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Evaluating the high-pressure structural response and crystal lattice interactions of the magnetically-bistable organic radical TTTA

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Table S1: Summary of the crystallographic information obtained from the high-pressure diffraction experiments on 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure	(ambient)	(0.33 GPa)	(0.94 GPa)	(1.30 GPa)
Crystal data				
Chemical formula	$C_2N_3S_3$	$C_2N_3S_3$	$C_2N_3S_3$	$C_2N_3S_3$
M_r	162.24	162.24	162.24	162.24
Crystal system, space group	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$
Temperature (K)	295	293	293	293
a, b, c (Å)	9.43880 (2), 3.710000 (14), 15.05490 (3)	9.409 (11), 3.6033 (3), 14.945 (3)	9.348 (13), 3.5161 (4), 14.834 (3)	9.327 (10), 3.4620 (3), 14.734 (3)
β (°)	104.552 (3)	104.55 (6)	104.56 (7)	104.37 (5)
V (Å^3)	510.28 (1)	490.5 (6)	471.9 (7)	460.9 (5)
Z	4	4	4	4
Radiation type	Mo $K\alpha$	Synchrotron, λ = 0.48590 Å	Synchrotron, λ = 0.48590 Å	Synchrotron, λ = 0.48590 Å
μ (mm^{-1})	1.32	0.46	0.48	0.49
Crystal size (mm)	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10
Data collection				
Diffractometer	Bruker Kappa Apex2	Synchrotron	Synchrotron	Synchrotron

Absorption correction	Multi-scan *SADABS* (Siemens, 1996)	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski & Minor, 1997)	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski & Minor, 1997)	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski & Minor, 1997)
T_{min}, T_{max}	0.75, 0.88	0.40, 0.95	0.01, 0.95	0.83, 0.95
No. of measured, independent and observed [$I > 2.0\sigma(I)$] reflections	4849, 1061, 1031	2336, 538, 455	2133, 502, 422	2294, 454, 373
R_{int}	0.022	0.038	0.028	0.023
$(\sin\theta/\lambda)_{max}$ (Å^{-1})	0.628	0.797	0.787	0.790
Refinement				
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.026, 0.067, 0.99	0.068, 0.157, 0.99	0.070, 0.202, 1.10	0.042, 0.094, 1.01
No. of reflections	1061	524	502	445
No. of parameters	73	73	73	73
No. of restraints	80	104	104	104
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.55, −0.43	0.31, −0.42	0.60, −0.62	0.25, −0.26

Pressure	(1.76 GPa)	(2.11 GPa)	(2.54 GPa)	(2.91 GPa)
Crystal data				
Chemical formula	$C_2N_3S_3$	$C_2N_3S_3$	$C_2N_3S_3$	$C_2N_3S_3$
M_r	162.24	162.24	162.24	162.24
Crystal system, space group	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$
Temperature (K)	293	293	293	293
a, b, c (Å)	9.323 (11), 3.4191 (4), 14.678 (3)	9.28 (1), 3.3957 (3), 14.640 (3)	9.240 (12), 3.3710 (4), 14.596 (3)	9.191 (12), 3.3508 (4), 14.562 (3)
β (°)	104.31 (6)	104.34 (5)	104.46 (6)	104.44 (6)
V (Å^3)	453.3 (5)	446.9 (5)	440.3 (6)	434.3 (6)
Z	4	4	4	4
Radiation type	Synchrotron, λ = 0.48590 Å	Synchrotron, λ = 0.48590 Å	Synchrotron, λ = 0.48590 Å	Synchrotron, λ = 0.48590 Å
μ (mm^{-1})	0.50	0.51	0.51	0.52
Crystal size (mm)	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10
Data collection				
Diffractometer	Synchrotron	Synchrotron	Synchrotron	Synchrotron
Absorption correction	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski

	& Minor, 1997)	& Minor, 1997)	& Minor, 1997)	& Minor, 1997)
T_{min}, T_{max}	0.84, 0.95	0.76, 0.95	0.63, 0.95	0.80, 0.95
No. of measured, independent and observed [$I > 2.0\sigma(I)$] reflections	1699, 343, 272	1902, 374, 303	2284, 463, 339	2098, 411, 305
R_{int}	0.018	0.019	0.027	0.040
$(\sin \theta/\lambda)_{max}$ (Å^{-1})	0.661	0.665	0.796	0.715
Refinement				
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.036, 0.057, 1.00	0.040, 0.110, 1.03	0.049, 0.094, 1.01	0.071, 0.177, 0.99
No. of reflections	338	374	458	403
No. of parameters	73	73	73	73
No. of restraints	104	104	104	104
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.25, −0.21	0.31, −0.38	0.43, −0.38	0.53, −0.48

