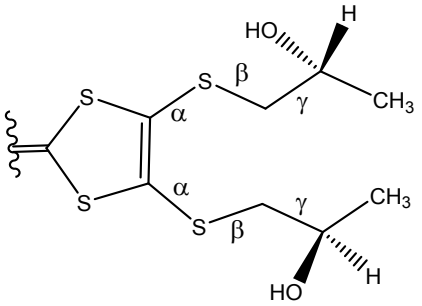
Supplementary Information.

Enantiopure and racemic radical-cation salts of bis(2'-hydroxylpropylthio)(ethylenedithio)TTF with polyiodide anions

Lee Martin,*[a] John D.Wallis,[a] Milena Guziak,[a] Peter Maksymiw,[a] Florence Konalian-Kempf,[a] Anthony Christian,[a] Shin'ichi Nakatsuji,[b] Jun'ichi Yamada[b] and Hiroki Akutsu[b,c]

[a] *School of Science and Technology, Nottingham Trent University, Clifton Lane, Nottingham, NG11 8NS, United Kingdom; E-mail: lee.martin@ntu.ac.uk*
[b] *Graduate School of Material Science, University of Hyogo, Kamigori-cho, Ako-gun, Hyogo, 678-1297, Japan*
[c] *Department of Chemistry, Graduate School of Science, Osaka University, 1-1 Machikaneyama-cho, Toyonaka, Osaka 560-0043, Japan*

Table S1. Torsion angles for the donor cation side chains in *RR*-**1**/ °, e.s.d.s in range 0.8-1.2°.



		Torsion α	Torsion β	Torsion γ
DONOR 1,	O1	-45.0	-78.2	-173.8
	O2	63.3	63.3	57.3
DONOR 2	O3	39.5	55.6	55.4
	O4	-72.3	-72.7	-165.8
DONOR 3	O5	-33.9	-90.5	81.6
	O6	119.9	-155.7	-51.8
DONOR 4	O7	57.6	74.0	70.2
	O8	-23.8	-178.8	66.0
DONOR 5	O9	-15.1	177.0	76.6
	O10	55.2	75.4	66.4
DONOR 6	O11	34.9	69.0	70.9
	O12	-98.3	133.8	173.5

Table S2. Torsion angles for the donor cation side chains in *S,S*-**2** / °, e.s.d.s 1-2°.

		Torsion α	Torsion β	Torsion γ
DONOR 1,	O1	40	64	172
	O2	-67	-81	-171
DONOR 2	O3	-57	-113	54
	O4	61	153	-168

Table S3. Torsion angles for the donor cation side chains in *SS*-**3** donor. / °, e.s.d.s 2-3°.

	Torsion α	Torsion β	Torsion γ
DONOR 1, O1	58	57	-173
O2	-9	169	174
DONOR 2 O3	-97	69	175
O4	7	168	174

Table S4. Torsion angles for the donor cation side chains in *rac*-**4**. / °, e.s.d.s 0.5-0.8°, main conformation only.

	Torsion α	Torsion β	Torsion γ
DONOR 1, O1	109.3	-82.4	-68.0
O2	12.6	-103.0	178.5

Table S5. Relative geometry of O atoms involved in hydrogen bonding for *R,R*-**1**, *S,*S-**2**, *S,S*-**3**, *rac*-**4**.

