

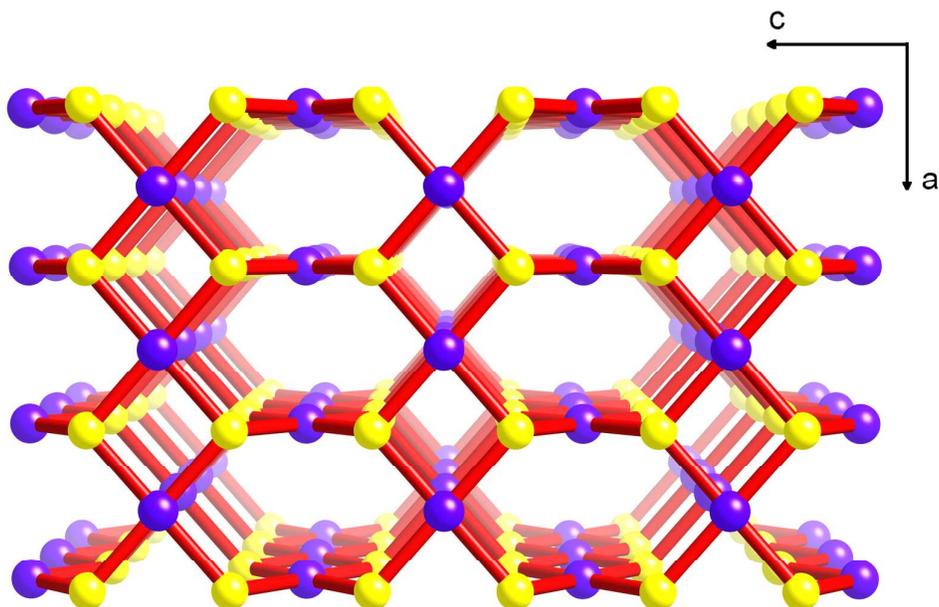
# **PtS-related $\{[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})]\}_\infty$ networks**

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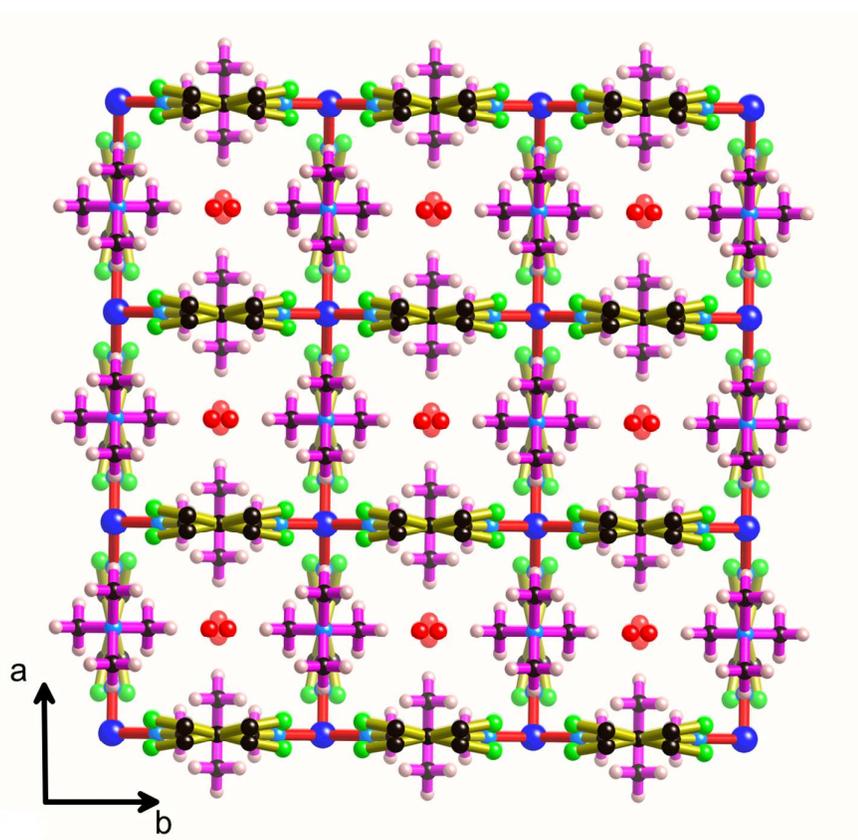
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## **Supporting Information**