Pressure	(3.94 GPa)	(4.56 GPa)
Crystal data		

Chemical formula	$C_2N_3S_3$	$C_2N_3S_3$
M_r	162.24	162.24
Crystal system, space group	Monoclinic, $P2_1/c$	Monoclinic, $P2_1/c$
Temperature (K)	293	293
a, b, c (Å)	9.084 (16), 3.3013 (5), 14.495 (4)	9.07 (2), 3.2680 (7), 14.418 (6)
β (°)	104.62 (8)	104.63 (11)
V (Å^3)	420.6 (8)	413.3 (10)
Z	4	4
Radiation type	Synchrotron, λ = 0.48590 Å	Synchrotron, λ = 0.48590 Å
μ (mm^{-1})	0.54	0.55
Crystal size (mm)	0.12 × 0.10 × 0.10	0.12 × 0.10 × 0.10
Data collection		
Diffractometer	Synchrotron	Synchrotron
Absorption correction	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski & Minor, 1997)	Multi-scan *DENZO*/*SCALEPACK* (Otwinowski & Minor, 1997)
T_{min}, T_{max}	0.36, 0.95	0.78, 0.95
No. of measured, independent and observed [$I > 2.0\sigma(I)$] reflections	2270, 454, 343	2149, 446, 337

R_{int}	0.031	0.032
$(\sin \theta/\lambda)_{max}$ (Å^{-1})	0.796	0.801
Refinement		
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.063, 0.131, 0.90	0.059, 0.173, 1.00
No. of reflections	427	443
No. of parameters	73	73
No. of restraints	104	104
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.37, −0.40	0.58, −0.52

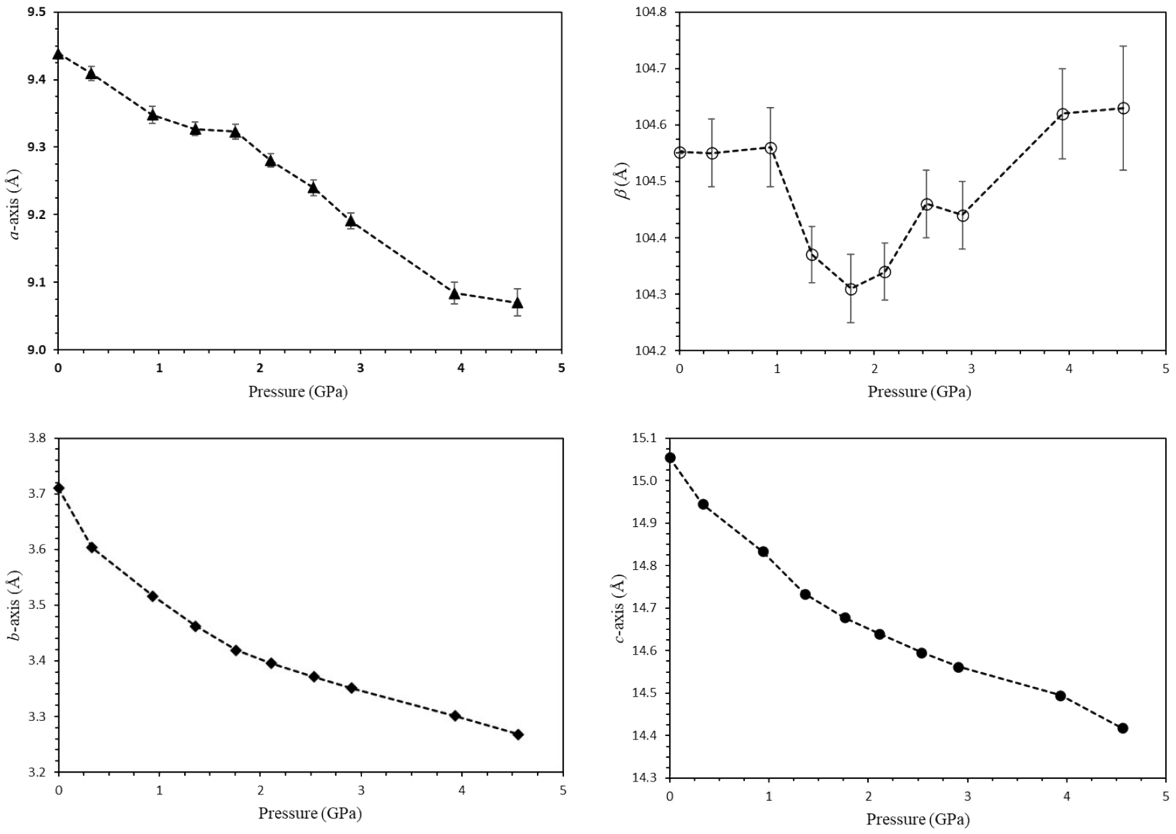

Figure S1: Unit cell parameter compression curves obtained from the high-pressure diffraction experiments on 1,3,5-triathia-2,4,6-triazapentalenyl.

