Electronic Supplementary Information

Bilayer conducting salts with polymeric anions

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Table S1. C–H···N≡C hydrogen bond parameters in $(CNB\text{-}EDT\text{-}TTF)_4A$ compounds with anions A. (**1-5**).

Anion	H···N [Å]	H···N - ΣVdW [Å]	C-H···N angle [°]	H_{type}
$Ag(CN)_2$ **(1)**	2.560	-0.190	157.59	H_α
	2.424	-0.326	152.52	H_β
$Ag(CN)_2 \cdot H_2O$ **(2)**	2.579	-0.171	155.88	H_α
	2.479	-0.271	152.43	H_β
$Ag_2(CN)_3$ **(3)**	2.614	-0.136	155.06	H_α
	2.509	-0.241	150.52	H_β
$Ag(SCN)_2$ **(4)**	2.583	-0.167	153.32	H_α
	2.457	-0.293	151.75	H_β
$Hg_2(SCN)_5$ **(5)**	2.623	-0.127	154.75	H_α
	2.536	-0.214	152.96	H_β

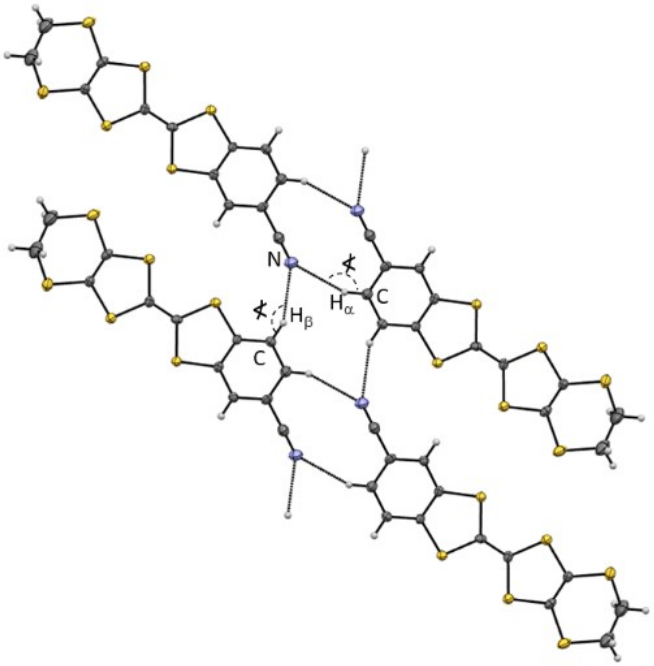
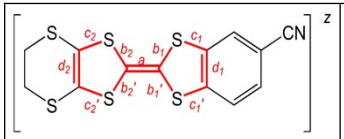
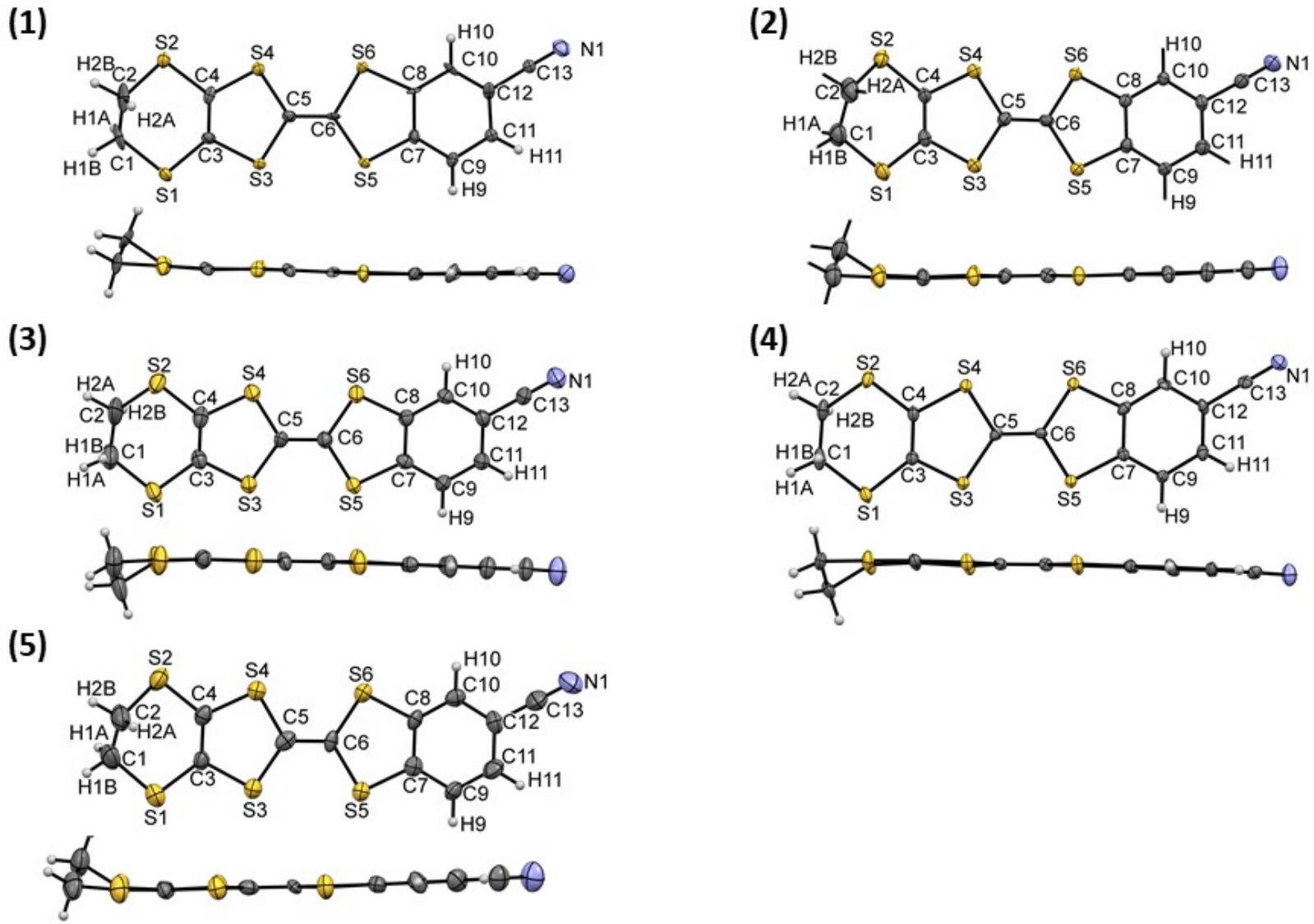