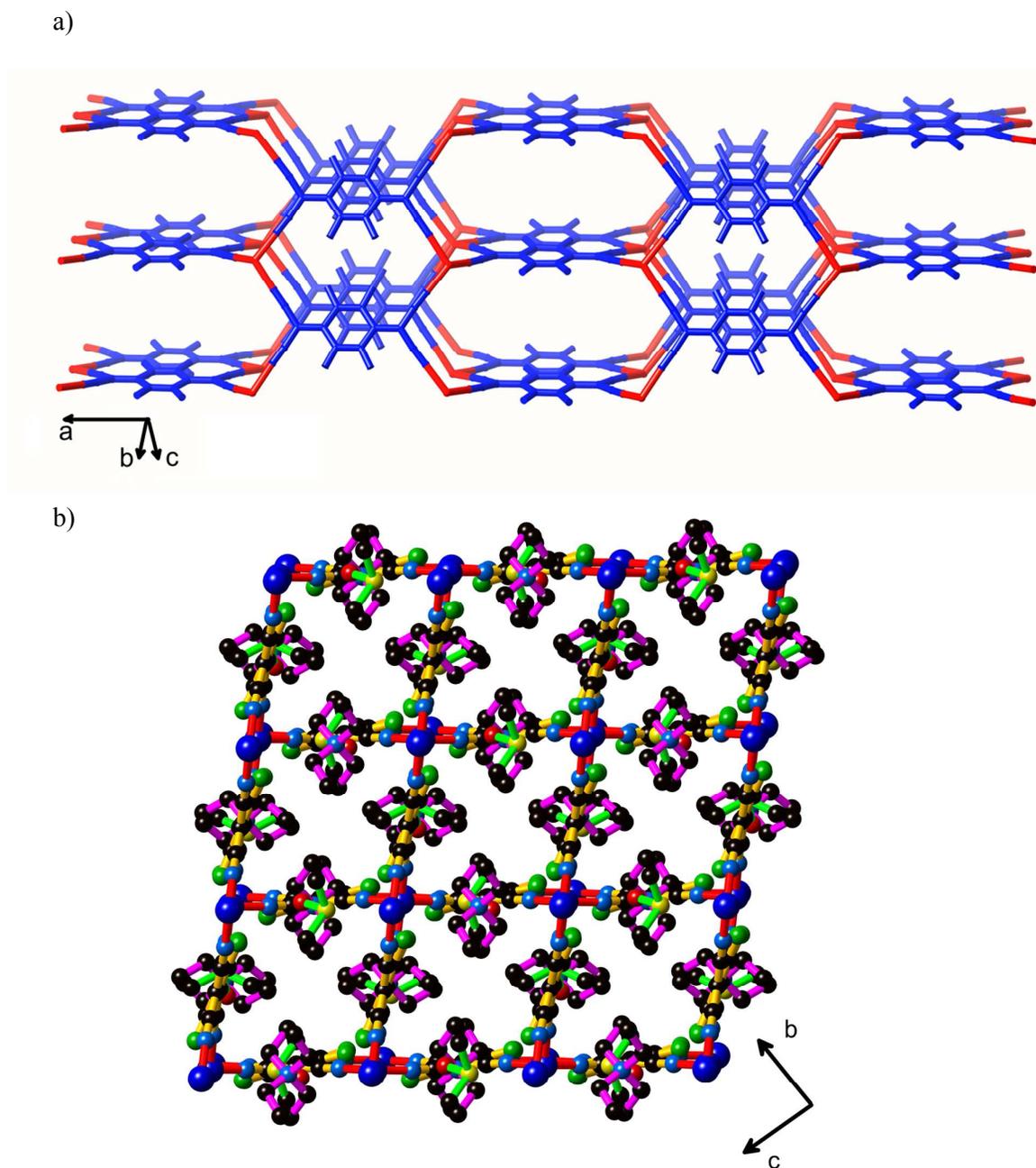
Supplementary Figures S1-S6



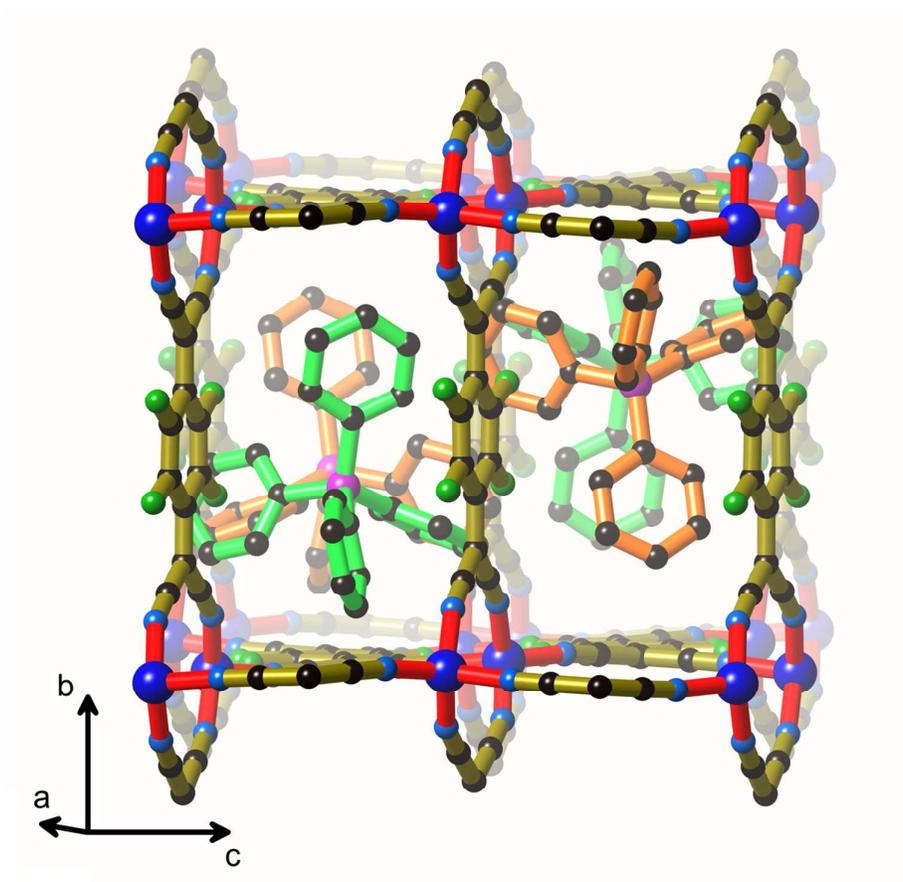
**Figure S1.** The structure of PtS viewed from an angle close to the *b* axis.



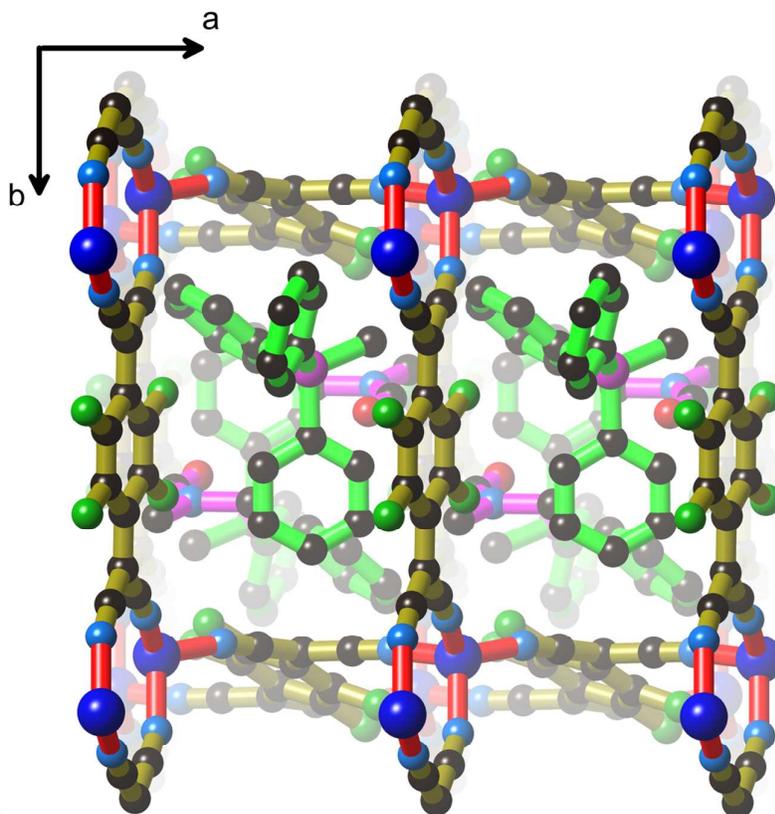
**Figure S2.** A view down the *c* axis of  $\text{NMe}_4[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})] \cdot 0.5\text{H}_2\text{O}$ .  $\text{F}_4\text{TCNQ}^{2-}$  units are indicated by gold connections, Cu-N bonds are indicated by red connections, tetramethylammonium cations are indicated by pink bonds. All sites occupied by the oxygen atom of the disordered water molecule are shown. Sphere color code: Cu blue, F green, O red, N light blue, C black, H pale pink.