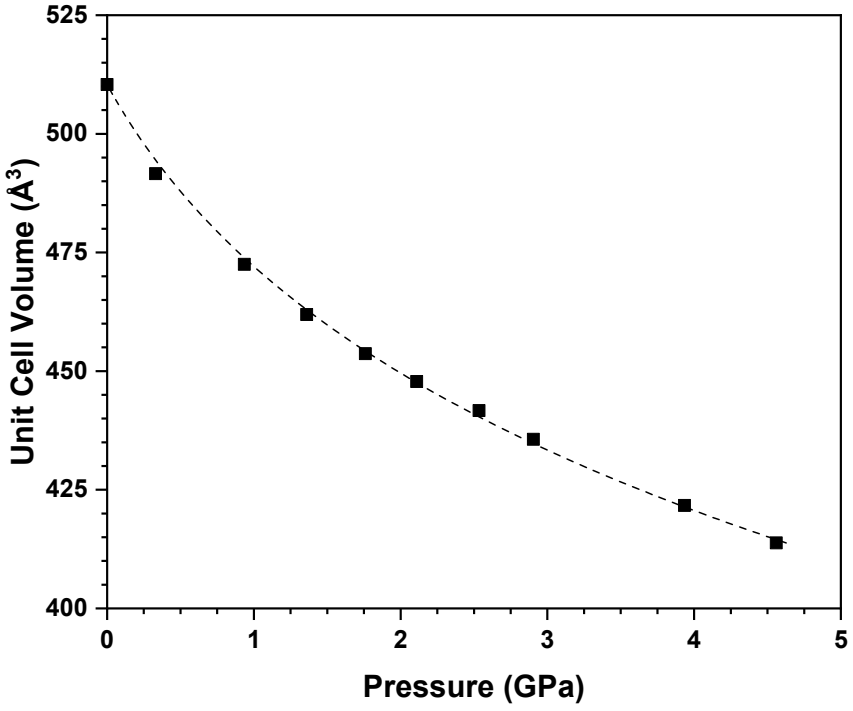

Figure S2: Volume-pressure data from the high-pressure diffraction experiments on 1,3,5-triathia-2,4,6-triazapentalenyl, line-fitted to the 3rd-order Birch-Murnaghan Equation of State.

Table S2: Component and total lattice energies for the ambient pressure $P2_1/c$ structure of TTTA with varying density step size of electron pixels and, thus, different number of superpixels. In all cases, superpixels were devised from the 3x3x3 combination of pixels. The emboldened line shows the density step size used to generate the results in the main text and the rest of the supplementary information; this value, combined with the condensation level of n=3, is in line with, or even exceeding, literature precedents.[1-3]

Pixel density step size (Å)	**# of superpixels per molecule**	**Lattice Energies (kJ mol^{-1})**				
		Coulombic	**Polarisation**	**Dispersion**	**Repulsion**	**Total**
0.25	1755	-104.1	-50.3	-138.6	160.7	-132.3
0.20	3168	-85.3	-42.5	-134.2	152.4	-109.7
0.16	6440	-71.1	-36.9	-131.2	147.4	-91.8
0.14	9152	-67.9	-33.4	-131.4	145.9	-86.7
0.12	14508	-63.5	-33.9	-131	144.6	-83.9
0.11	18480	-61.9	-31.3	-130.9	144.0	-80.1
0.10	25234	-61.1	-32.3	-130.5	143.5	-80.4
0.09	34440	-59.1	-32.2	-130.5	142.7	-79.1
0.08	**48438**	**-56.8**	**-30.8**	**-130.8**	**142.9**	**-75.5**
0.07	72540	-54.5	-30.0	-130.4	142.0	-72.9
0.06	114192	-51.2	-28.7	-130.4	142.1	-68.3

Table S3: Component and total lattice energies calculated for the $P2_1/c$ polymorph of TTTA across the pressure series up to the structure obtained at 4.56 GPa.

Pressure (GPa)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Energy (kJ mol^{-1})
Ambient	-56.8	-30.8	-130.8	142.9	-75.5
0.33	-67.7	-39.0	-144.7	178.1	-73.4
0.94	-83.2	-51.1	-160.8	225.2	-69.9
1.36	-93.3	-57.4	-170.3	254.9	-66.1
1.76	-102.9	-63.6	-178.5	283.6	-61.5
2.11	-108.6	-66.2	-184	302.1	-56.6
2.54	-117.7	-71.5	-190.9	327.8	-52.3
2.91	-128.7	-76.6	-198.3	357.1	-46.6
3.94	-154.4	-93.5	-215.2	425.1	-38.0
4.56	-169.5	-104	-225.1	465.6	-32.9

