Supplementary information

A Comprehensive Study on the Influence of Ortho/Para-Substituents, Stimulated Medium and External Field Wavelength on Photophysical and Non-Linear Optical Properties of Thienyl-Chalcone Derivatives

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1. NMR

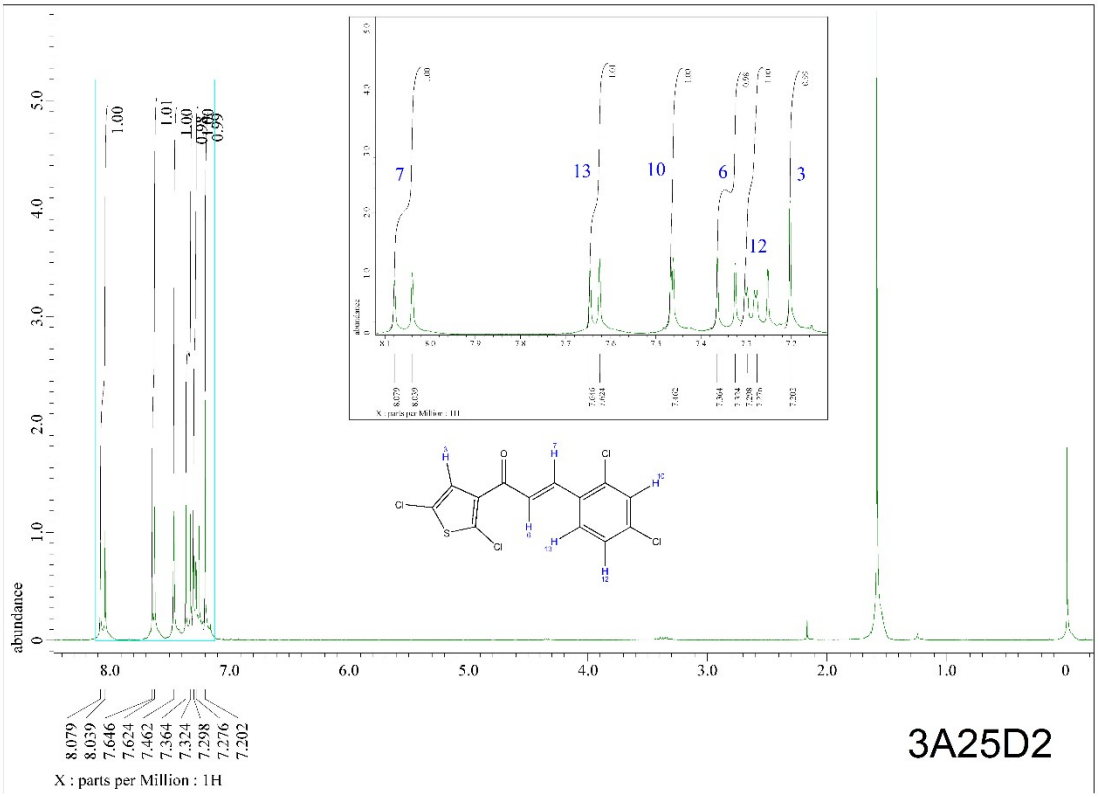


Figure S1 ^{1}H NMR spectrum of **3A25D2**.

