β''-(CNB-EDT-TTF)$_4$BF$_4$; Anion disorder effects in bilayer molecular metals.

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Supplementary Materials

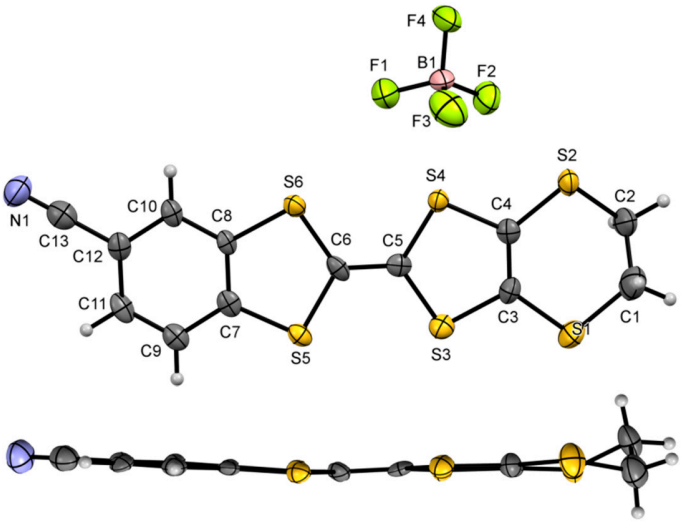

Figure S1. ORTEP and atomic numbering schemes (top and side views) **of (CNB-EDT-TTF)BF$_4$ (**1**).** Thermal ellipsoids drawn at 50 % probability level.

Table S1. Bond lengths (Å) in the crystal structure of (CNB-EDT-TTF)BF$_4$ (**1**).

S1-C3	1.742(6)	S1-C1	1.833(7)
S2-C4	1.744(6)	S2-C2	1.803(7)
S3-C5	1.720(6)	S3-C3	1.728(7)
S4-C5	1.715(6)	S4-C4	1.734(6)
S5-C6	1.739(6)	S5-C7	1.740(6)
S6-C6	1.732(6)	S6-C8	1.740(6)
N1-C13	1.138(9)	C1-C2	1.449(8)
C3-C4	1.366(8)	C5-C6	1.374(9)
C7-C8	1.387(8)	C7-C9	1.417(8)
C8-C10	1.401(8)	C9-C11	1.369(9)
C10-C12	1.401(8)	C11-C12	1.395(8)
C12-C13	1.473(10)		
F1-B1	1.390(10)	F2-B1	1.380(9)
F3-B1	1.403(10)	F4-B1	1.389(8)

Table S2. Short S···S , S···C and S···F contacts and hydrogen bonds in the crystal structure of (CNB-EDTTTF)BF_4 (**1**).

	Symm. op.	Length (Å)
S2···S4*	-x,1-y,1-z	3.540(2)
S2···S6*	-x,1-y,1-z	3.503(2)
S4···S4*	-x,1-y,1-z	3.575(2)
S3···S6*	-1-x,1-y,1-z	3.471(2)
S4···S5*	1-x,1-y,1-z	3.369(2)
S4···C7*	1-x,1-y,1-z	3.439(6)
S5···C9*	1-x,2-y,1-z	3.484(6)
C6···C6*	1-x,1-y,1-z	3.204(9)
C7···C7*	1-x,2-y,1-z	3.332(9)
S2···F2*	x,y,z	3.147(5)
S4···F2*	x,y,z	3.026(5)
F3···S5*	1-x,1-y,1-z	3.005(5)
B1···S5*	1-x,1-y,1-z	3.587(9)

Donor-H.....Acceptor		
C10-H10···N1*	-x+1,+y-1/2,-z+1/2	2.546 (136.31°)
C1-H1B···F4*	-x,-y,-z+1	2.667 (134.79°)
C2-H2A···F3*	-x,-y,-z+1	2.700 (129.54°)
C1-H1A···F4*	x,-y+1/2,+z+1/2	2.284 (141.06°)
C1-H1A···F1*	x,-y+1/2,+z+1/2	2.447 (144.27°)
C2-H2B···F1*	-x,-y+1,-z+1	2.672 (144.25°)
C9-H9···F2*	-x+1,-y+1,-z+1	2.735 129.57°)
C9-H9···F3*	-x+1,-y+1,-z+1	2.444 (139.15°)
C11-H11···F2*	x+1,+y+1,+z	2.429 (149.36°)
C11-H11···F4*	x+1,+y+1,+z	2.310 (142.60°)

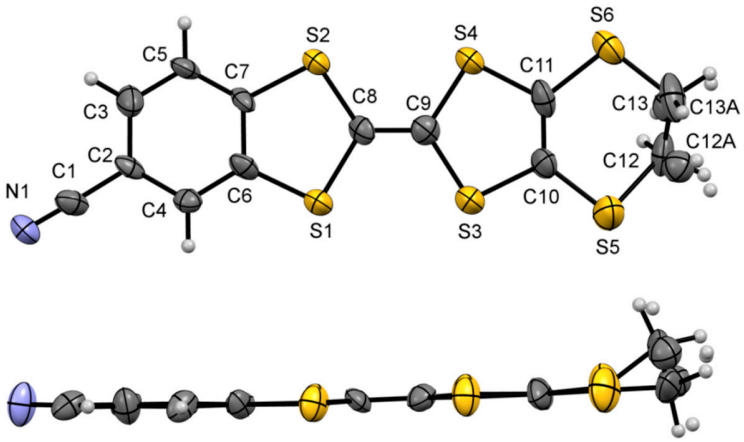

Figure S2. ORTEP and atomic numbering schemes (top and side views) **of (CNB-EDT-TTF)$_4$BF$_4$ (2).** Thermal ellipsoids drawn at 50 % probability level.

Table S3. Bond lengths(Å) of the cation and anion in the crystal structure of (CNB-EDT-TTF)4BF4 (**2**).

S1-C6	1.738(12)	S1-C8	1.774(11)
S2-C8	1.729(12)	S2-C7	1.734(11)
S3-C10	1.745(12)	S3-C9	1.747(12)
S4-C11	1.737(13)	S4-C9	1.736(12)
S5-C10	1.715(12)	S5-C12A	1.78(4)
S5-C12	1.90(3)	S6-C13	1.72(3)
S6-C11	1.746(12)	S6-C13A	1.93(6)
N1-C1	1.132(14)	C1-C2	1.430(18)
C2-C3	1.390(15)	C2-C4	1.410(15)
C3-C5	1.352(15)	C4-C6	1.386(15)
C5-C7	1.387(15)	C6-C7	1.404(15)
C8-C9	1.341(15)	C10-C11	1.367(16)
C12-C13	1.49(6)	C12A-C13A	1.43(9)

Table S4. Short S···S and S···C contacts and hydrogen bonds in the crystal structure of (CNB-EDTTTF)4BF4 (**2**).

	Symm. op.	Length (Å)
S5···S6*	x,y,-1+z	3.575(5)
S5···C13A*	x,y,-1+z	3.47(6)
Donor-H.....Acceptor		
C3 –H3···N1*	3-x,1-y,1-z	2.624 (142.7°)
C5 –H5···N1*	x-1,+y,+z+1	2.506 (153.4°)
C12–H12C···S5*	x-1,+y,+z	2.955 (177.2°)
C13–H13B···S5*	x-1,+y,+z	2.963 (162.4°)
C13–H13B···C10*	x-1,+y,+z	2.756 (148.3°)