Supplementary Information

Martensitic phase transition and stimuli responsive effects in thermosalient cocrystal of 9,10-dimethylanthracene with F_2TCNQ

Kamil Ivshin,[a] Anton Fedonin,[a] Kirill Metlushka,[a] Dmitry Zakharychev,[a] Nadir Garif'yanov,[b] Andrey Kamashev,[b] Airat Kiiamov,[a] Anastasia Efimova,[c] Marco Naumann,[d] Stanislav Avdoshenko,[d] Sandra Schiemenz,[d] Alexey A. Popov,[d] Vladislav Kataev,[d] Martin Knupfer[d] and Olga Kataeva*[a]

[a]Arbuzov Institute of Organic and Physical Chemistry, FRC Kazan Scientific Center, Russian Academy of Sciences, Arbuzov str. 8, 420088 Kazan, Russian Federation

[b]Zavoisky Physical-Technical Institute, FRC Kazan Scientific Center, Russian Academy of Sciences, Sibirsky tr. 10/7, 420029 Kazan, Russian Federation

[c]Chair of Inorganic Chemistry, Brandenburg University of Technology (BTU) Cottbus – Senftenberg, 01968, Senftenberg, Germany

[d]Leibniz Institute for Solid State and Materials Research, Helmholtzstraße 20, 01069 Dresden, Germany

*Corresponding author:

Email: olga-kataeva@yandex.ru

Table of contents:

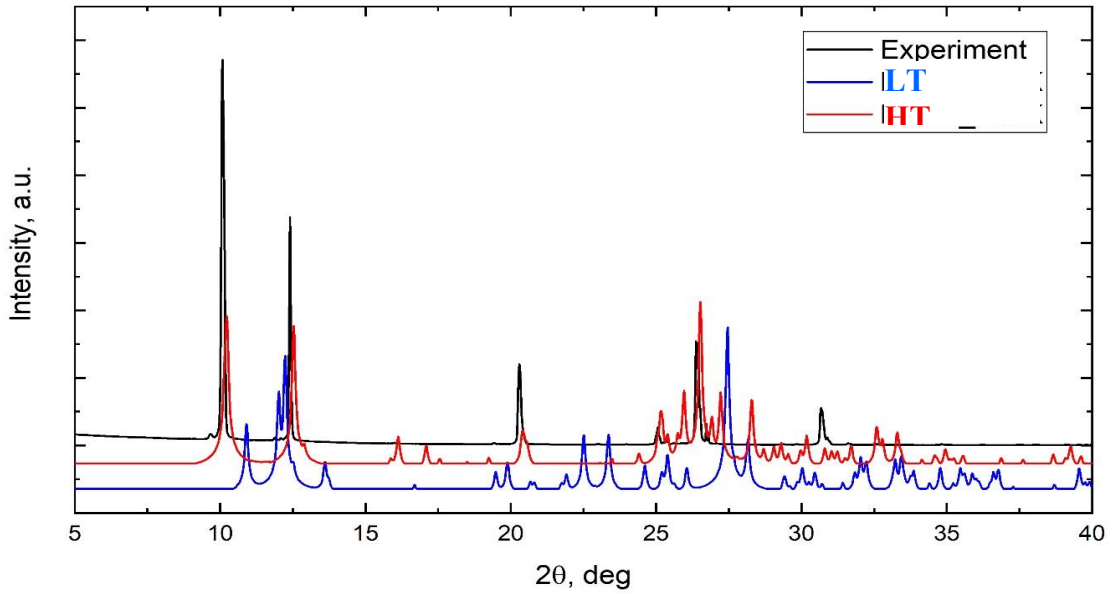


Figure S1. Powder XRD pattern collected at 300 K in comparison with XRD pattern simulated from single crystal data for **HT** and **LT** polymorphs.