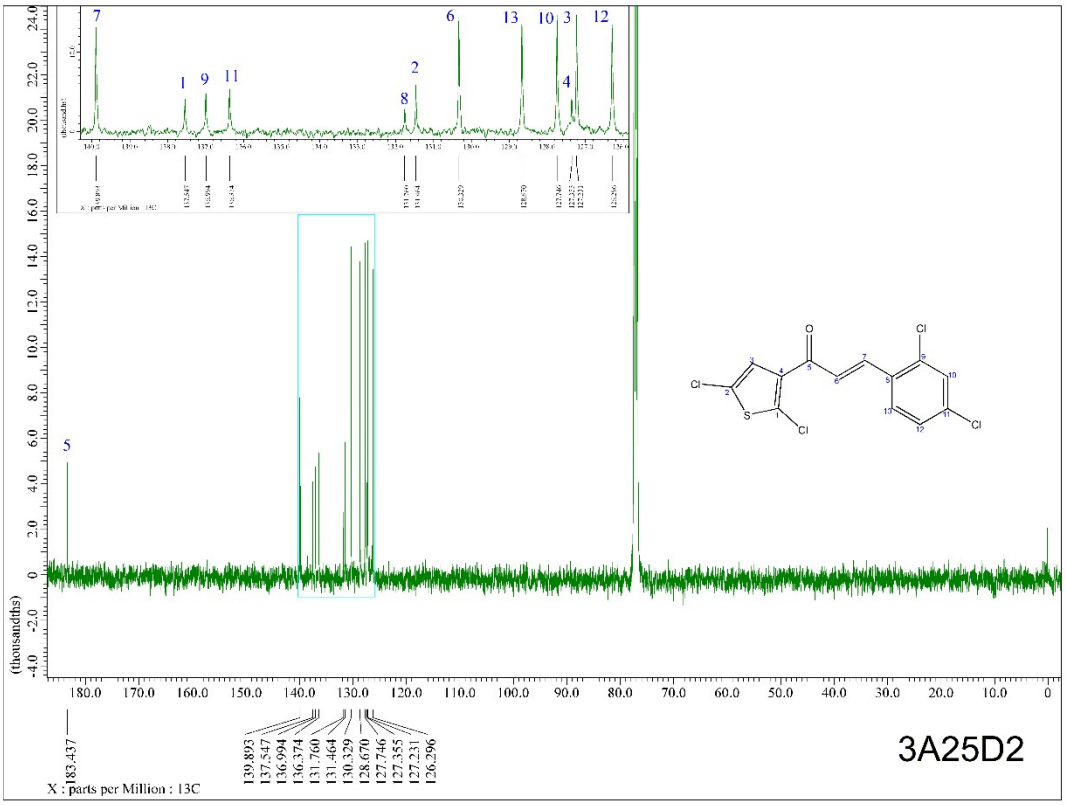


Figure S2 ^{13}C NMR spectrum of **3A25D2**

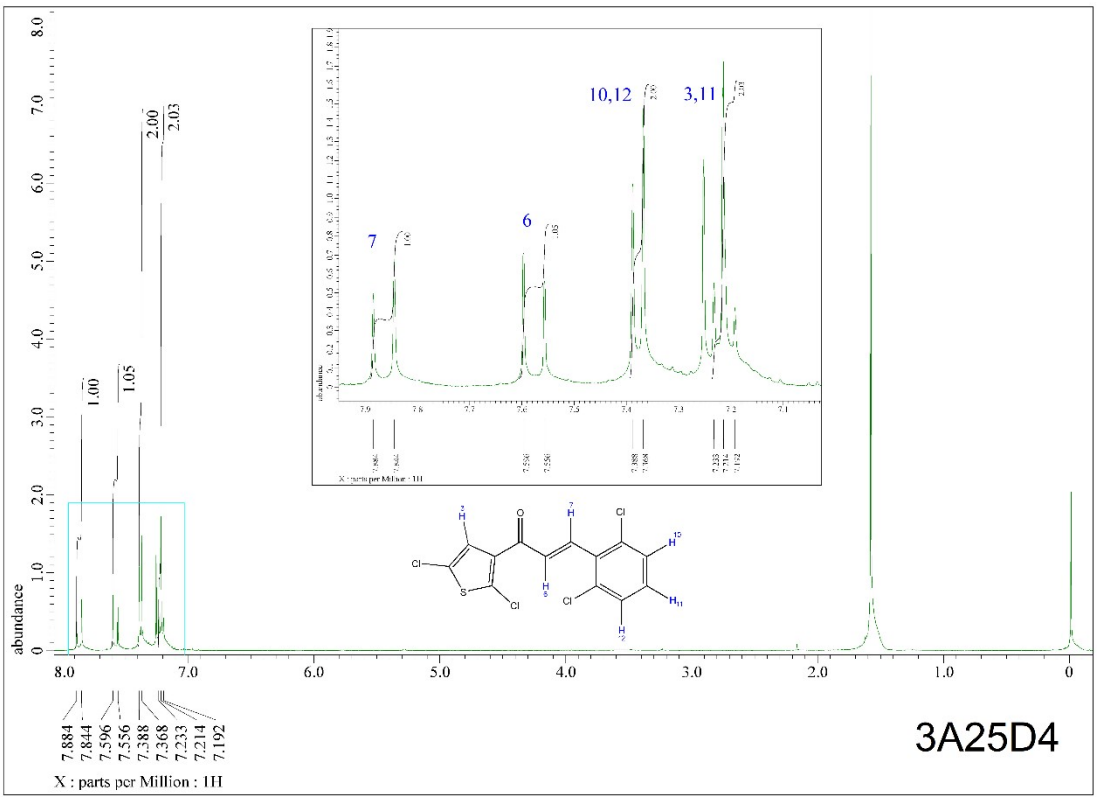