R,R-**1**	O(8)-H(8)··· O(3)-H(3)(x-1, y, z)··· O(6)-H(6)(x-1, y-1, z)··· O(9)-H(9)(x, y-1, z)	O(8)···O(3): 2.594(16) O(3)···O(6): 2.781(10) O(6)···O(9): 2.742(18) O(8)···O(3)···O(6): 98.6(5)° O(3)···O(6)···O(9): 125.4(3)°
	O(11)-H(11)··· O(2)-H(2)(x-1, y, z+1) ··· O(5)-H(5)(x, y, z+1)··· O(4)-H(4)(x-1, y+1, z+1)	O(11)···O(2): 2.940(13) O(2)···O(5): 2.832(16) O(5)···O(4): 2.914(13) O(11)···O(2)···O(5): 104.1(4)° O(2)···O(5)···O(4): 106.9(4)°
	O(10)-H(10)··· O(7)-H(7)(x, y + 1, z)··· O(12)-H(12)(x + 1, y + 1, z)	O(10)···O(7): 2.777(13) O(7)···O(12): 2.703(15) O(10)···O(7)···O(12): 117.1(4)°
S,S-**2**	O(3)-H(3)···O(4)-H(4)(x+1, y, z)··· O(1)-H(1)(x+1, y+1, z-1)···O(2)-H(2)(x+1, y+1, z-1)	O(3)···O(4): 2.890(14) O(4)···O(1): 2.652(14) O(1)···O(2): 2.619(14) O(3)···O(4)···O(1): 94.2(5)° O(4)···O(1)···O(2): 114.1(5)°
S,S-**3**	O(3)-H(3)···O(2)(x, y, z + 1) O(4)-H(4)···O(3) (x, y-1, z)	O(3)···O(2): 3.01(3) O(4)···O(3): 2.85(3) O(2)···O(3)···O(4): 121.3(9)
rac-**4**	O(1)-H(1)···O(2) (-x-1, -y, -z)	O(1)···O(2): 2.751(10)