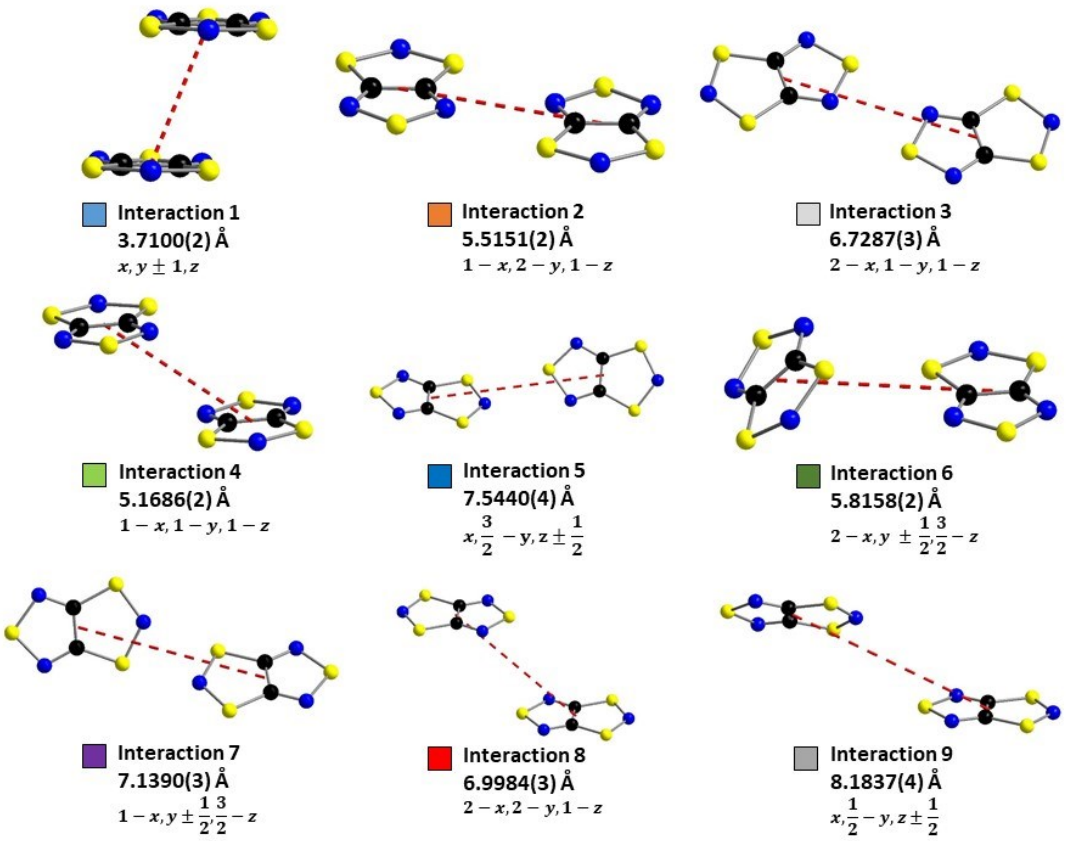

Figure S3: Identified intermolecular interactions in the ambient pressure $P2_1/c$ phase of TTTA with total interaction energy of at least -2.5 kJ mol^{-1}, with colour identifiers to match those used in the energy framework diagram in Figures 3 and S4. Also provided for each interaction is the intermolecular centroid···centroid separation and symmetry operation relating the two molecules in each interaction dimer.

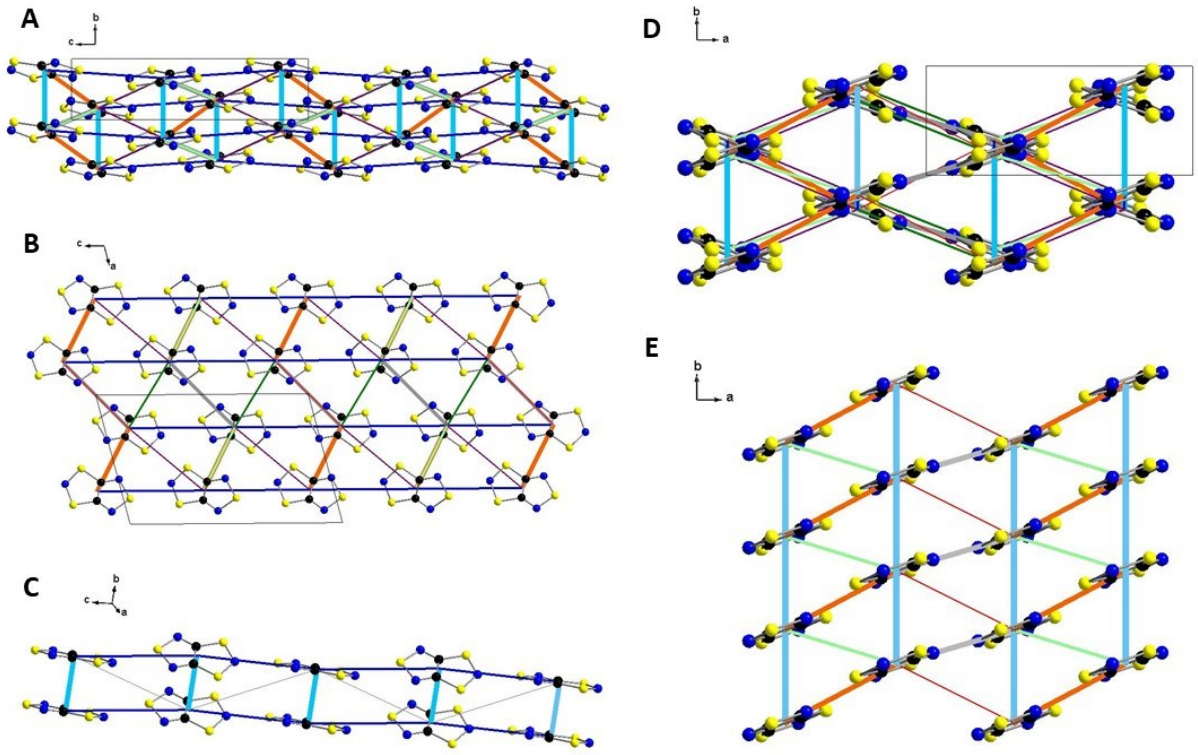

Figure S4: Various views of the energy framework of the ambient pressure HT phase structure of TTTA, consisting of all nine strongest interactions, as calculated by the PIXEL method. Interaction rod thickness is scaled according to the total interaction energy, and are coloured to match the interactions shown in Figure 3. A, B and D view the crystal lattice along the a, b, and c axes respectively. C and E show extended segments of the crystal lattice, to provide a clearer view of the combination of inter-column interactions mostly aligned with the bc and ab planes respectively.

