$C_3N_6H_7SO_3NH_2$: Non-π-Conjugated Tetrahedra Decoupling π-Conjugated Groups Achieving Large Optical Anisotropy and Wide Band Gap

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Table S1. Atomic coordinates and equivalent isotropic displacement parameters for $C_3N_6H_7SO_3NH_2$.

Atom	*x*	*y*	*z*	U(eq)
C1	0.5140(5)	0.7750(3)	0.5894(2)	0.0254(6)
C2	0.2975(5)	0.6264(3)	0.4034(2)	0.0262(6)
C3	0.6809(5)	0.8157(3)	0.4032(2)	0.0262(6)
S1	0.14325(14)	0.75661(9)	0.01911(6)	0.0284(2)
O1	0.2650(4)	0.9252(2)	0.07626(18)	0.0408(5)
O2	0.1390(4)	0.7274(3)	-0.11927(17)	0.0418(6)
O3	0.2553(4)	0.6185(2)	0.08195(17)	0.0338(5)
N1	0.5305(5)	0.8116(3)	0.7158(2)	0.0369(6)
H1A	0.415132	0.763521	0.757067	0.044
H1B	0.656614	0.883575	0.756924	0.044
N2	0.3108(4)	0.6608(3)	0.53115(19)	0.0284(6)
N3	0.7033(4)	0.8534(3)	0.53052(19)	0.0283(6)
N4	0.1077(4)	0.5165(3)	0.3377(2)	0.0344(6)
H4A	-0.008892	0.466354	0.377234	0.041
H4B	0.100156	0.494800	0.255238	0.041
N5	0.4782(4)	0.7040(3)	0.3378(2)	0.0297(6)
H5	0.461(6)	0.686(4)	0.257(3)	0.035
N6	0.8518(5)	0.8871(3)	0.3359(2)	0.0355(6)
H6A	0.980320	0.959303	0.374445	0.043
H6B	0.834995	0.861481	0.253404	0.043
N7	-0.1605(5)	0.7513(3)	0.0550(2)	0.0314(6)
H7A	-0.238(5)	0.649(3)	0.022(3)	0.038
H7B	-0.237(5)	0.835(3)	0.021(3)	0.038

U_{eq} is defined as 1/3 of the trace of the orthogonalized U_{ij} tensor.

Table S2. Bond lengths [Å] and angles [°] for $C_3N_6H_7SO_3NH_2$.

Atom–Atom	Length [Å]	Atom–Atom–Atom	Angle [°]
C1–N1	1.317(3)	N1–C1–N2	116.8(2)
C1–N2	1.352(3)	N1–C1–N3	117.1(2)
C1–N3	1.352(3)	N2–C1–N3	126.1(2)
C2–N4	1.314(3)	N4–C2–N2	120.4(2)
C2–N2	1.332(3)	N4–C2–N5	118.5(2)
C2–N5	1.361(3)	N2–C2–N5	121.1(2)
C3–N6	1.317(3)	N6–C3–N3	121.0(2)
C3–N3	1.326(3)	N6–C3–N5	117.9(2)
C3–N5	1.363(3)	N3–C3–N5	121.1(2)
S1–O2	1.4438(19)	O2–S1–O1	113.55(12)
S1–O1	1.444(2)	O2–S1–O3	111.63(13)
S1–O3	1.461(2)	O1–S1–O3	112.26(12)
S1–N7	1.648(3)	O2–S1–N7	109.62(12)
N1–H1A	0.8600	O1–S1–N7	104.29(13)
N1–H1B	0.8600	O3–S1–N7	104.81(12)
N4–H4A	0.8600	C1–N1–H1A	120.0
N4–H4B	0.8600	C1–N1–H1B	120.0
N5–H5	0.84(3)	H1A–N1–H1B	120.0
N6–H6A	0.8600	C2–N2–C1	115.7(2)
N6–H6B	0.8600	C3–N3–C1	115.9(2)
N7–H7A	0.878(17)	C2–N4–H4A	120.0
N7–H7B	0.883(17)	C2–N4–H4B	120.0
		H4A–N4–H4B	120.0
		C2–N5–C3	120.0(2)
		C2–N5–H5	120(2)
		C3–N5–H5	120(2)
		C3–N6–H6A	120.0
		C3–N6–H6B	120.0
		H6A–N6–H6B	120.0
		S1–N7–H7A	106(2)
		S1–N7–H7B	108.6(19)
		H7A–N7–H7B	112(3)

Table S3. Hydrogen bonds for $C_3N_6H_7SO_3NH_2$.