Fig S1a Hydrogen bonding in ***R,R*-1**

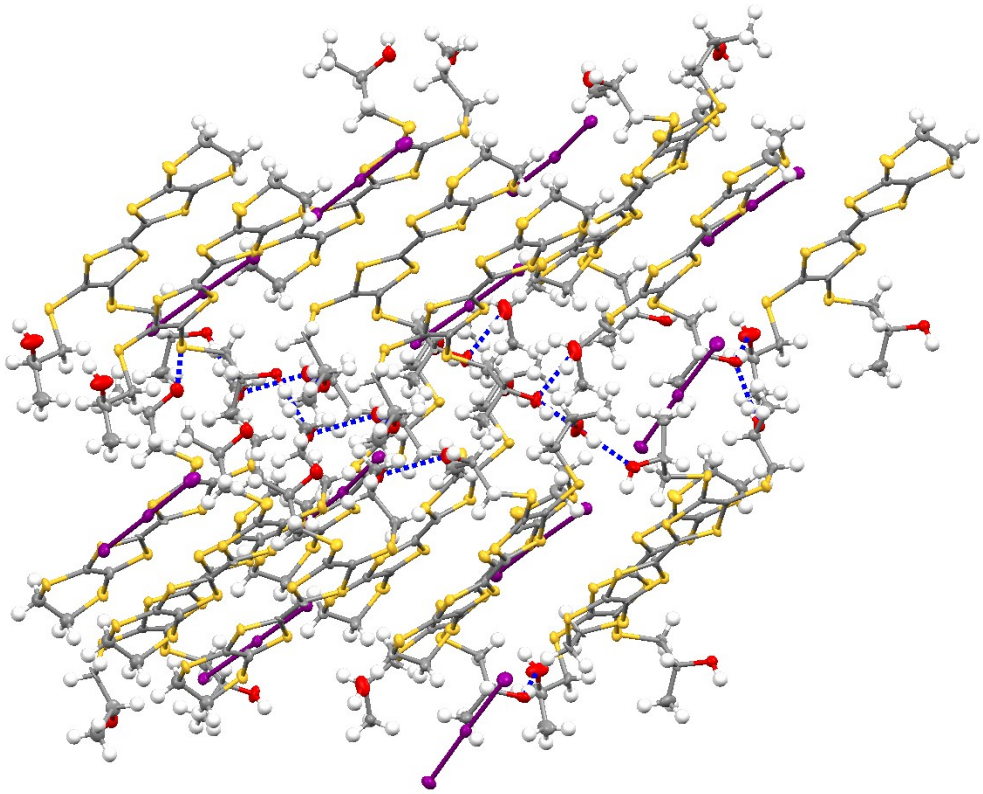


Fig S1b Hydrogen bonding in ***R,R*-1**

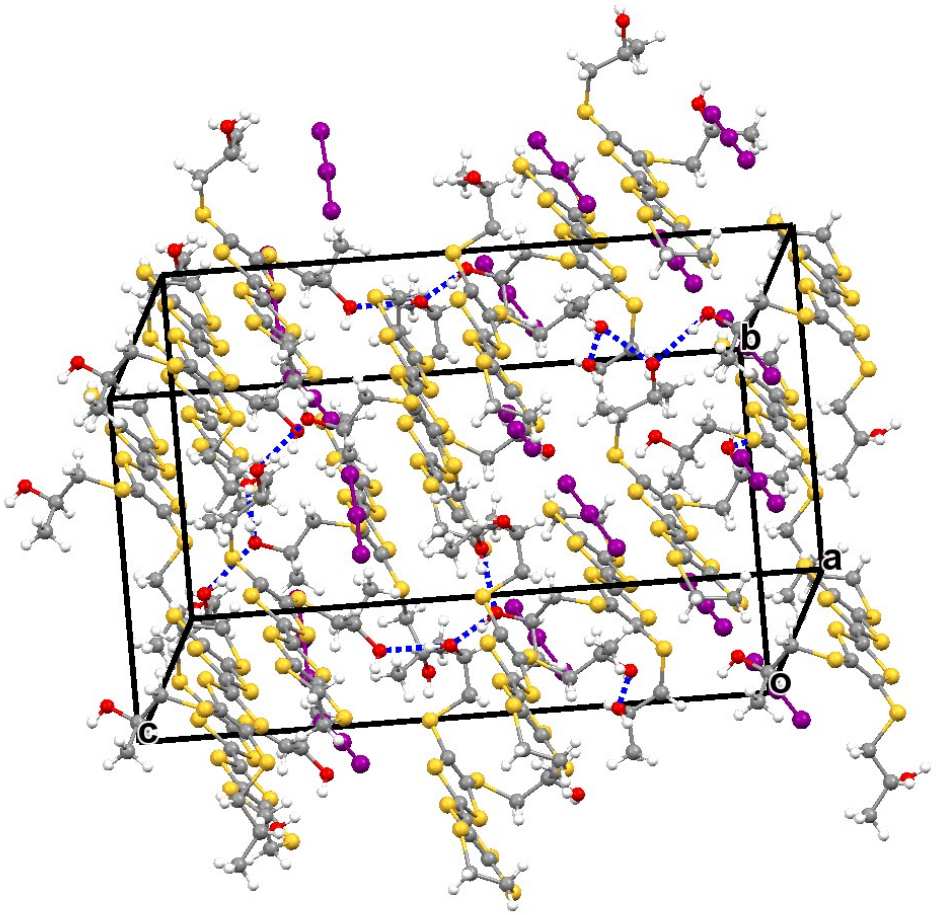