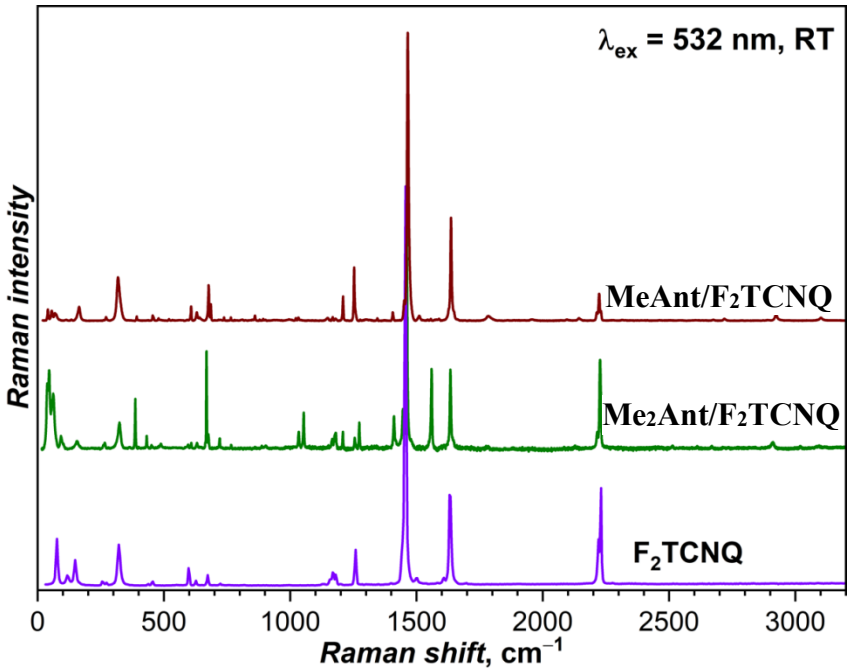
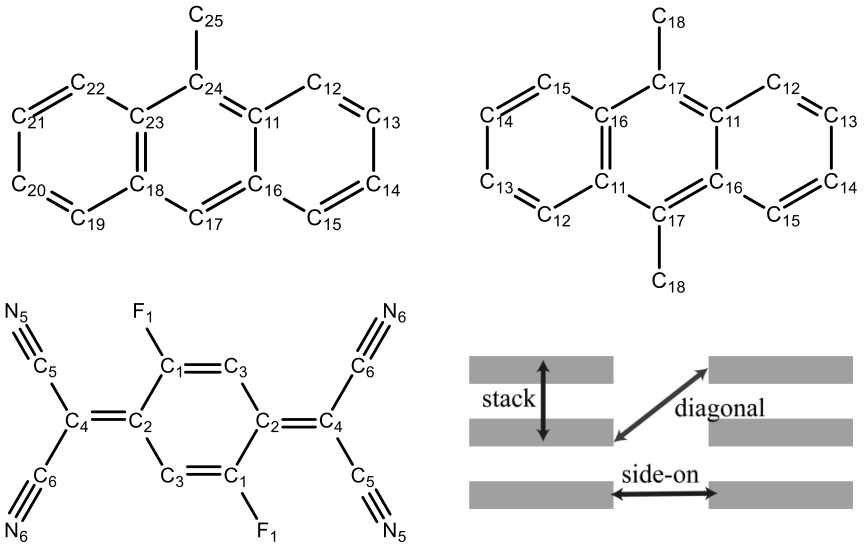


Figure S2. Raman spectra for MeAnt/F_2TCNQ and Me_2Ant/F_2TCNQ cocrystals in comparison with F_2TCNQ.

Table S1. Crystal data and structure refinement for cocrystals of Me_2Ant/F_2TCNQ.

	LT	**HT**
Chemical formula	$C_{28}H_{16}F_2N_4$	$C_{28}H_{16}F_2N_4$
Formula weight (g mol–1)	446.45	446.45
Radiation, wavelength (Å)	0.71073	0.71073
Temperature (K)	100.0(5)	300(2)
Crystal system	Triclinic	Triclinic
Space group	$P\bar{1}$	$P\bar{1}$
a, b, c (Å); α, β, γ (deg)	7.8372(4), 9.0718(4), 9.2216(4); 106.995(2), 102.260(2), 115.527(2)	6.9487(6), 8.9390(7), 9.0709(7); 103.910(3), 97.390(3), 92.321(3)
Volume (Å^3)	520.51(4)	540.87(8)
Z and Z'	1 and 0.5	1 and 0.5
Calculated density (g cm^{-3})	1.424	1.371
F(000)	230	230
Absorption coefficient (mm^{-1})	0.099	0.095
Crystal size (mm^3)	0.301 x 0.168 x 0.122	0.231 x 0.113 x 0.112
Absorption correction	Multi-scan	Multi-scan
Max. and min. transmission	0.988 and 0.971	0.7458 and 0.7085
θ range for data collection θ (deg)	2.510 to 55.838	2.964 to 28.777
Resolution range sin(θ)/λ, (Å^{-1})	0.062 to 1.164	0.073 to 0.677
Index ranges	$-17 \le h \le 18$, $-21 \le k \le 21$ $-21 \le l \le 21$	$-9 \le h \le 9$, $-12 \le k \le 12$ $-12 \le l \le 12$

Completeness to θ_{max}	99.3%	97.8%
	LT	**HT**
Reflections collected	247363	35453
Independent reflections	13667	2755
R_{int}	0.0338	0.0351
Rσ	0.0122	0.0155
Spherical atom refinement (SHELX)		
Refinement method	Full-matrix least-squares on F^2	Full-matrix least-squares on F^2
Observed Data [I ≥ 2σ(I)]	10409	2296
Parameters	155	155
Goodness-of-fit on F^2	1.052	1.069
Final R indices for I ≥ 2σ(I) [all data]	$R(F^2)$ = 0.0432 [0.0570], $wR(F^2)$ = 0.1451 [0.1575]	$R(F^2)$ = 0.0538 [0.0606], $wR(F^2)$ = 0.1695 [0.1756]
Largest diff. peak and hole [r.m.s] (e Å^{-3})	0.807 (–0.535) [0.133]	0.233 (–0.135) [0.030]
Max shift/e.s.d. in the last cycle	0.001	0.001
Multipole refinement (MoPro)		
Refinement method	Full-matrix least-squares on F	
Observed Data [I ≥ 2σ(I)]	13644	
Parameters	387	
Final R indices for I ≥ 2σ(I)	R(F) = 0.0469, wR(F) = 0.0200 $R(F^2)$ = 0.0269, $wR(F^2)$ = 0.0369	
Largest diff. peak and hole [r.m.s] (e Å^{-3})	0.558 (–0.398) [0.129]	
Max shift/e.s.d. in the last cycle	0.004	



Scheme S1. Numbering scheme of donors and acceptor molecules, and the directions of interactions in crystals of Me_2Ant/F_2TCNQ.

