

4-cyanobenzene-ethylenedithio-TTF electron donor and its (1:1)  
triiodide radical cation salt; isomer effects in C–N···H–C  
interactions

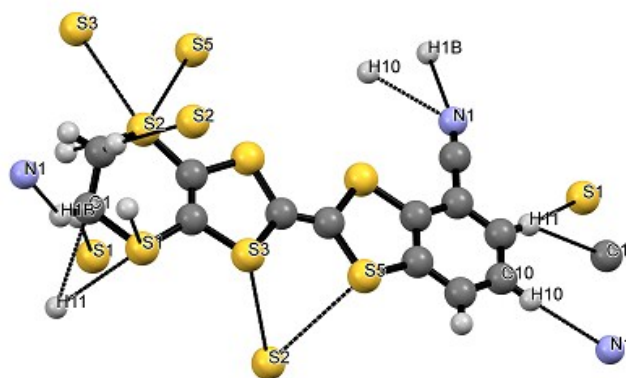
Gonçalo Lopes<sup>a</sup>, Vasco da Gama<sup>a</sup>, Dulce Belo<sup>a</sup>, Dulce Simão<sup>b</sup>, Isabel  
Cordeiro Santos<sup>a</sup>, Manuel Almeida<sup>a,\*</sup> and Sandra Rabaça<sup>a,\*</sup>

<sup>a</sup> *C<sup>2</sup>TN, Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, E.N. 10 ao km 139,7, 2695-066 Bobadela LRS, Portugal*

<sup>b</sup> *Centro de Química Estrutural, Instituto Superior Técnico, Universidade de Lisboa, P-1049-001 Lisboa, Portugal*

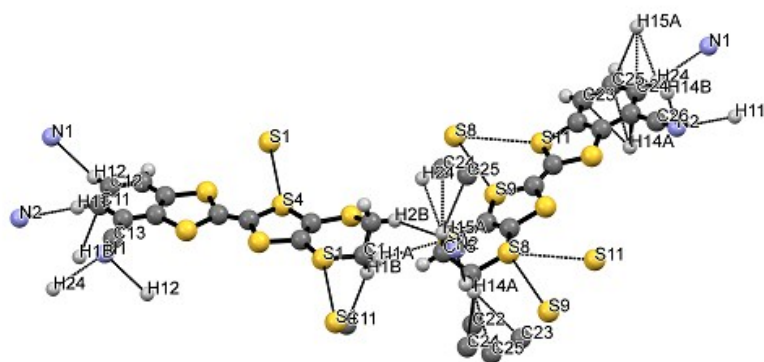
**Keywords:**

Dissymmetric tetrathiafulvalene donor; Cyanobenzene; C–N···H–C interactions; Synthesis; X-ray crystal structure; Cyclic voltammetry



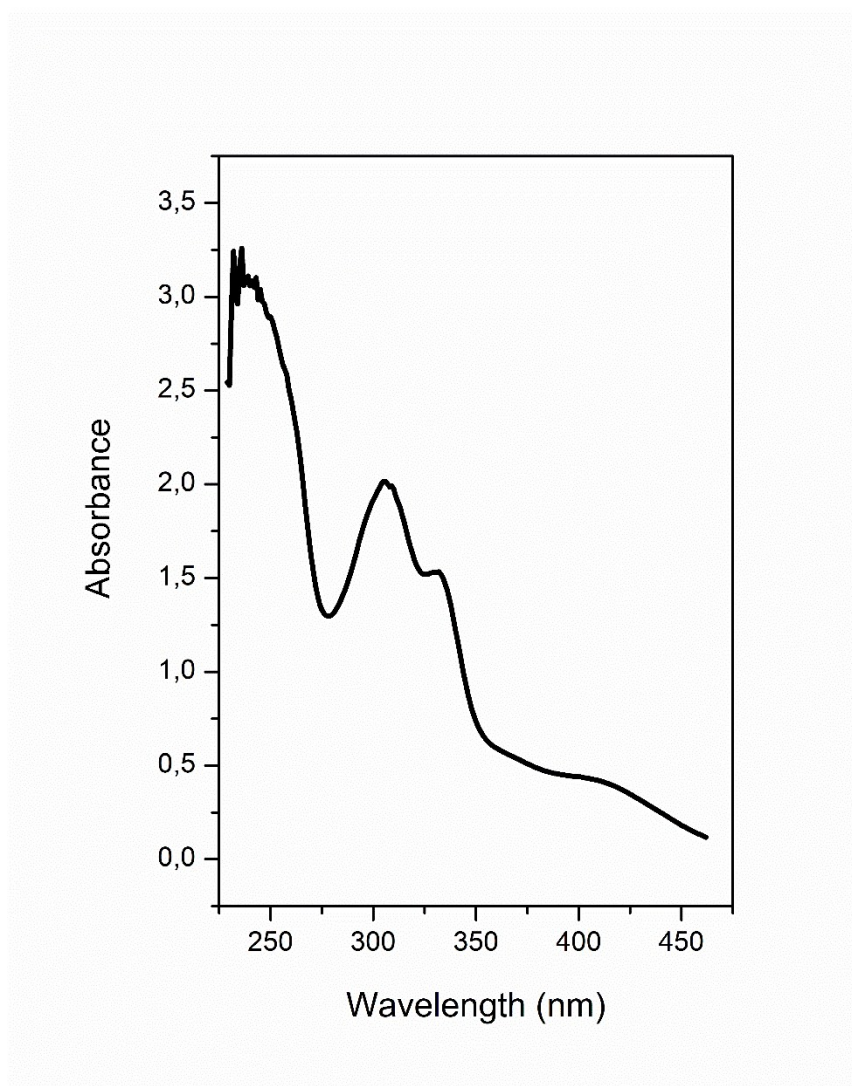
**Table S1.** Short contact list of compound  $\alpha$ -4-CNB-EDT-TTF ( $\alpha$ -1)