Table S4: Interaction energy terms, calculated by PIXEL, for interaction 1 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	3.710	-10.8	-8.5	-46.3	47.4	-18.2
0.33	3.605	-15.6	-12.2	-52.6	63.7	-16.6
0.94	3.518	-20.1	-16.9	-58.6	81.7	-13.9
1.36	3.466	-24.9	-19.9	-62.6	95.0	-12.4
1.76	3.424	-28.7	-22.3	-65.9	107.7	-9.2
2.11	3.398	-31.1	-23.5	-68.6	116.7	-6.5
2.54	3.374	-33.5	-24.5	-70.9	125.7	-3.2
2.91	3.354	-37.1	-25.7	-73	135	-0.8
3.94	3.302	-46.3	-32.6	-79.0	159.5	1.7
4.56	3.267	-53.4	-38.5	-83.5	179.2	3.8

Table S5: Interaction energy terms, calculated by PIXEL, for interaction 2 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid	Coulombic Energy	Polarisation Energy	Dispersion Energy	Repulsion Energy	Total Interaction

	Separation (Å)	(kJ mol^{-1})	(kJ mol^{-1})	(kJ mol^{-1})	(kJ mol^{-1})	Energy (kJ mol^{-1})
Ambient	5.515	-25.4	-8.9	-24.6	41.8	-17.1
0.33	5.467	-28.1	-10.8	-26.2	48.7	-16.4
0.94	5.437	-29.9	-11.8	-27.3	53.7	-15.3
1.36	5.403	-32.2	-12.6	-28.3	59.3	-13.8
1.76	5.392	-31.9	-12.2	-28.7	59.7	-13.2
2.11	5.391	-32.6	-12.6	-28.8	62.3	-11.7
2.54	5.365	-35.0	-13.5	-29.9	67.1	-11.3
2.91	5.347	-36.8	-14.2	-30.9	71.3	-10.6
3.94	5.272	-45.2	-19.5	-34.4	90.8	-8.3
4.56	5.236	-50.3	-22.5	-36.3	102.6	-6.5

Table S6: Interaction energy terms, calculated by PIXEL, for interaction 3 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	5.169	-4.6	-2.1	-17.5	13.2	-10.9
0.33	5.14	-6.1	-2.8	-19.7	18.2	-10.4
0.94	5.118	-8.0	-3.8	-21.7	23.8	-9.7
1.36	5.09	-9.7	-4.2	-23.2	29	-8.1
1.76	5.08	-11.2	-4.8	-24.6	32.8	-7.8
2.11	5.072	-12.0	-5.7	-25.2	35.9	-7.0
2.54	5.068	-12.0	-5.8	-25.3	35.8	-7.3
2.91	5.045	-12.9	-6.2	-26.2	38.3	-6.9
3.94	4.981	-15.8	-7.9	-28.9	47.0	-5.6
4.56	4.951	-17.0	-8.6	-29.8	50.8	-4.7

Table S7: Interaction energy terms, calculated by PIXEL, for interaction 4 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	6.729	-4.6	-2.1	-17.5	13.2	-10.9
0.33	5.14	-6.1	-2.8	-19.7	18.2	-10.4
0.94	5.118	-8.0	-3.8	-21.7	23.8	-9.7
1.36	5.09	-9.7	-4.2	-23.2	29.0	-8.1
1.76	5.08	-11.2	-4.8	-24.6	32.8	-7.8
2.11	5.072	-12.0	-5.7	-25.2	35.9	-7.0
2.54	5.068	-12.0	-5.8	-25.3	35.8	-7.3
2.91	5.045	-12.9	-6.2	-26.2	38.3	-6.9
3.94	4.981	-15.8	-7.9	-28.9	47.0	-5.6
4.56	4.951	-17.0	-8.6	-29.8	50.8	-4.7

Table S8: Interaction energy terms, calculated by PIXEL, for interaction 5 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	7.544	-8.8	-4.4	-12.5	16.8	-8.9
0.33	7.484	-10.6	-5.4	-14	21.6	-8.4
0.94	7.431	-12.5	-6.9	-15.4	26.5	-8.4
1.36	7.386	-13.7	-8.2	-16.5	30.1	-8.3
1.76	7.365	-15.5	-9.5	-17.4	34.4	-8.0
2.11	7.338	-15.1	-9.4	-17.6	34.3	-7.7
2.54	7.311	-17.2	-10.5	-18.5	39	-7.2
2.91	7.297	-18.3	-11.2	-19.2	42.2	-6.5
3.94	7.261	-19.9	-11.6	-20.1	46.3	-5.2
4.56	7.223	-21.1	-12.7	-20.9	49.9	-4.8