D-H	d(D-H)	d(H..A)	∠DHA	d(D..A)
N1–H1A···O2$^{\#1}$	0.86	2.06	2.887(3)	162.0
N1–H1B···O1$^{\#2}$	0.86	2.15	2.863(3)	140.2
N4–H4A···N2$^{\#3}$	0.86	2.15	3.009(3)	177.7
N4–H4B···O2$^{\#4}$	0.86	2.29	2.903(3)	128.7
N4–H4B···O3	0.86	2.34	3.049(3)	139.4
N6–H6A···N3$^{\#5}$	0.86	2.16	3.019(3)	178.3
N6–H6B···N7$^{\#6}$	0.86	2.17	3.014(3)	167.7
N5–H5···S1	0.84(3)	2.96(3)	3.669(3)	144(2)
N5–H5···O3	0.84(3)	2.00(3)	2.780(3)	154(3)
N7–H7A···O3$^{\#4}$	0.878(17)	2.24(2)	3.061(3)	155(3)
N7–H7B···O1$^{\#7}$	0.883(17)	2.25(2)	3.086(3)	157(3)

Symmetry transformations used to generate equivalent atoms:

#1: +X, +Y, 1+Z; #2: 1-X, 2-Y, 1-Z; #3: -X, 1-Y, 1-Z; #4: -X, 1-Y, -Z; #5: 2-X, 2-Y, 1-Z; #6: 1+X, +Y, +Z; #7: -X, 2-Y, -Z;

Table S4. Optical properties of compounds containing [$C_3N_6H_7$] groups.

No.	Crystals	Band gap	Birefringence @546 nm	UV Cut-off (nm)	reference
1	$(C_3N_6H_7)_3HgCl_5$	4.19[e]	0.093@1064nm[c]	290	[1]
2	$(C_3N_6H_7)_2SiF_6·H_2O$	4.76[e]	0.152[c]	220	[2]
3	$(C_3N_6H_6)_2(C_3N_6H_7)PF_6·H_2O$	4.20[e]	0.243[c]	300	[3]
4	$(C_3N_6H_7)(C_3N_6H_6)HgCl_3$	4.40[e]	0.246@1064nm[c]	278	[1]
5	$(H_7C_3N_6)·(H_6C_3N_6)ZnCl_3$	3.95[e]	0.255@1064nm[c]	245	[4]
6	$C_3H_6N_6$	4.60[e]	0.26[c]	248	
7	$(C_3N_6H_6)_4HPF_6$	4.12[e]	0.264[c]	300	[3]
8	$2(C_3H_7N_6)^+·2Cl^-·H_2O$	4.75[e]	0.277[c]	245	[5]
9	$(C_3H_8N_6)PbBr_4$	3.13[e]	0.322[e]	374	[6]
10	$(C_3H_7N_8)SnCl_4$	3.71[e]	0.294[e]	334	[7]
11	$(C_3H_7N_6)_2Cl_2·H_2O$	4.70[e]	0.33[c]	230	[8]
12	**$C_3N_6H_7SO_3NH_2$**	**5.53[e]**	**0.34[c]**	**206**	**This work**
13	$(C_3N_6H_7)_2SbF_5·H_2O$	4.74[e]	0.38[c]	220	[9]
14	$(C_3H_7N_6)F·H_2O$	4.72[e]	0.38[c]	220	[8]
15	$[C_3N_6H_7]_2[B_3O_3F_4(OH)]$	4.72[e]	0.44[c]	240	[10]
16	$(C_3N_6H_7)_6(H_2PO_4)_4(HPO_4)·4H_2O$	4.60[e]	0.22[c]	/	[11]

Note: the upper corner labeled "e" stands for experimentally obtained and the upper corner labeled "c" indicates theoretically calculated.

Table S5. The shortest distance of $[C_3N_6H_7]$ units and band gaps of crystals**.**

Crystals	Band gap (eV)	Distance [Å]
$(H_7C_3N_6)\cdot(H_6C_3N_6)ZnCl_3$	3.95[e]	6.1363
$(C_3N_6H_6)_2(C_3N_6H_7)PF_6\cdot H_2O$	4.20[e]	6.2095
$(C_3N_6H_7)_2SiF_6\cdot H_2O$	4.76[e]	6.1955
$C_3N_6H_7SO_3NH_2$	**5.53**[e]	**6.2269**

Note: the upper corner labeled "e" for experimentally obtained.

Figure S1. SEM images and EDX microanalysis for $C_3N_6H_7SO_3NH_2$.

