

## Proton Switching of Polarity in Metalloamphiphile Crystals

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### SUPPLEMENTARY INFORMATION

Fig. Supp. 1 Atom separations and the implied H-bonding network (dashed lines) within complex **1** in the vicinity of lattice water molecules, the protonated external amino group and its nearest anions.

Fig. Supp. 2 A partial view of the doubly disordered array of methyl-cap coordinated NH units and their nearest perchlorate anions, showing their 3:3 mutual surrounding. The shortest N...O separation is 2.95 Å.

Fig. Supp. 3 A partial view of the lattice of complex **2** down *a* with perchlorate anions omitted, so that the presence of hydrocarbon moieties within both “interfaces” is more obvious.

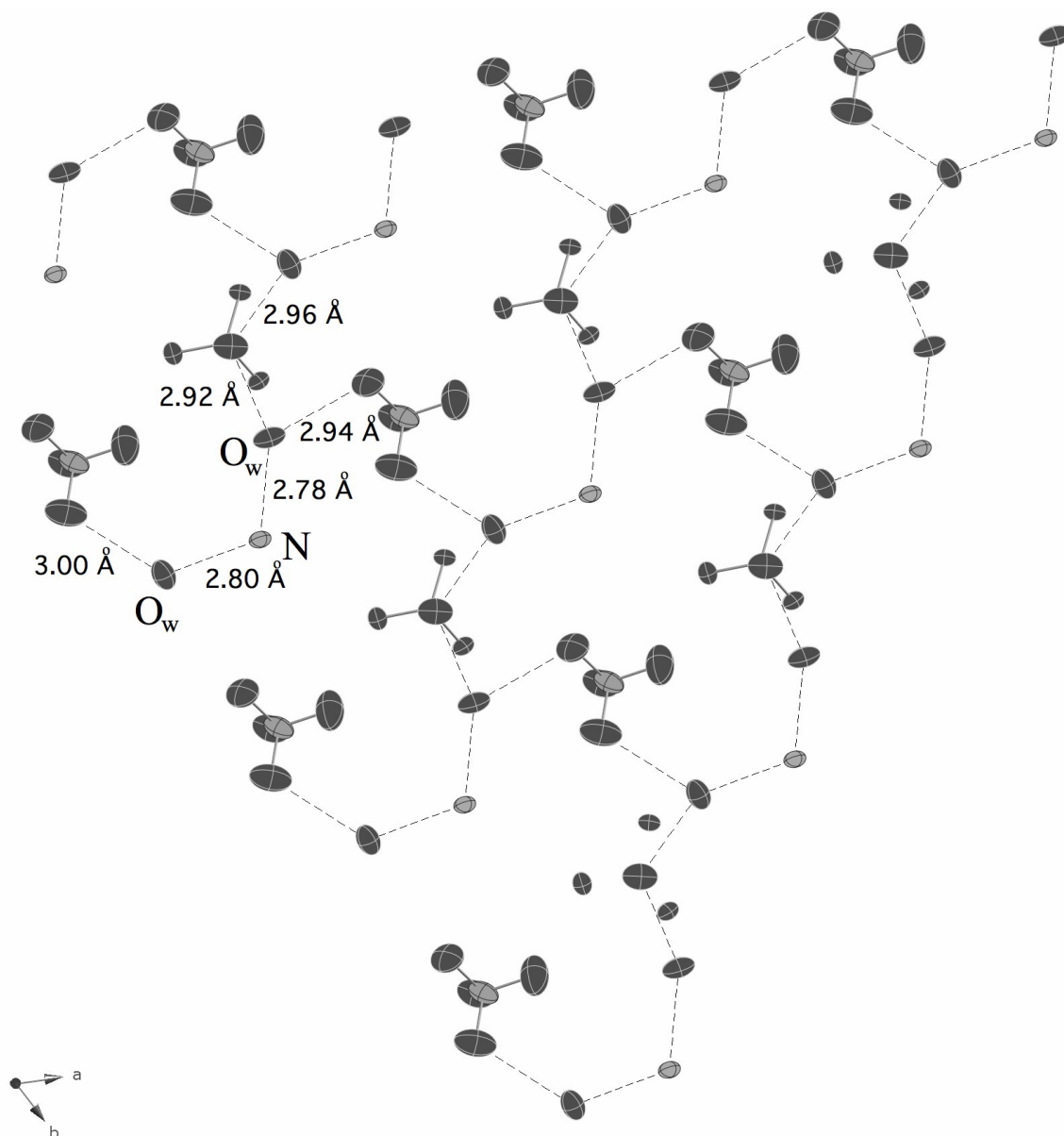


Fig. Supp. 1 Atom separations and the implied H-bonding network (dashed lines) within complex **1** in the vicinity of lattice water molecules, the protonated external amino group and its nearest anions. (View down *c*;  $O_w$  = water-molecule oxygen atoms; for clarity, only the external, protonated amino group nitrogen atoms (*N*) are shown; displacement ellipsoids are at the 50 % probability level.)