Table S9: Interaction energy terms, calculated by PIXEL, for interaction 6 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	5.816	-6.8	-5.9	-17.1	22.5	-7.4
0.33	5.791	-8.0	-7.2	-18.4	26.5	-7.1
0.94	5.715	-10.7	-9.6	-21.0	35.0	-6.3
1.36	5.708	-11.1	-9.8	-21.4	36.1	-6.3
1.76	5.679	-12.9	-11.6	-22.7	41.5	-5.7
2.11	5.656	-13.0	-11.6	-23.1	42.1	-5.6
2.54	5.628	-15.0	-13.2	-24.4	47.6	-5.0
2.91	5.595	-16.4	-14.5	-25.5	52.0	-4.4
3.94	5.539	-20.1	-17.7	-27.8	62.5	-3.1
4.56	5.510	-20.7	-18.1	-28.5	64.4	-2.8

Table S10: Interaction energy terms, calculated by PIXEL, for interaction 7 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	7.139	-5.1	-3	-12.8	14.3	-6.5
0.33	7.092	-5.6	-3.7	-13.9	17.1	-6.1
0.94	7.039	-7.1	-4.7	-15.4	21.5	-5.8
1.36	6.999	-8.1	-5.5	-16.6	25.3	-4.9
1.76	6.976	-8.5	-5.6	-17.2	27.3	-4.0
2.11	6.971	-9.0	-5.8	-17.3	27.8	-4.3
2.54	6.946	-9.7	-6.3	-18.1	30.5	-3.6
2.91	6.915	-11.2	-7.0	-19.2	35.1	-2.4
3.94	6.850	-14.3	-8.9	-21.4	43.8	-0.8
4.56	6.826	-16.1	-9.8	-22.2	48.1	-0.1

Table S11: Interaction energy terms, calculated by PIXEL, for interaction 8 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	6.998	-0.1	-0.3	-5.2	1.4	-4.3
0.33	6.955	-0.2	-0.3	-5.6	1.7	-4.5
0.94	6.879	-0.5	-0.5	-6.6	2.5	-5.0
1.36	6.846	-0.6	-0.5	-7.1	3.1	-5.2
1.76	6.813	-0.8	-0.6	-7.4	3.4	-5.4
2.11	6.782	-1.1	-0.7	-7.9	4.1	-5.6
2.54	6.747	-1.2	-0.8	-8.4	4.6	-5.7
2.91	6.727	-1.4	-1.0	-8.8	5.5	-5.8
3.94	6.680	-1.6	-1.1	-9.5	6.4	-5.8
4.56	6.665	-1.8	-1.3	-9.9	7.3	-5.8

Table S12: Interaction energy terms, calculated by PIXEL, for interaction 9 throughout the pressure series on the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl.

Pressure (GPa)	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
Ambient	8.184	-0.4	-0.2	-2.7	0.5	-2.8
0.33	8.096	-0.5	-0.2	-3.1	0.7	-3.1
0.94	8.014	-0.6	-0.3	-3.5	0.9	-3.4
1.36	7.955	-0.6	-0.3	-3.8	1.1	-3.5
1.76	7.919	-0.7	-0.3	-4.0	1.3	-3.7
2.11	7.878	-0.8	-0.4	-4.2	1.5	-3.8
2.54	7.857	-0.8	-0.4	-4.3	1.6	-3.9
2.91	7.834	-0.8	-0.4	-4.6	1.9	-4.0
3.94	7.787	-0.9	-0.5	-4.9	2.2	-4.1
4.56	7.742	-1.1	-0.5	-5.2	2.5	-4.3

Table S13: Lattice energy terms, calculated by PIXEL, for the $P2_1/c$ phase of 1,3,5-triathia-2,4,6-triazapentalenyl throughout the entire pressure series.

Pressure (GPa)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Lattice Energy (kJ mol^{-1})
Ambient	-56.8	-30.6	-130.8	142.9	-75.4
0.33	-67.7	-39.0	-144.7	178.1	-73.4
0.94	-83.2	-51.1	-160.8	225.2	-69.9
1.36	-93.3	-57.4	-170.3	254.9	-66.1
1.76	-102.9	-63.6	-178.5	283.6	-61.5
2.11	-108.6	-66.2	-184.0	302.1	-56.6
2.54	-117.7	-71.5	-190.9	327.8	-52.3
2.91	-128.7	-76.6	-198.3	357.1	-46.6
3.94	-154.4	-93.5	-215.2	425.1	-38.0
4.56	-169.5	-104.0	-225.1	465.6	-32.9