Table S2. Selected geometric parameters and theoretical quantum-topological properties at the bond critical points of non-covalent interactions for Me_2Ant/F_2TCNQ single crystal (symmetry independent part) at **LT**. The first line shows data for experimental multipole refinement model, the second line is for theoretical calculations, the third line shows data for multipole modeling.

Intramolecular non-covalent interaction								
	Geometry		(3, –1) in ρ					
Interaction atoms	d_1	d_2	ρ_b	$\nabla^2\rho_b$	$G_b\ 10^3$	$-V_b\ 10^3$	$H_b\ 10^3$	E_b
F1···C5	1.266	1.328	0.10474	1.69	14.44	11.38	3.06	3.57
	1.290	1.304	0.1006	1.7305	14.34	10.73	3.61	3.37
	1.289	1.310	0.095	1.68	13.97	10.49	3.48	3.29
H12···C18	0.997	1.560	0.106	1.477	12.411	10.459	1.952	3.28
H12···H18A	1.051	1.039	0.1089	1.3504	12.10	10.19	1.91	3.26
	0.997	0.995	0.123	1.42	13.40	12.11	1.29	3.80
H15···C18	1.003	1.542	0.114	1.326	11.79	10.69	1.10	3.35
H15···H18B	1.166	1.362	0.0966	1.2271	11.25	9.76	1.49	3.06
	1.056	1.124	0.110	1.29	11.87	10.42	1.45	3.27
Intermolecular non-covalent interaction								
***F_2TCNQ-F_2TCNQ* interactions**								
N6···H3[a]	1.418	0.984	0.068	1.139	8.74	6.41	2.33	2.01
	1.505	0.919	0.0680	0.9184	7.23	4.94	2.29	1.55
	1.501	0.913	0.039	1.11	7.23	4.93	2.30	1.55
N5···C6[b]	1.584	1.609	0.038	0.538	3.99	2.75	1.24	0.86
	1.591	1.625	0.0446	0.5640	4.73	3.61	1.12	1.13
	1.568	1.591	0.041	0.57	4.50	3.11	1.39	0.98
C5···C5[b]	1.727	1.727	0.043	0.745	5.47	3.70	1.77	1.16
	1.986	1.986	0.0356	0.4185	3.49	2.63	0.86	0.83

	1.703	1.703	0.033	0.42	3.30	2.27	1.03	0.71
N6···N6[c]	1.614	1.614	0.033	0.499	3.64	2.42	1.22	0.76
	1.605	1.605	0.0384	0.5572	4.51	3.23	1.28	1.01
	1.600	1.600	0.035	0.51	3.94	2.64	1.30	0.83
Sum								**5.79**
								4.52
								4.07
***F₂TCNQ-Me₂Ant* side-on interactions**								
F1···H13[d]	1.354	0.988	0.049	1.079	7.77	5.05	2.73	1.58
	1.371	1.018	0.0539	0.9362	7.17	4.64	2.53	1.46
	1.370	1.027	0.044	0.98	7.39	4.67	2.73	1.47
N5···H14[e]	1.556	1.058	0.043	0.745	5.47	3.70	1.77	1.16
	1.590	1.030	0.0595	0.7352	6.17	4.71	1.46	1.48
	1.536	1.047	0.047	0.81	6.31	4.25	2.06	1.33
N5···H12[d]	1.604	1.048	0.026	0.643	4.44	2.62	1.82	0.82
	1.632	1.088	0.0376	0.4674	3.66	2.47	1.19	0.77
	1.654	1.074	0.016	0.55	3.92	2.15	1.77	0.67
N5···H18A[f]	1.628	1.250	0.028	0.403	2.92	1.91	1.00	0.60
	1.656	1.120	0.0381	0.4398	3.53	2.49	1.04	0.78
	1.656	1.135	0.020	0.47	3.44	1.98	1.46	0.62
N6···H12[g]	1.706	1.441	0.021	0.310	2.20	1.40	0.81	0.44
	1.719	1.435	0.0249	0.3069	2.48	1.84	0.64	0.56
	1.700	1.377	0.020	0.31	2.33	1.42	0.91	0.45
N6···H18C[c]	1.486	0.951	0.056	0.847	6.47	4.70	1.77	1.47
	1.776	1.420	0.0229	0.2962	2.37	1.66	0.71	0.52
	1.755	1.385	0.020	0.29	2.20	1.36	0.84	0.43
N6···H14[e]	1.848	1.242	0.020	0.345	2.42	1.48	0.94	0.46