**Figure S3.** The structure of  $\text{NEt}_4[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})] \cdot \text{DMSO} (4 \cdot \text{DMSO})$ . a) A stick representation of the anionic  $[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})]^-$  framework viewed from an angle close to the  $[0\ 1\ -1]$  direction; the  $\text{F}_4\text{TCNQ}^{\text{II-}}$  ligands are represented by blue connections, Cu-N bonds are represented by red connections. b) A view down the  $a$  axis.  $\text{F}_4\text{TCNQ}^{2-}$  units are indicated by gold connections, Cu-N bonds are indicated by red connections, tetraethylammonium cations are indicated by pink bonds, dimethylsulfoxide molecules are indicated by green bonds. Only one orientation for each dimethylsulfoxide molecule is shown. Sphere color code: Cu blue, S yellow, F green, O red, N light blue, C black, H pale pink.

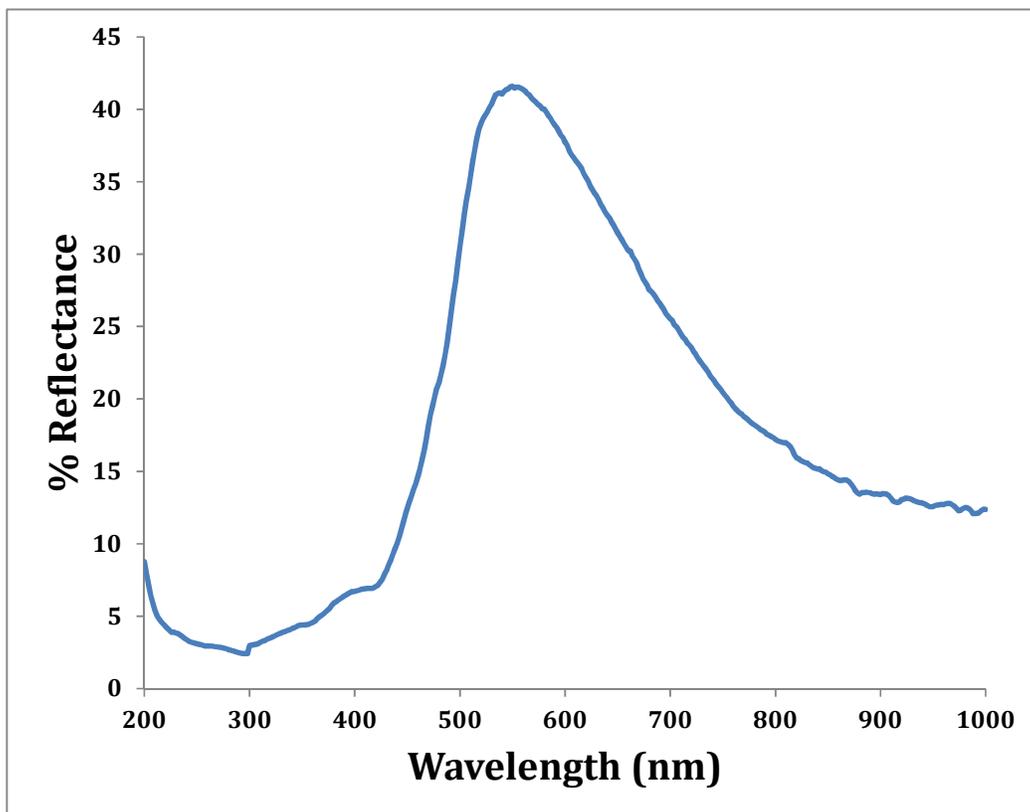


**Figure S4.** A view of  $\text{PPh}_4[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II}})] \cdot 0.5 \text{ DMF}$  (**5**·0.5 DMF) along the rectangular channels in which all  $\text{F}_4\text{TCNQ}^{2-}$  units are viewed almost edge-on.  $\text{F}_4\text{TCNQ}^{2-}$  units are indicated by gold connections, Cu-N bonds are indicated by red connections, tetraphenylphosphonium cations are indicated by green and orange connections. Sphere color code: Cu blue, P pink, F green, N light blue, C black. H atoms have been omitted for clarity.