Fig S1c Hydrogen bonding in ***R,R*-1**

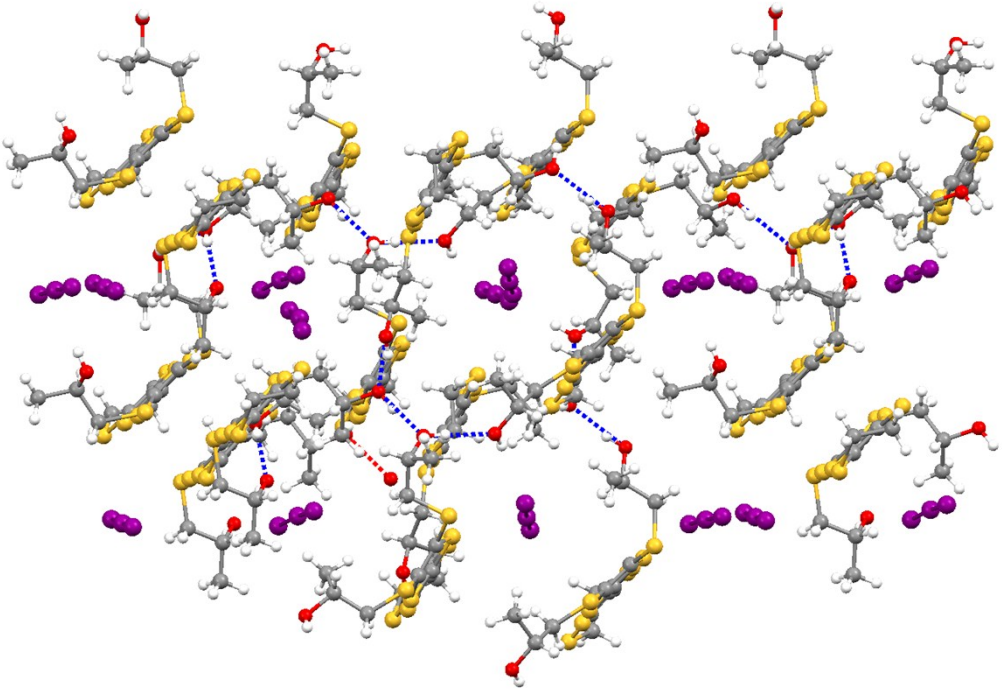


Fig S1d Hydrogen bonding in ***R,R*-1**

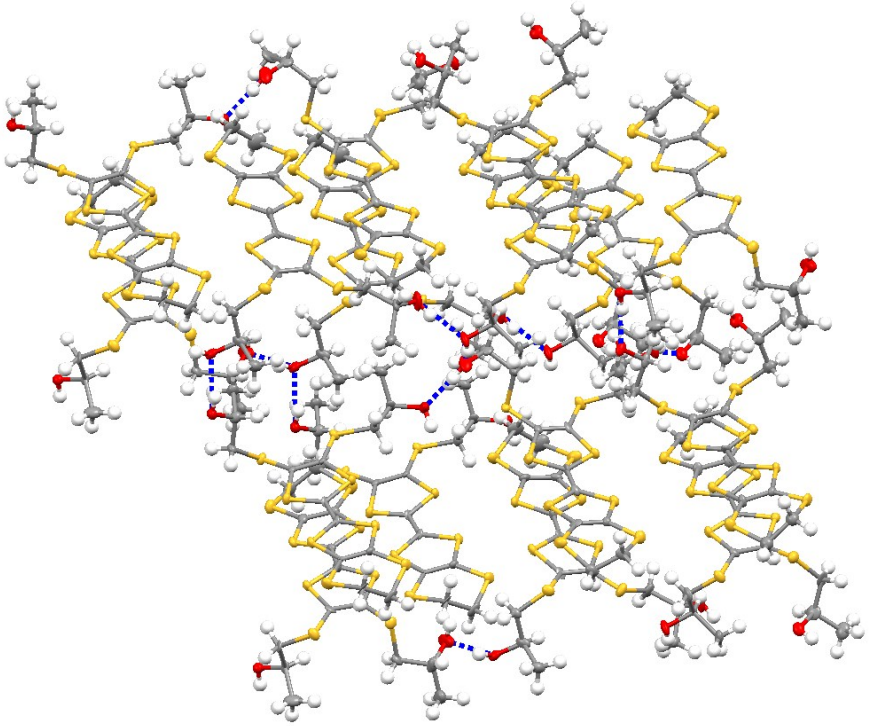


Fig S2. Hydrogen bonding in ***rac*-4**