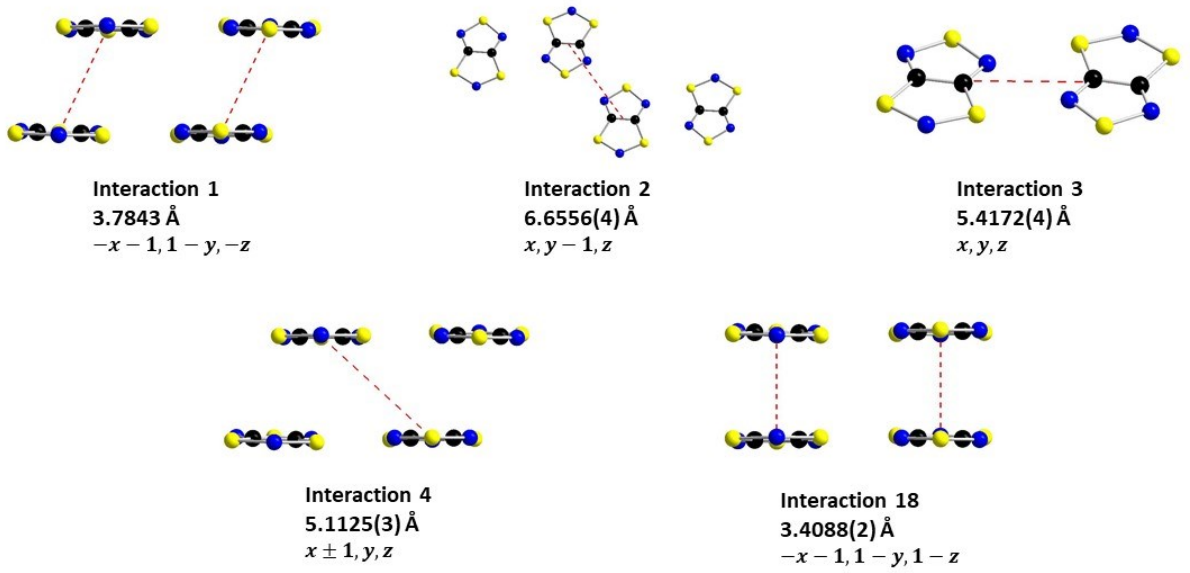

Figure S5: Selection of the intermolecular interactions identified in the ambient pressure $P\bar{1}$ phase of TTTA, with the dashed lines indicating the specific interaction between the dimers of asymmetric units. C, N and S atoms are coloured black, blue and yellow respectively. Also provided for each interaction is the intermolecular centroid···centroid separation and symmetry operation relating the two molecules in each interaction dimer.

Table S14: Component and total lattice energies calculated for the room-temperature ambient pressure structure of the $P\bar{1}$ polymorph of TTTA.[4] Rows of table indicate the interactions illustrated in Figure S5.

Interaction	Intramolecular Centroid Separation (Å)	Coulombic Energy (kJ mol^{-1})	Polarisation Energy (kJ mol^{-1})	Dispersion Energy (kJ mol^{-1})	Repulsion Energy (kJ mol^{-1})	Total Interaction Energy (kJ mol^{-1})
1	3.784	-10.1	-7.8	-44.1	43	-18.9
2	6.656	-22.6	-7.7	-18.6	31.5	-17.4
3	5.417	-34.1	-13.2	-28.9	58.9	-17.2
4	5.112	-2.9	-1.2	-14.8	8.1	-10.8
5	7.531	-6.6	-3.4	-11.3	13.7	-7.5
6	7.531	-4.8	-2.6	-10	10.1	-7.3
7	7.538	-4.5	-2.3	-9.5	9.2	-7.2
8	5.912	-7.7	-6.8	-17.8	25.5	-6.8
9	7.202	-4.3	-3.7	-13.2	14.7	-6.6
10	5.886	-9.5	-8.7	-19.7	31.5	-6.4
11	7.212	-3.9	-3.1	-12.3	13	-6.4
12	6.890	-9	-4.8	-17.8	26.6	-5.0
13	7.121	-0.2	-0.3	-5.4	1.7	-4.2
14	6.381	-0.4	-0.1	-3.9	0.2	-4.1
15	6.420	-0.3	-0.1	-3.5	0.2	-3.7
16	7.491	-0.6	-0.1	-2.2	0.1	-2.8
17	7.464	-0.6	-0.1	-2.2	0.1	-2.7
18	3.409	-26.7	-22.3	-61.7	108.5	-2.2
19	6.511	0	0	-2.2	0	-2.2
20	7.453	0.1	0	-2.4	0.2	-2.2