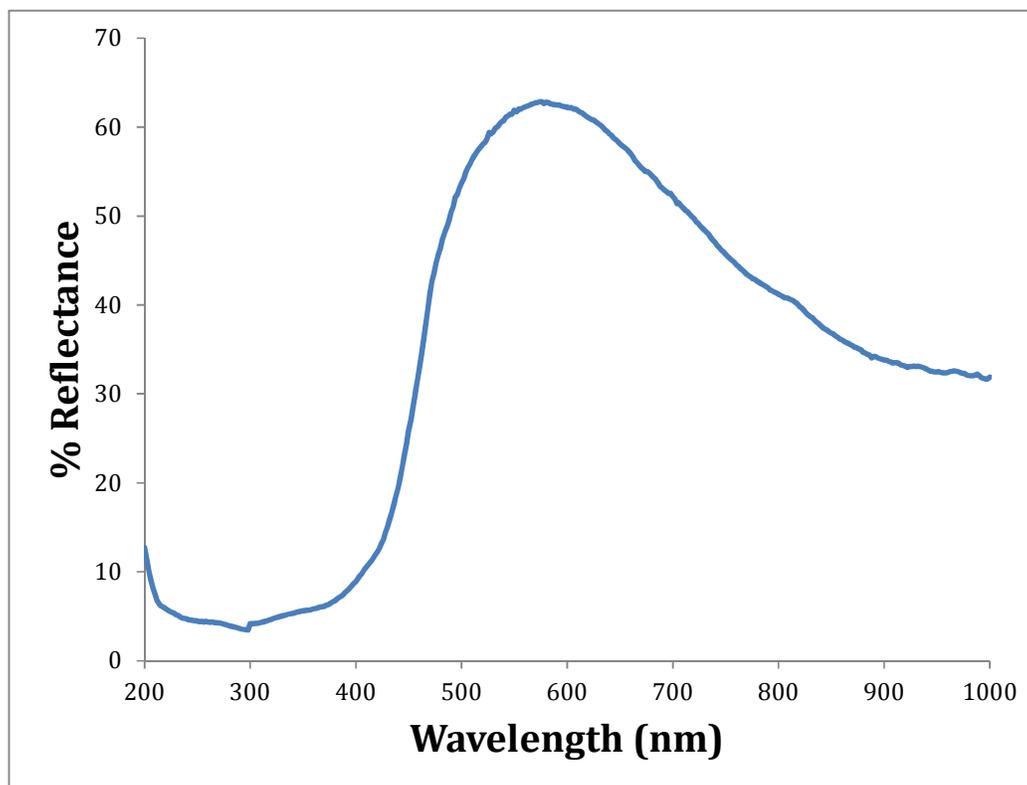


**Figure S5.** A view of  $\text{PPh}_4[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})] \cdot \text{DMF}$  (**6**·DMF) along the rectangular channels in which all  $\text{F}_4\text{TCNQ}^{2-}$  units are viewed almost edge-on.  $\text{F}_4\text{TCNQ}^{2-}$  units are indicated by gold connections, Cu-N bonds are indicated by red connections, tetraphenylphosphonium cations are indicated by green connections, dimethylformamide by pink connections. Sphere color code: Cu blue, P pink, F green, O red, N light blue, C black. H atoms have been omitted for clarity.

a)



b)



**Figure S6.** UV/Vis reflectance spectra for a)  $\text{PPh}_4[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})] \cdot 0.5 \text{ DMF}$  (5·0.5 DMF) and b)  $\text{PPh}_4[\text{Cu}^{\text{I}}(\text{F}_4\text{TCNQ}^{\text{II-}})] \cdot \text{DMF}$  (6·DMF).