Figure S3 ^{1}H NMR spectrum of **3A25D4**.

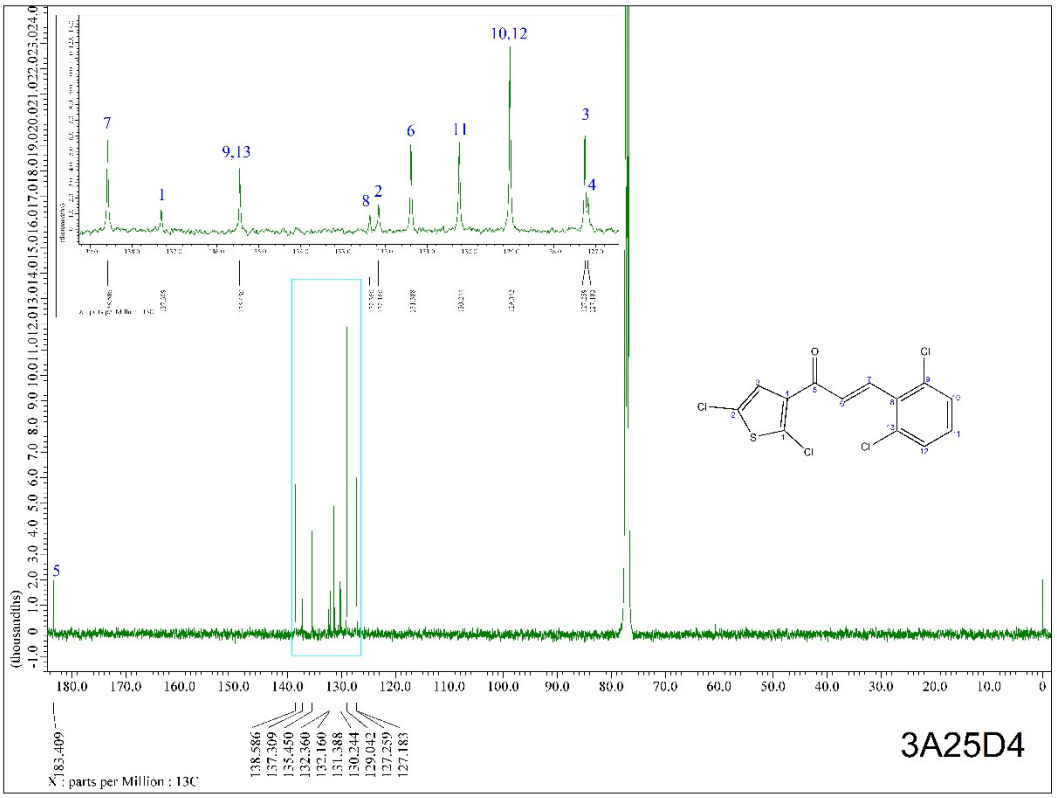


Figure S4 ^{13}C NMR spectrum of **3A25D4**.