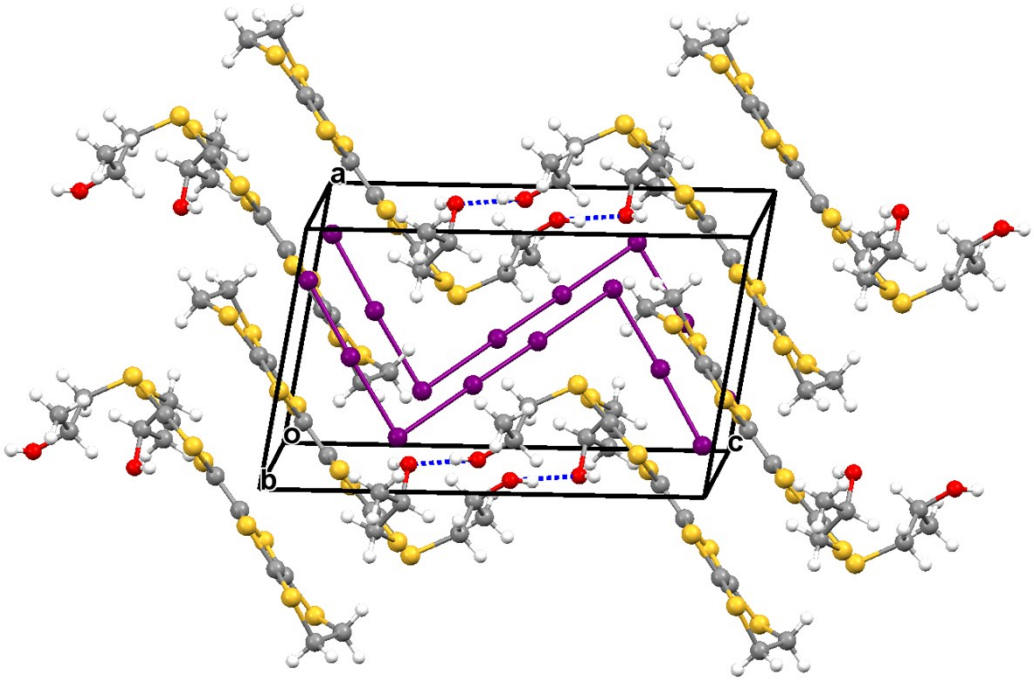


Fig S3a. Asymmetric unit of ***S,S*-1**.

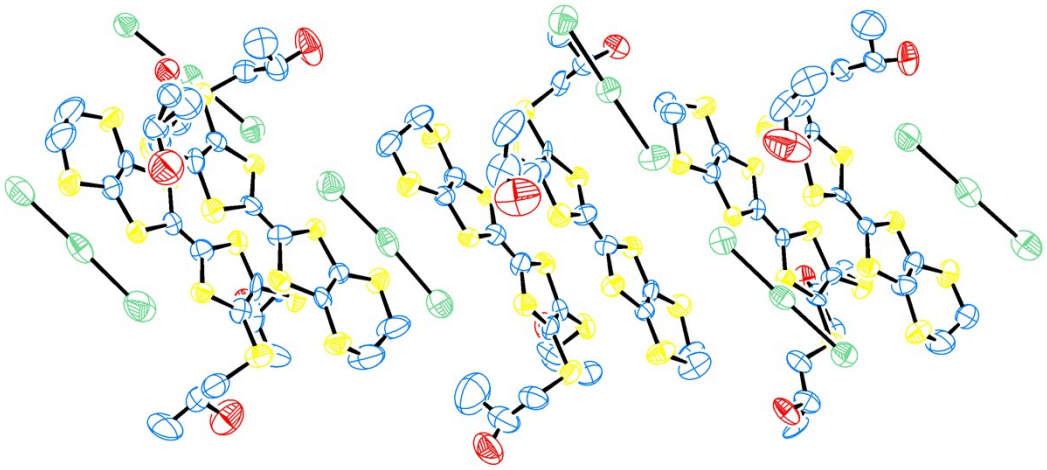


Fig S3b. Asymmetric unit of ***S,S*-2**.