	2.360	1.262	0.0247	0.2773	2.25	1.62	0.63	0.51
	1.872	1.289	0.017	0.29	2.17	1.29	0.88	0.40
N6···H13[e]	1.809	1.466	0.013	0.217	1.50	0.88	0.61	0.28
	1.811	1.393	0.0164	0.2070	1.62	1.09	0.53	0.34
	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
H3···H18C[e]	1.408	1.322	0.017	0.248	1.74	1.07	0.67	0.34
	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.518	1.364	0.007	0.17	1.23	0.66	0.57	0.21
Sum								**7.15**
								6.42
								5.58
***F₂TCNQ-Me₂Ant* stack interactions**								
C4···C16	1.691	1.730	0.043	0.509	3.94	2.93	1.01	0.92
	1.685	1.729	0.0429	0.4330	4.05	3.61	0.44	1.09
	1.696	1.710	0.040	0.43	3.51	2.58	0.93	0.81
C3···C11[h]	1.767	1.681	0.048	0.539	4.27	3.29	0.98	1.03
	1.734	1.646	0.0446	0.4610	4.29	3.79	0.50	1.19
	1.680	1.667	0.044	0.45	3.75	2.86	0.88	0.90
N5···C12	n.f.	n.f.	n.f.	n.f.	n.f.	n.f.	n.f.	n.f.
	1.835	2.133	0.0288	0.3539	2.91	2.15	0.76	0.67
	1.806	1.851	0.028	0.32	2.50	1.71	0.79	0.54
C1···C15[i]	1.722	1.870	0.045	0.493	3.87	2.95	0.92	0.92
	1.722	1.899	0.0422	0.4156	3.80	3.29	0.51	1.03
	1.719	1.848	0.040	0.42	3.48	2.59	0.89	0.81
C2···C16	1.683	1.737	0.041	0.41	3.67	2.67	1.00	0.84
	1.772	1.683	0.0415	0.4252	3.90	3.39	0.51	1.06
	1.683	1.736	0.041	0.41	3.39	2.55	0.84	0.71

Sum								**3.71**
								5.04
								3.77
***F_2TCNQ-Methyl* interactions within stack**								
F1···C18^{h}	1.490	1.890	0.040	0.553	4.15	2.92	1.23	0.91
C3···H18B^{i}	1.208	2.200	0.0438	0.4196	3.88	3.40	0.48	1.07
C1···H18B^{i}	1.720	1.250	0.040	0.49	3.95	2.81	1.14	0.88
N6···C18	1.672	2.127	0.033	0.350	2.67	1.95	0.73	0.61
N6···H18C	1.500	1.011	0.0659	0.8042	6.82	5.30	1.52	1.66
	1.499	1.036	0.054	0.86	6.87	4.83	2.04	1.52
Sum								**1.52**
								2.73
								2.40
***Me_2Ant-Me_2Ant* interactions**								
C18···C18^{c}	1.834	1.834	0.049	0.557	4.40	3.38	1.02	1.06
H18B···H18B^{c}	1.343	1.343	0.0424	0.4626	4.17	3.53	0.64	1.12
	1.258	1.258	0.036	0.54	4.23	2.80	1.43	0.88
H13···H13^{d}	1.544	1.544	0.011	0.162	1.12	0.66	0.46	0.21
	1.614	1.614	0.0110	0.1520	1.17	0.77	0.40	0.24
	1.534	1.534	0.007	0.14	1.00	0.55	0.45	0.17
C14···C14^{j}	1.822	1.822	0.027	0.318	2.36	1.62	0.74	0.51
	1.897	1.897	0.0312	0.3489	3.04	2.47	0.57	0.77
	1.781	1.781	0.028	0.35	2.72	1.83	0.89	0.57
Sum								**1.78**
								2.13
								1.62

Symmetry codes are as follows: *a* = –*x*, 1 – y, –*z*, *b* = –*x*, 1 – *y*, 1 – *z*, *c* = –*x*, 1 – *y*, –*z*, *d* = –*x*, –*y*, 1 – *z*, *e* = 1 – *x*, 1 – *y*, 1 – *z*, *f* = *x*, *y*, 1 + *z*, *g* = *x*, 1 + *y*, *z*, *h* = –*x*, –*y*, –*z*, *i* = *x* – 1, *y*, *z*, *j* = –*x*, 1 – *y*, 1 – *z* The properties are presented in the following units: distances d_1, d_2 [Å]; electron density

ρ_b [e Å^{-3}]; Laplacian of electron density $\nabla^2\rho_b$ [e A^{-5}]; kinetic, potential and total energy densities G_b, V_b, H_b [H Bohr^{-3}]; Interaction energy (Using electron potential energy at BCP and Espinosa equation) E_b kcal/mol; n.f – not found.

Table S3. Selected geometric parameters and theoretical quantum-topological properties at the bond critical points of non-covalent interactions for Me_2Ant/F_2TCNQ single crystal (symmetry independent part) at **HT**. The first line is for theoretical calculations; the second line shows data for multipole modeling.