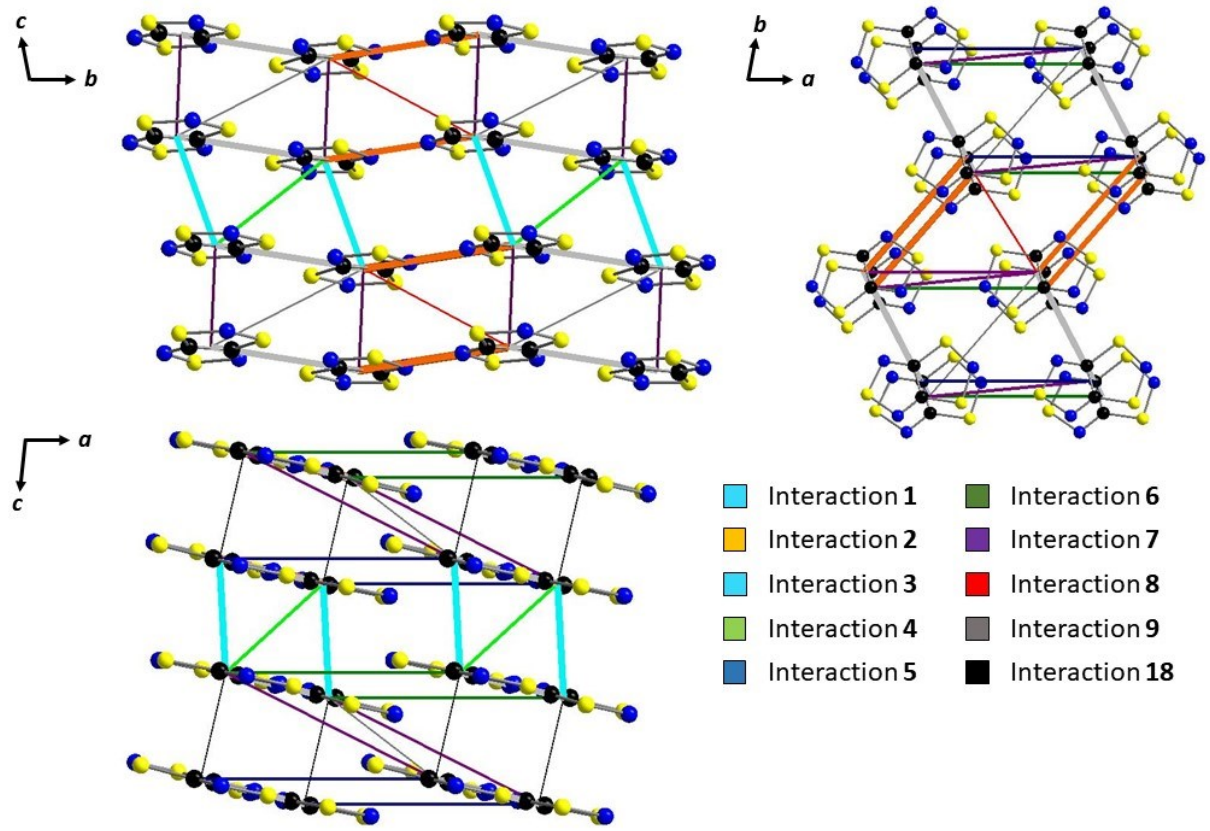

*Figure S6: Various views of the energy framework of the ambient pressure $P\bar{1}$ phase structure of TTTA, consisting of interactions **1**-**9** and **18**, as calculated by the PIXEL method. Interaction rod thickness is scaled according to the total interaction energy.*

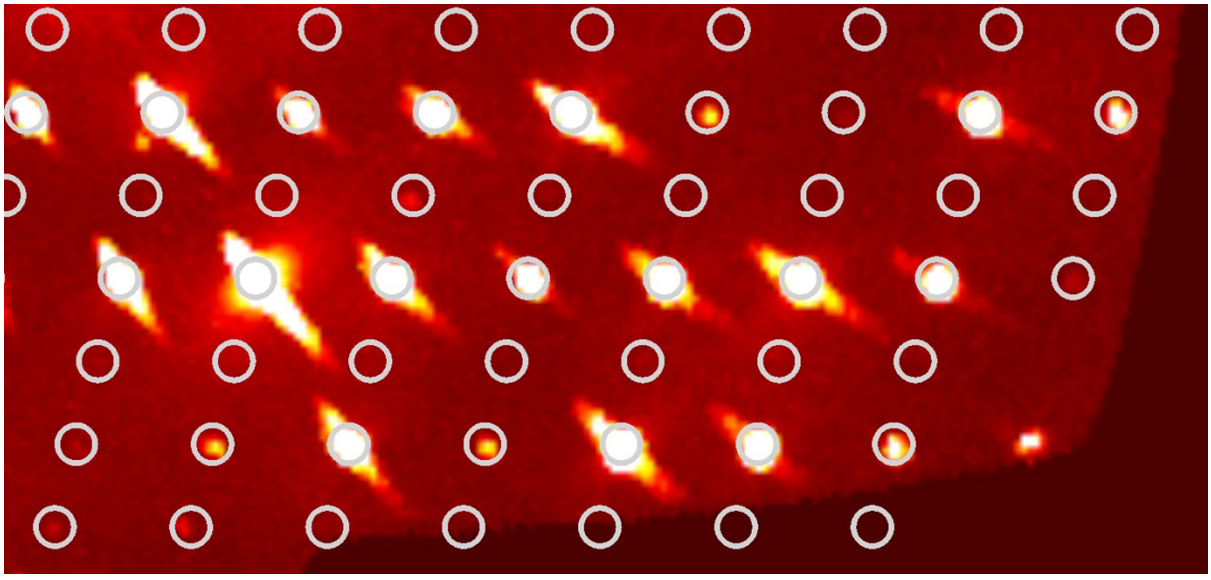

Figure S7: Calculated precession image from the ambient temperature and pressure data collection of TTTA. Circles indicate the predicted overlay of the indexed monoclinic (P21/c) polymorph. Note the streaking of reflections caused by pair exchange dynamic (PED) along the π stacking direction.

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