of [dineopentyl-biferrocene]<sub>2</sub>[Cl<sub>1</sub>TCNQ]<sub>7</sub> (**1**) viewed along the *a*-axis.



**Fig. S2.** ESR spectra of polycrystalline samples of **1** recorded at (a) room temperature and (b) 77 K.



**Fig. S3.** Temperature dependence of the ESR spectra of oriented crystals of **1** recorded under magnetic fields applied normal to the (101) plane. The data points and fitting curves are indicated by circles ( $\circ$ ) and bold lines, respectively. Fitting was done via summation of the TCNQ component (thin line), ferrocenium component (dotted line), and impurity (dashed line). A photograph of a plate-like single crystal of **1** (~0.3 mm length) is shown on the right.



**Fig. S4.** Absorption spectrum of **1** in the NIR-IR region at room temperature (KBr pellet).