Intramolecular non-covalent interaction								
	Geometry		(3, –1) in ρ					
Interaction atoms	d_1	d_2	ρ_b	$\nabla^2\rho_b$	$G_b\ 10^3$	$-V_b\ 10^3$	$H_b\ 10^3$	E_b
F1···C5	1.303	1.316	0.0953	1.6349	13.70	9.98	3.72	3.13
	1.300	1.320	0.087	1.61	13.15	9.62	3.53	3.02
H12···H18B	1.045	1.049	0.1079	1.3305	11.95	10.10	1.85	3.17
	0.993	0.995	0.120	1.40	13.14	11.80	1.34	3.34
H15···H18C	1.206	1.878	0.1004	1.2561	11.53	10.02	1.51	3.14
	1.030	1.102	0.113	1.30	12.17	10.83	1.34	3.40
Intermolecular non-covalent interaction								
***F₂TCNQ-F₂TCNQ* interactions**								
N5···N6[a]	1.747	1.756	0.0217	0.3027	2.29	1.44	0.75	0.45
	1.723	1.778	0.019	0.28	2.09	1.29	0.80	0.40
N6···H3[b]	1.479	0.902	0.0744	1.0114	8.07	5.65	2.42	1.77
	1.472	0.901	0.046	1.23	9.20	5.64	3.56	1.77
N6···N6[b]	1.627	1.627	0.0354	0.5029	4.03	2.84	1.19	0.89
	1.621	1.621	0.031	0.46	3.53	2.31	1.22	0.72
F1···N5[c]	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.807	1.990	0.006	0.10	0.71	0.39	0.32	0.12
Sum								**3.11**
								3.01

***F₂TCNQ-Me₂Ant* side-on interactions**								
N5···H12[c]	1.651	1.125	0.0371	0.4469	3.58	2.53	1.05	0.79
	1.653	1.120	0.018	0.490	3.57	2.01	1.56	0.63
F1···H13[c]	1.503	1.208	0.0278	0.4155	3.21	2.11	1.10	0.66
	1.513	1.216	0.021	0.45	3.31	1.93	1.38	0.61
N5···H18B[d]	1.526	0.978	0.0618	0.7844	6.27	4.40	1.87	1.38
	1.521	0.979	0.04	0.90	6.75	4.20	2.55	1.32
N5···H14[a]	1.610	1.075	0.0456	0.5583	4.53	3.28	1.25	1.03
	1.597	1.086	0.031	0.62	4.66	2.88	1.78	0.90
N6···H18A[b]	1.638	1.102	0.0382	0.4308	3.43	2.39	1.04	0.75
	1.651	1.075	0.019	0.53	3.80	2.14	1.66	0.67
N6···H13[a]	1.852	1.416	0.0129	0.1680	1.27	0.79	0.48	0.25
	1.901	1.455	0.009	0.16	1.13	0.63	0.50	0.20
N6···H14[a]	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	2.103	1.563	0.008	0.12	0.87	0.49	0.38	0.15
N6···H18C[e]	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.929	1.600	0.007	0.13	0.90	0.50	0.40	0.02
Sum								**4.86**
								4.50
***F₂TCNQ-Me₂Ant* stack interactions**								
C3···C17[f]	1.781	1.851	0.0399	0.4010	3.69	3.22	0.47	1.01
	1.742	1.702	0.041	0.38	3.32	2.47	0.76	0.77
C3···C17	1.805	2.052	0.0378	0.3714	3.39	2.93	0.46	0.92
	1.768	1.799	0.037	0.36	2.98	2.24	0.74	0.70
C5···C14[f]	1.806	1.854	0.0309	0.3425	2.92	2.28	0.64	0.72
	1.740	1.779	0.03	0.32	2.56	1.79	0.77	0.56
C4···C14[f]	1.709	2.286	0.0408	0.3843	3.60	3.21	0.39	1.01

	1.715	1.847	0.038	0.38	3.31	2.33	0.79	0.73
C4···C15	1.738	1.757	0.0375	0.3725	3.45	3.03	0.42	0.95
	1.742	1.742	0.036	0.35	2.90	2.14	0.76	0.67
C2···C16	1.815	1.749	0.0368	0.3938	3.53	2.98	0.55	0.93
	1.747	1.709	0.037	0.35	2.93	2.21	0.73	0.69
Sum								**5.54**
								4.12
***F₂TCNQ-Methyl* interactions within stack**								
N6···H18C	2.337	1.291	0.0219	0.2321	1.89	1.37	0.52	0.43
C6···H18C	1.856	1.306	0.013	0.24	1.76	1.01	0.75	0.32
***Me₂Ant-Me₂Ant* interactions**								
H12···H13^{g}	1.393	1.910	0.0216	0.2496	2.08	1.57	0.61	0.49
	1.381	1.483	0.016	0.26	1.92	1.15	0.77	0.36
H14···H14^{h}	1.464	1.464	0.0165	0.1872	1.53	1.11	0.42	0.35
	1.414	1.414	0.010	0.20	1.47	0.81	0.66	0.25
C18···H15^{i}	1.206	1.878	0.0245	0.2893	2.38	1.76	0.62	0.55
	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
H18C···H18C^{i}	1.587	1.587	0.0239	0.3034	2.55	1.96	0.59	0.61
	1.441	1.441	0.014	0.30	2.17	1.23	0.94	0.39
H15···H18A^{e}	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.160	1.584	0.010	0.33	2.37	1.26	1.10	0.40
Sum								**2.00**
								1.40