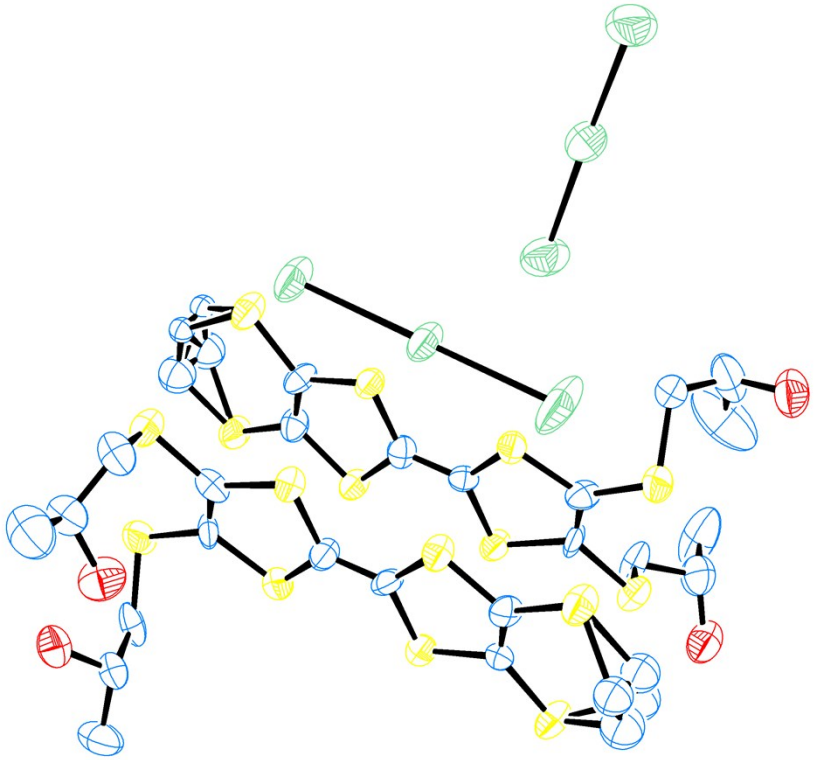


Fig S3c. Asymmetric unit of ***S,S*-3**; two parts of two different octaiodide dianions are shown in the asymmetric unit.

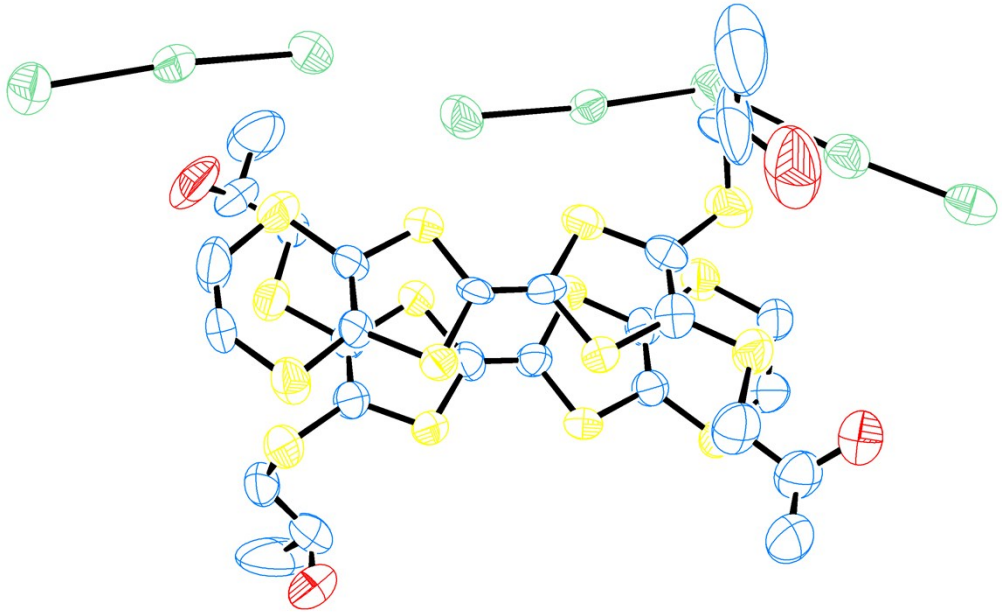


Fig S3d. Asymmetric unit of ***rac*-4**; the octaiodide dianion sits on a centre of symmetry so only half is crystallographically unique.