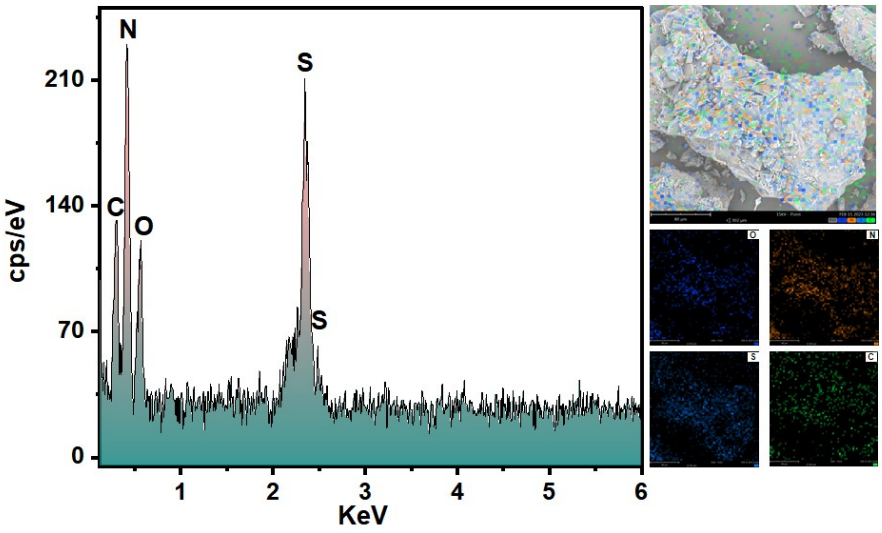


Figure S2. The IR spectrum for $C_3N_6H_7SO_3NH_2$

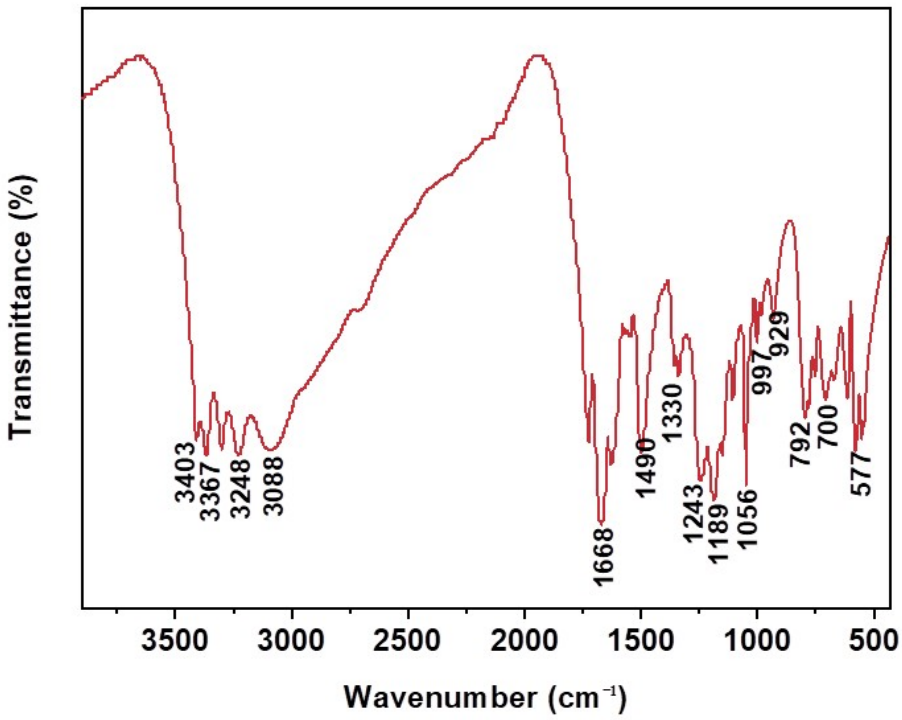


Figure S3. The UV-vis-NIR diffuse reflectance spectrum of $C_3N_6H_6$.