Table 1Scheme.
View of 4 donor molecules. Dash lines represent C–H···N≡C hydrogen bonds, with indication of C-H···N angles and H_α, H_b localization in $(CNB\text{-}EDT\text{-}TTF)_4A$ compounds.

Table S2. Bond lengths (Å) of the (CNB-EDT-TTF) donor in compounds $(CNB\text{-}EDT\text{-}TTF)_4A$ with anions A (**1-5**).

 A	a (Å)	b1 (Å)	b1′ (Å)	b2 (Å)	b2′ (Å)	c1 (Å)	c1′ (Å)	c2 (Å)	c2′ (Å)	d1 (Å)	d2 (Å)	δ=(b+c)-(a+d)
$Ag(CN)_2$ **(1)**	1.357(19)	1.760(16)	1.732(15)	1.746(17)	1.744(18)	1.766(14)	1.743(15)	1.721(16)	1.759(15)	1.42(2)	1.34(2)	0.75575
$Ag(CN)_2 \cdot H_2O$ **(2)**	1.327(12)	1.756(8)	1.763(8)	1.747(8)	1.768(8)	1.746(7)	1.749(8)	1.755(8)	1.751(8)	1.400(11)	1.335(12)	0.81425
$Ag_2(CN)_3$ **(3)**	1.352(6)	1.747(5)	1.741(5)	1.753(5)	1.733(5)	1.753(5)	1.741(5)	1.748(5)	1.748(5)	1.399(6)	1.352(7)	0.7635
$Ag(SCN)_2$ **(4)**	1.347(9)	1.749(6)	1.753(6)	1.750(6)	1.751(7)	1.751(7)	1.744(7)	1.756(7)	1.754(7)	1.396(9)	1.336(10)	0.789
$Hg_2(SCN)_5$ **(5)**	1.369(13)	1.729(10)	1.757(9)	1.738(10)	1.733(11)	1.751(9)	1.737(10)	1.752(10)	1.763(10)	1.401(14)	1.357(14)	0.742



Figures S1. ORTEP plots (50% probability level), with atom labeling scheme for the CNB-EDT-TTF molecules in compounds $(CNB\text{-}EDT\text{-}TTF)_4A$ with anions A (**1-5**) structure at 150 K. Hydrogen atoms are drawn as spheres with arbitrary radius. Up, top view, down, lateral view.