Symmetry codes are as follows: $a = 1 - x, 1 - y, -z$, $b = 1 - x, 1 - y, 1 - z$, $c = 1 - x, 2 - y, -z$, $d = x-1, y, z - 1$, $e = 2 - x, 1 - y, 1 - z$, $f = x - 1, y, z$, $g = 2 - x, 2 - y, 2 - z$, $h = 2 - x, 1 - y, -z$, $i = x, 1+y, z$. The properties are presented in the following units: distances d_1, d_2 [Å]; electron density ρ_b [e Å^{-3}]; Laplacian of electron density $\nabla^2\rho_b$ [e A^{-5}]; kinetic, potential and total energy densities G_b, V_b, H_b [H Bohr^{-3}]; Interaction energy (Using electron potential energy at BCP and Espinosa equation) E_b kcal/mol; n.f – not found.

Table S4. Selected geometric parameters and theoretical quantum-topological properties at the bond critical points of non-covalent interactions for MeAnt/F_2TCNQ single crystal (symmetry independent part). The first line is for theoretical calculations; the second line shows data for multipole modeling.

Intramolecular non-covalent interaction								
	Geometry		(3, –1) in ρ					
Interaction atoms	d_1	d_2	ρ_b	$\nabla^2\rho_b$	$G_b\ 10^3$	$-V_b\ 10^3$	$H_b\ 10^3$	E_b
C5A···F1A	1.322	1.294	0.08898	1.571	12.99	9.66	3.32	3.03
	1.319	1.280	0.0961	1.6502	13.62	10.11	3.50	3.17
C5B···F1B	1.324	1.298	0.08749	1.55	12.80	9.48	3.32	2.97
	1.318	1.295	0.0949	1.6262	13.41	9.96	3.50	3.12
H12···H25C	1.050	1.025	0.116	1.36	12.71	11.27	1.44	3.54
	1.105	1.102	0.1046	1.2632	11.38	9.67	1.70	3.03
C25···H22	1.499	1.132	0.09313	1.27	11.05	8.95	2.11	2.81
	1.495	1.128	0.0879	1.1983	10.86	9.29	1.60	2.91
Intermolecular non-covalent interaction								
***F_2TCNQ-F_2TCNQ* interactions**								
N6A···H3A	1.411	0.885	0.06656	1.41	11.05	7.48	3.57	2.35
	1.410	0.884	0.0913	1.2319	10.28	7.78	2.50	2.44
N6B···H3B	1.402	0.873	0.06936	1.48	11.61	7.90	3.71	2.48
	1.401	0.873	0.0949	1.2781	10.72	8.18	2.50	2.57
C6A···N5A	1.575	1.559	0.04483	0.63	5.05	3.54	1.52	1.11
C4A···N5A	1.551	1.550	0.0508	0.6193	5.51	4.59	0.90	1.44
C6B···N5B	1.604	1.555	0.04591	0.65	5.16	3.63	1.53	1.14
C4B···N5B	1.551	1.532	0.0521	0.6319	5.65	4.74	0.90	1.49
C5A…C5A	1.614	1.614	0.04443	0.54	4.40	3.20	1.21	1.00

	1.612	1.612	0.0460	0.5408	4.60	3.60	1.00	1.13
C5B…C5B	1.607	1.607	0.04529	0.55	4.50	3.28	1.22	1.03
	1.606	1.606	0.0468	0.5505	4.69	3.67	1.00	1.15
Sum								**9.11**
								10.22
***F₂TCNQ-MeAnt* side-on interactions**								
F1A···H20	1.411	0.979	0.02717	0.96	6.95	3.91	3.04	1.23
	1.412	0.981	0.0467	0.8369	6.20	3.71	2.50	1.17
F1B···H14	1.387	0.973	0.03344	1.04	7.60	4.42	3.18	1.39
	1.386	0.973	0.0513	0.9386	7.01	4.28	2.70	1.34
N5B···H12	1.570	1.139	0.03258	0.6	4.52	2.85	1.67	0.90
	1.567	1.135	0.0452	0.5505	4.53	3.34	1.20	1.05
N5A···H22	1.571	1.174	0.03428	0.58	4.43	2.86	1.57	0.90
	1.570	1.168	0.0436	0.5294	4.39	3.30	1.10	1.03
N5A···H21	1.686	1.249	0.02948	0.45	3.44	2.23	1.22	0.70
	1.685	1.249	0.0359	0.4258	3.53	2.64	0.90	0.83
N6B···H13	1.740	1.114	0.01649	0.48	3.42	1.90	1.52	0.60
	1.737	1.113	0.0355	0.4089	3.22	2.19	1.00	0.69
N6A···H21	1.783	1.144	0.01763	0.44	3.16	1.79	1.37	0.56
	1.781	1.139	0.0341	0.3784	3.01	2.10	0.90	0.66
N5B···H15	1.684	1.298	0.02034	0.36	2.64	1.59	1.05	0.50
	1.682	1.288	0.0290	0.3376	2.76	2.02	0.70	0.63
N5A···H19	1.697	1.267	0.01791	0.35	2.54	1.49	1.05	0.47
	1.695	1.260	0.0287	0.3309	2.68	1.92	0.80	0.60
N6A···H25A	1.755	1.235	0.01236	0.3	2.13	1.19	0.95	0.37
	1.747	1.231	0.0270	0.2850	2.32	1.69	0.60	0.53
N6B···H13	1.114	1.740	0.01649	0.48	3.42	1.90	1.52	0.60