Short Contacts	Symmetry operation	Length(Å)	Length- $\Sigma$ VdW(Å)
S1...H1A	$x, -1+y, z$	2.725	-0.275
H2A...S2	$x, -1+y, z$	2.710	-0.290
S1...H11	$1-x, 1-y, -1/2+z$	2.972	-0.028
C1...H11	$1-x, 1-y, -1/2+z$	2.835	-0.065
S2...S3	$-1/2+x, 1-y, z$	3.552	-0.048
S2...S5	$-1/2+x, 1-y, z$	3.578	-0.022
C13-N1...H10-C10	$-1/2+x, 1-y, z$	2.619	-0.131
C1-H1B...N1-C13	$1/2-x, -1+y, -1/2+z$	2.632	-0.118

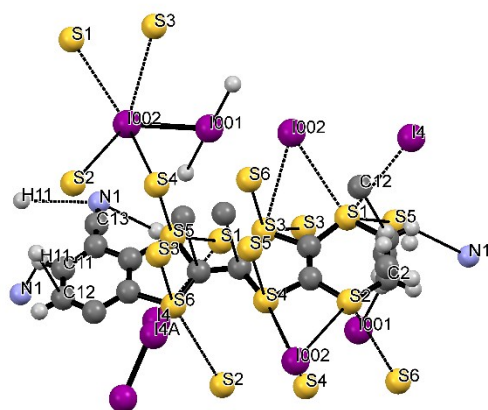


**Table S2.** Short contact list of compound  $\beta$ -4-CNB-EDT-TTF ( $\beta$ -1)

Short Contacts	Symmetry operation	Length(Å)	Length- $\Sigma$ VdW(Å)
S4...S1	-1+x,y,z	3.522	-0.078
H1B...C11	-x,2-y,-z	2.700	-0.200
H1A...S7	x,y,z	2.939	-0.061
H2B...S7	x,y,z	2.835	-0.165
S9...S8	-1+x,y,z	3.553	-0.047
S11...S8	-1+x,y,z	3.587	-0.013
H15A...C24	1-x,-y,1-z	2.663	-0.237
H15A...H24	1-x,-y,1-z	2.391	-0.009
H15A...C25	1-x,-y,1-z	2.740	-0.160
H14A...C22	1-x,1-y,1-z	2.827	-0.073
H14A...C23	1-x,1-y,1-z	2.810	-0.090
H14A...C24	1-x,1-y,1-z	2.702	-0.198
H14A...C25	1-x,1-y,1-z	2.712	-0.188
C26-N2...H14B	2-x,-y,1-z	2.745	-0.005
C25-H25...N2-C26	-1+x,y,z	2.831	0.081
C12-H12...N1-C13	-1+x,y,z	2.637	-0.113
C11-H11...N2-C26	-1+x,1+y,-1+z	2.489	-0.261
C13-N1...H24-C24	x,1+y,-1+z	2.586	-0.164



**Figure S1.** UV-vis absorption spectra in DCM of 4-CNB-EDT-TTF donor.



**Table S3.** Short contact list of compound [4-CNB-EDT-TTF]I<sub>3</sub> (2)

Short Contacts	Symmetry operation	Length(Å)	Length- $\Sigma$ VdW(Å)
S4...S5	1/2-x,1.5-y,1-z	3.362	-0.238
S3...S6	1/2-x,1.5-y,1-z	3.330	-0.270
C6...C5	1/2-x,1.5-y,1-z	3.400	-0.000
C12...H1B-C1	1/2-x,1.5-y,1-z	2.694	-0.206
S2...S6	-x,1-y,1-z	3.560	-0.040
S4...S4	-x,1-y,1-z	3.557	-0.043
S5...S1	1-x,1-y,1-z	3.541	-0.059
S3...S3	1-x,1-y,1-z	3.576	-0.024
C13-N1...H1A-C1	1-x,1-y,1-z	2.667	-0.083
C11-H11...N1-C13	1-x,y,1/2-z	2.749	-0.001
C12-H12...N1A-C12	-x, y, 1/2-z	2.842	+0.092
S1...I4	1-x,1-y,1-z	3.592	-0.188
I001...H2B	1/2+x,1/2+y,z	3.132	-0.048
I002...S2	1/2-x,1.5-y,1-z	3.673	-0.107
I002...S4	1/2-x,1.5-y,1-z	3.594	-0.186
I002...S3	1.5-x,1.5-y,1-z	3.640	-0.140
I002...S1	1.5-x,1.5-y,1-z	3.642	-0.138