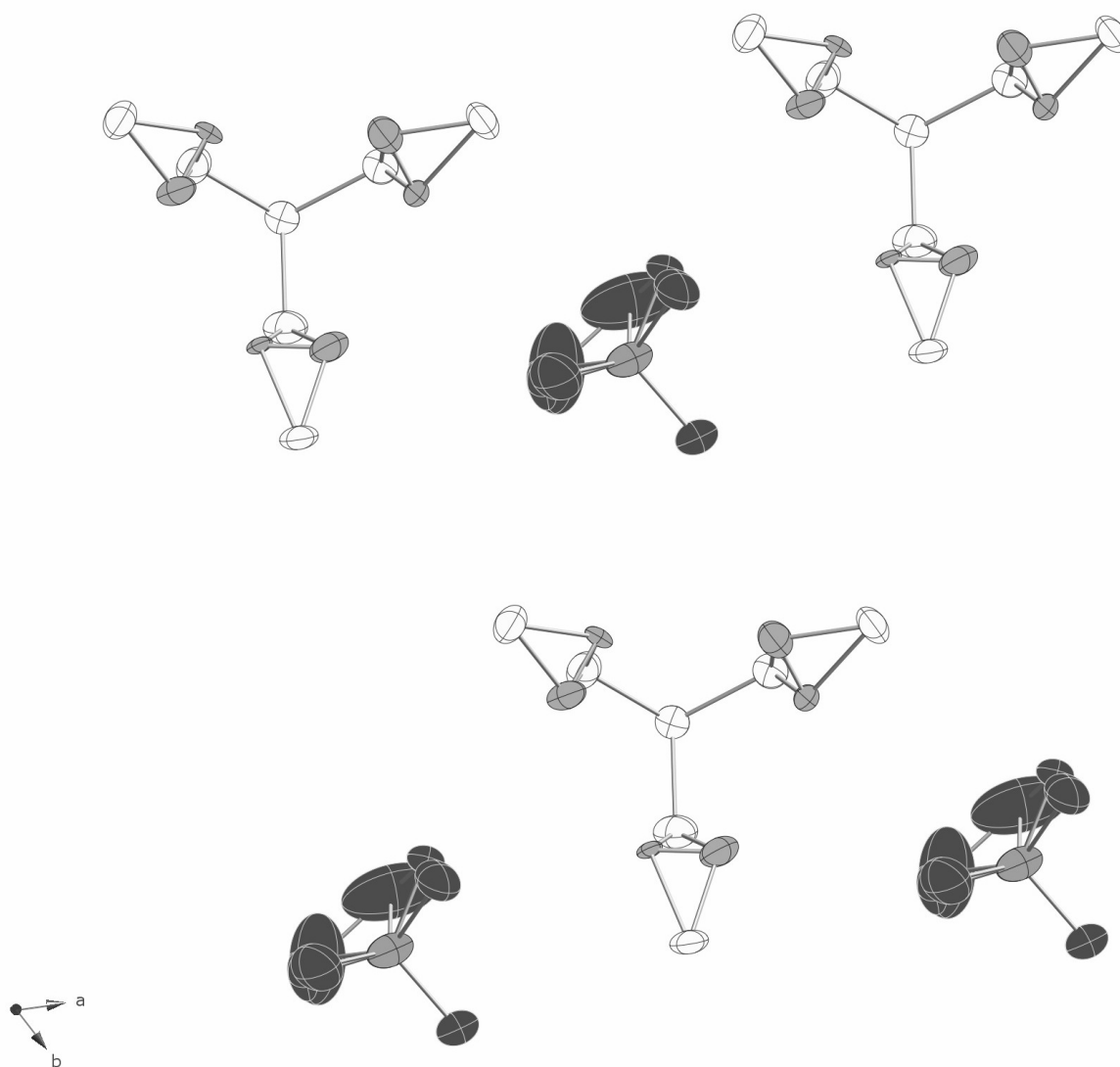


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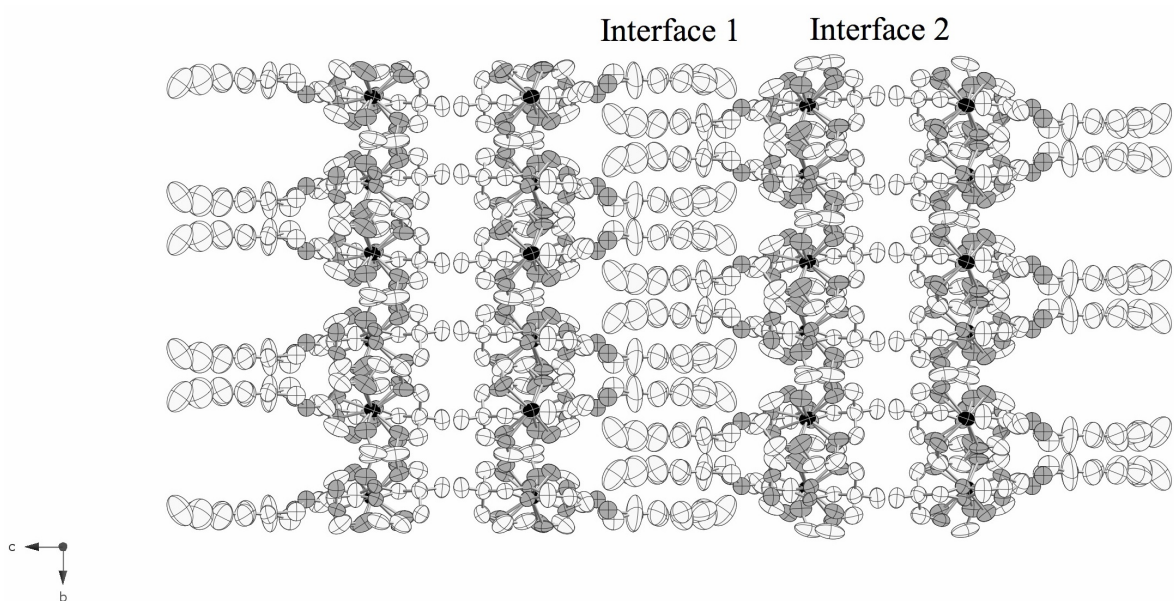


Fig. Supp. 3 A partial view of the lattice of complex **2** down *a* with perchlorate anions omitted, so that the presence of hydrocarbon moieties within both “interfaces” is more obvious. (*Cf.* Fig. 3 of main text.)  
(Displacement ellipsoids again at the 50 % level.)