	1.115	1.689	0.0226	0.2851	2.29	1.63	0.70	0.51
N6B···H25B	1.406	1.740	0.01671	0.27	1.97	1.18	0.79	0.37
	1.402	1.739	0.0230	0.2599	2.13	1.56	0.60	0.49
N5B···H14	1.409	1.740	0.01763	0.28	2.09	1.26	0.83	0.40
	1.402	1.738	0.0209	0.2874	2.26	1.55	0.70	0.49
N6B···N6B	1.752	1.752	0.01755	0.27	2.02	1.22	0.80	0.38
	1.749	1.749	0.0216	0.3297	2.43	1.44	1.00	0.45
N6A···N6A	1.762	1.762	0.01679	0.26	1.93	1.16	0.77	0.37
	1.759	1.759	0.0208	0.3186	2.34	1.37	1.00	0.43
Sum								**9.74**
								10.90
***F_2TCNQ-MeAnt* stack interactions**								
C4A···C17	1.649	1.711	0.04617	0.52	4.33	3.23	1.10	1.01
	1.639	1.689	0.0529	0.5069	4.86	4.47	0.40	1.40
C4B···C17	1.649	1.711	0.04617	0.52	4.33	3.23	1.10	1.01
	1.638	1.674	0.0532	0.5093	4.89	4.49	0.40	1.41
C2A···C18	1.757	1.769	0.04012	0.45	3.66	2.67	0.99	0.84
C3A···C18	1.732	1.734	0.0433	0.4540	4.20	3.70	0.50	1.16
C3B···C16	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.731	1.735	0.0433	0.4549	4.22	3.71	0.50	1.16
C1A···C21	1.667	1.802	0.04211	0.46	3.77	2.80	0.97	0.88
	1.610	1.774	0.0461	0.4433	4.10	3.61	0.50	1.13
C1B···C12	1.675	1.767	0.04182	0.45	3.73	2.77	0.97	0.87
	1.668	1.731	0.0459	0.4386	4.06	3.57	0.50	1.12
C1B···C24	1.762	1.785	0.03805	0.42	3.41	2.47	0.93	0.78
	1.755	1.766	0.0413	0.4172	3.82	3.31	0.50	1.04
N5B···C19	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f

	1.701	1.711	0.0343	0.4277	3.61	2.78	0.80	0.87
N5A···C13	1.698	1.847	0.03053	0.38	2.96	2.02	0.95	0.63
	1.690	1.802	0.0320	0.4205	3.50	2.63	0.90	0.83
F1B···H25B	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.903	1.203	0.0265	0.3995	3.21	2.28	0.90	0.71
Sum								**6.02**
								9.67
***F_2TCNQ-Methyl* interactions within stack**								
C1A···H25A	1.795	1.322	0.03164	0.39	3.10	2.12	0.99	0.66
C3A···H25A	1.775	1.302	0.0362	0.3630	3.24	2.72	0.50	0.85
F1B···H25B	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.903	1.203	0.0265	0.3995	3.21	2.28	0.90	0.71
Sum								**0.66**
								1.56
***MeAnt-MeAnt* interactions**								
C22-H14	1.835	1.420	0.02811	0.34	2.66	1.79	0.86	0.56
	1.835	1.388	0.0287	0.3233	2.78	-2.20	0.60	0.69
C15-H21	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.877	1.307	0.0251	0.2923	2.47	-1.91	0.60	0.60
H12-H20	1.415	1.477	0.02235	0.29	2.23	1.43	0.80	0.45
	1.415	1.497	0.0244	0.2765	2.34	-1.81	0.50	0.57
H13-H19	1.484	1.489	0.01998	0.26	1.96	1.24	0.72	0.39
	1.474	1.477	0.0214	0.2536	2.11	-1.58	0.50	0.50
C25-C17	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.809	1.820	0.0158	0.1863	1.52	-1.11	0.40	0.35
H15-H25C	n.f	n.f	n.f	n.f	n.f	n.f	n.f	n.f
	1.405	1.408	0.0086	0.0955	0.76	-0.52	0.20	0.16

Sum								**1.40**
								2.87

The properties are presented in the following units: distances d_1, d_2 [Å]; electron density ρ_b [e Å^{-3}]; Laplacian of electron density $\nabla^2\rho_b$ [e A^{-5}]; kinetic, potential and total energy densities G_b, V_b, H_b [H Bohr^{-3}]; Interaction energy (Using electron potential energy at BCP and Espinosa equation) E_b kcal/mol; n.f – not found.

Table S5. Selected properties integrated over the topological atoms for the Me_2Ant/F_2TCNQ cocrystals.