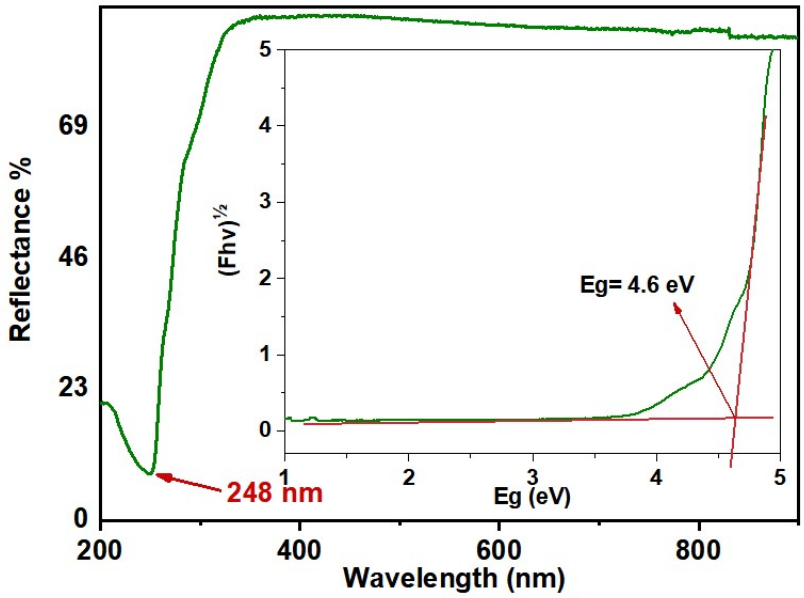


Figure S4. Crystal picture under polarizing microscope. (a) The original colour of crystal; (b) Crystal achieving complete extinction; (c) The thickness of crystal.

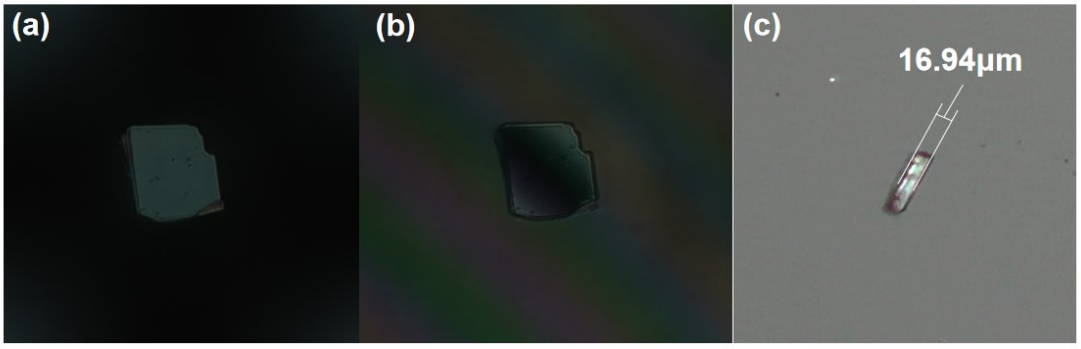
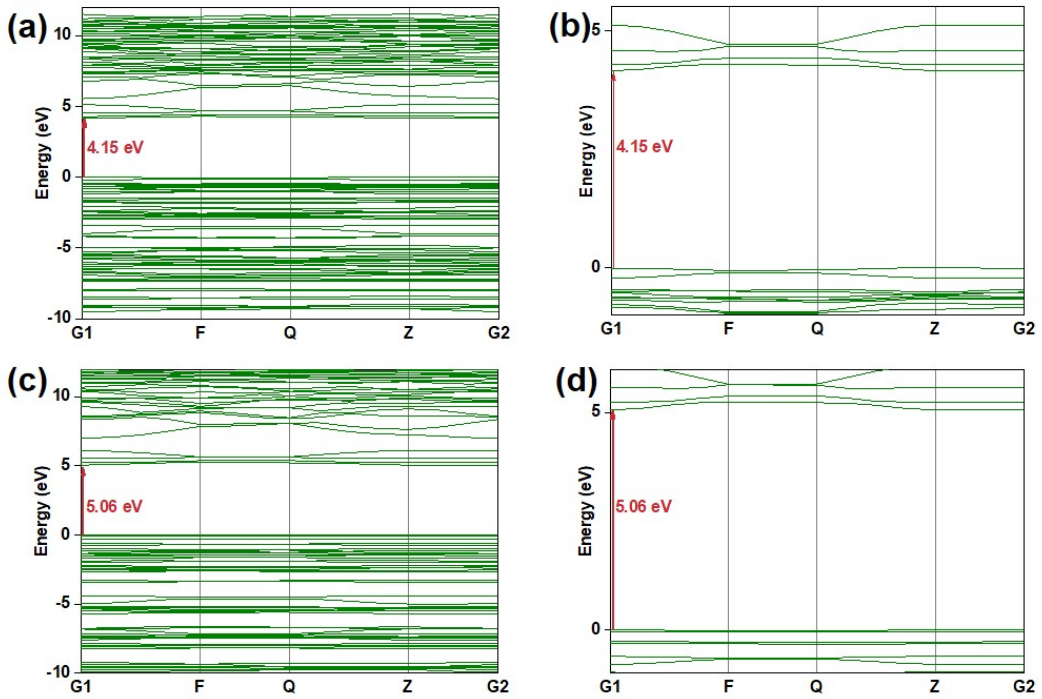


Figure S5. The calculated band gaps using GGA (a and b) and HSE06 functionals (c and d) for $C_3N_6H_7SO_3NH_2$.



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