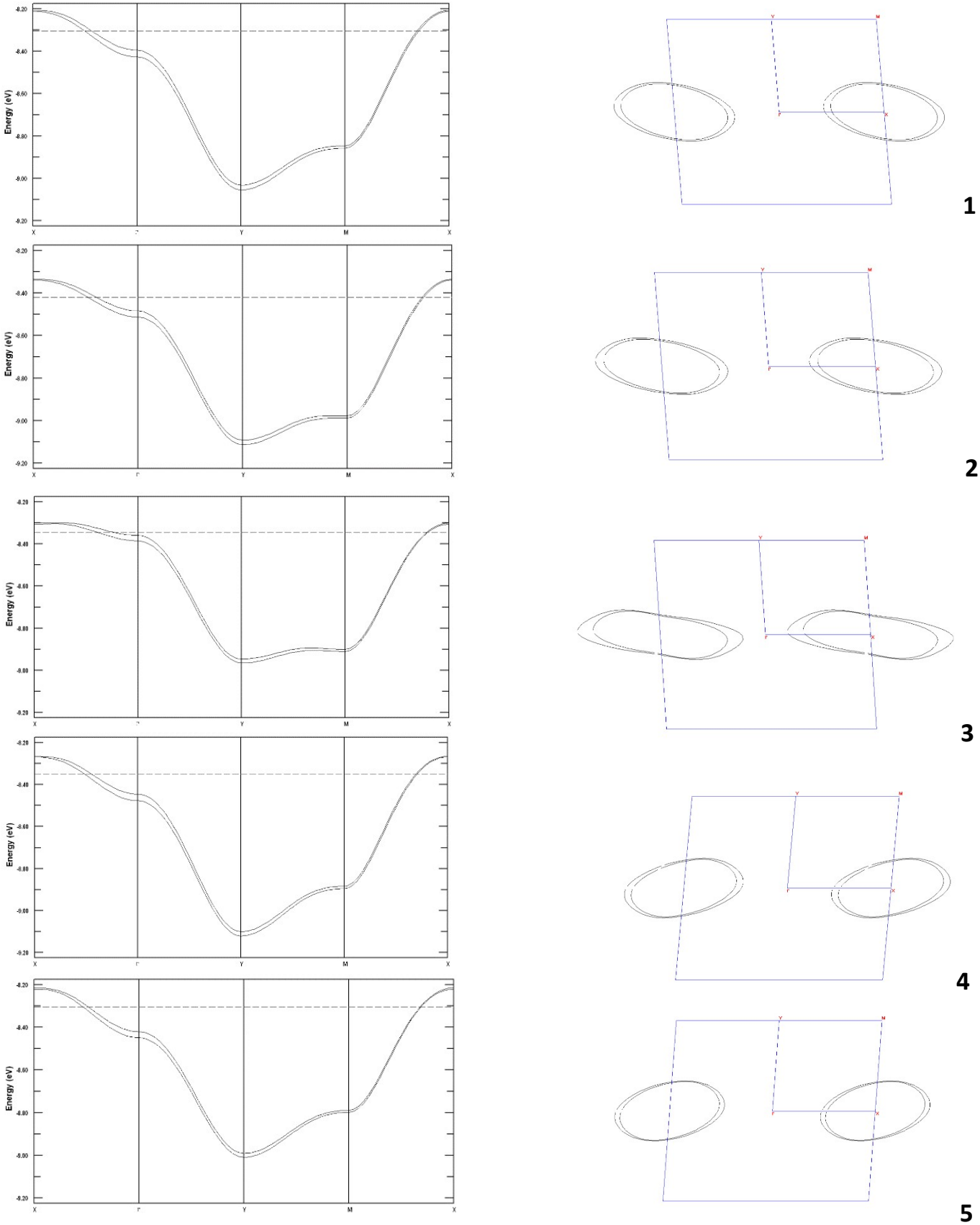


Figure S2. Calculated electronic band structures (left) and Fermi surfaces (right), for compounds **1** - **5** .
The dashed line represents the Fermi-level. Γ = (0, 0), X = (*a**/2, 0), Y = (0, *b**/2), and M = (*a**/2, *b**/2) except for **4** where, Y = (0, *c**/2), and M = (*a**/2, *c**/2).