Atom	Net charge (q_{Pval})		Bader charge		Volume	
	LT	**HT**	**LT**	**HT**	**LT**	**HT**
F1	0.111(3)	0.112(3)	0.548	0.552	16.533	17.886
N5	0.112(4)	0.102(4)	0.832	0.856	21.962	24.082
N6	0.116(4)	0.105(4)	0.828	0.845	22.153	24.660
C1	0.001(6)	-0.008(6)	-0.403	-0.417	8.007	8.422
C2	0.011(6)	0.023(7)	-0.049	-0.042	8.908	8.991
C3	-0.011(7)	-0.130(7)	-0.025	-0.021	10.692	10.606
C4	-0.091(6)	-0.087(6)	-0.147	-0.151	9.753	9.450
C5	0.073(5)	0.077(5)	-0.649	-0.671	10.643	11.022
C6	0.070(5)	0.082(5)	-0.647	-0.662	11.005	12.147
H3	-0.086(6)	-0.088(3)	-0.150	-0.150	6.297	6.141
C11	0.162(5)	0.165(5)	0.062	0.051	9.181	9.262
C12	-0.035(5)	-0.029(5)	0.051	0.051	12.462	12.050
C13	-0.028(5)	-0.031(5)	0.030	0.022	12.765	11.859
C14	-0.031(5)	-0.025(5)	0.033	0.026	11.340	11.654
C15	-0.029(5)	-0.028(5)	0.061	0.051	11.103	11.159
C16	0.167(5)	0.167(5)	0.065	0.060	8.858	9.104
C17	-0.277(6)	-0.300(6)	-0.050	-0.051	9.380	9.464
C18	-0.115(8)	0.120(8)	-0.026	-0.036	9.356	9.361
H12	-0.037(4)	-0.037(3)	-0.083	-0.070	6.893	6.721
H13	-0.030(4)	-0.034(3)	-0.079	-0.077	8.379	8.291
H14	-0.027(4)	-0.024(3)	-0.064	-0.059	6.591	8.419
H15	-0.026(4)	-0.025(3)	-0.074	-0.055	7.339	7.345
H18A	0.002(6)	0.065(5)	-0.038	-0.004	7.649	8.942

Atom	Net charge (q_{Pval})		Bader charge		Volume	
	LT	**HT**	**LT**	**HT**	**LT**	**HT**
H18B	0.057(6)	-0.007(6)	-0.001	-0.034	6.804	7.034
H18C	0.043(6)	0.073(5)	-0.014	0.007	7.474	7.311
Acceptor	ca. 0.42	ca. 0.38	0.27	0.27		
Donor	ca. -0.42	ca. -0.38	-0.25	-0.24		
All	ca. 0.00	ca. 0.00	-0.02	-0.02		

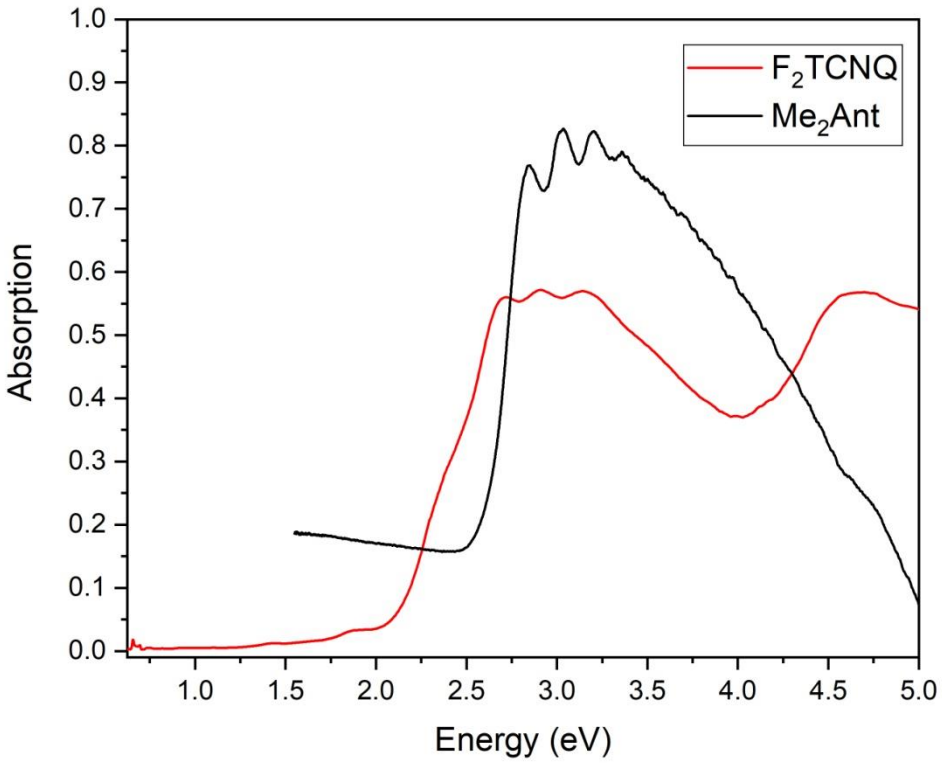


Figure S3. UV/Vis spectra of F_2TCNQ and Me_2Ant.