2. FTIR

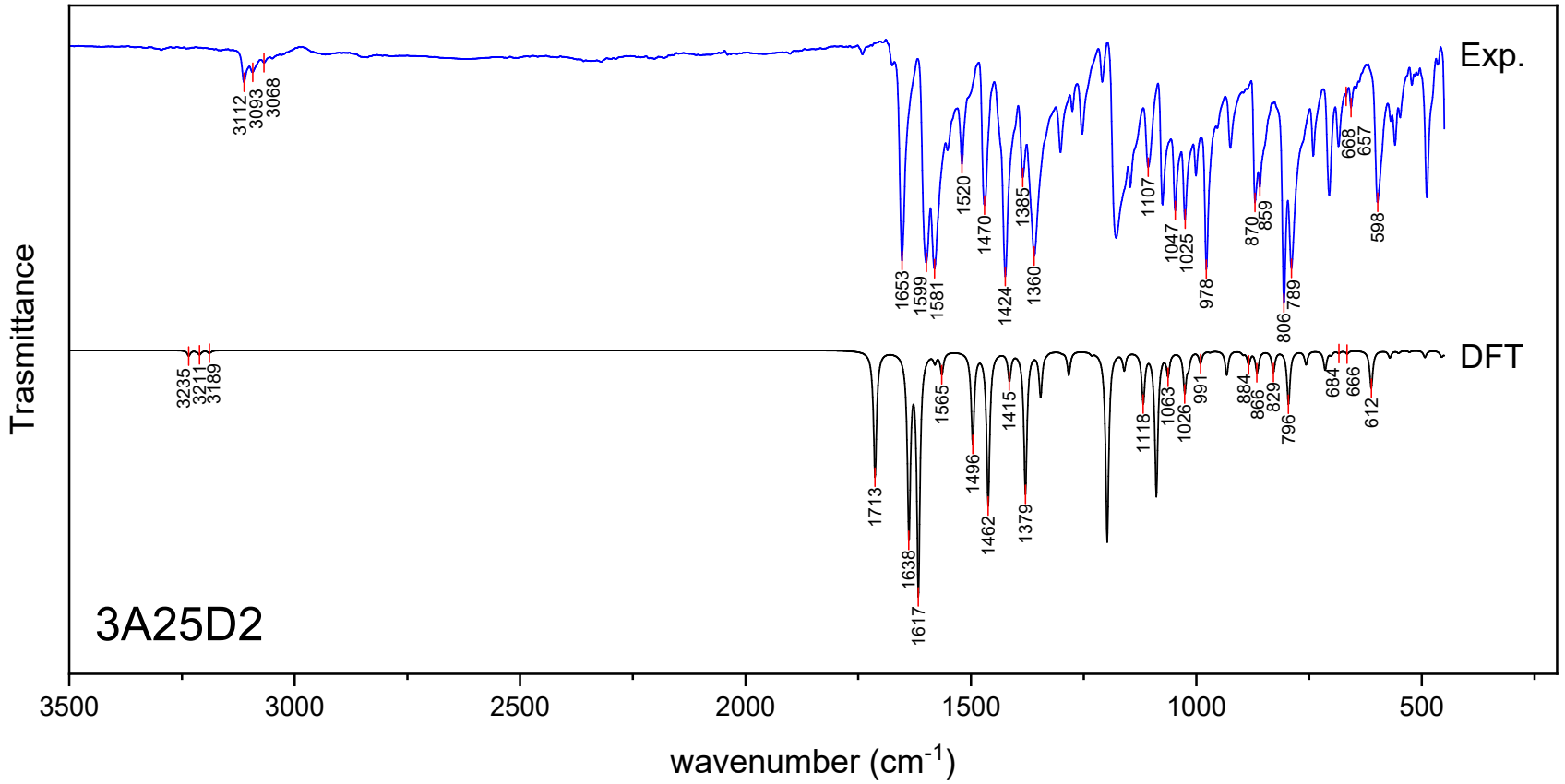


Figure S5 Experimental (top) and calculated (DFT) (below) FT-IR spectra of **3A25D2**.

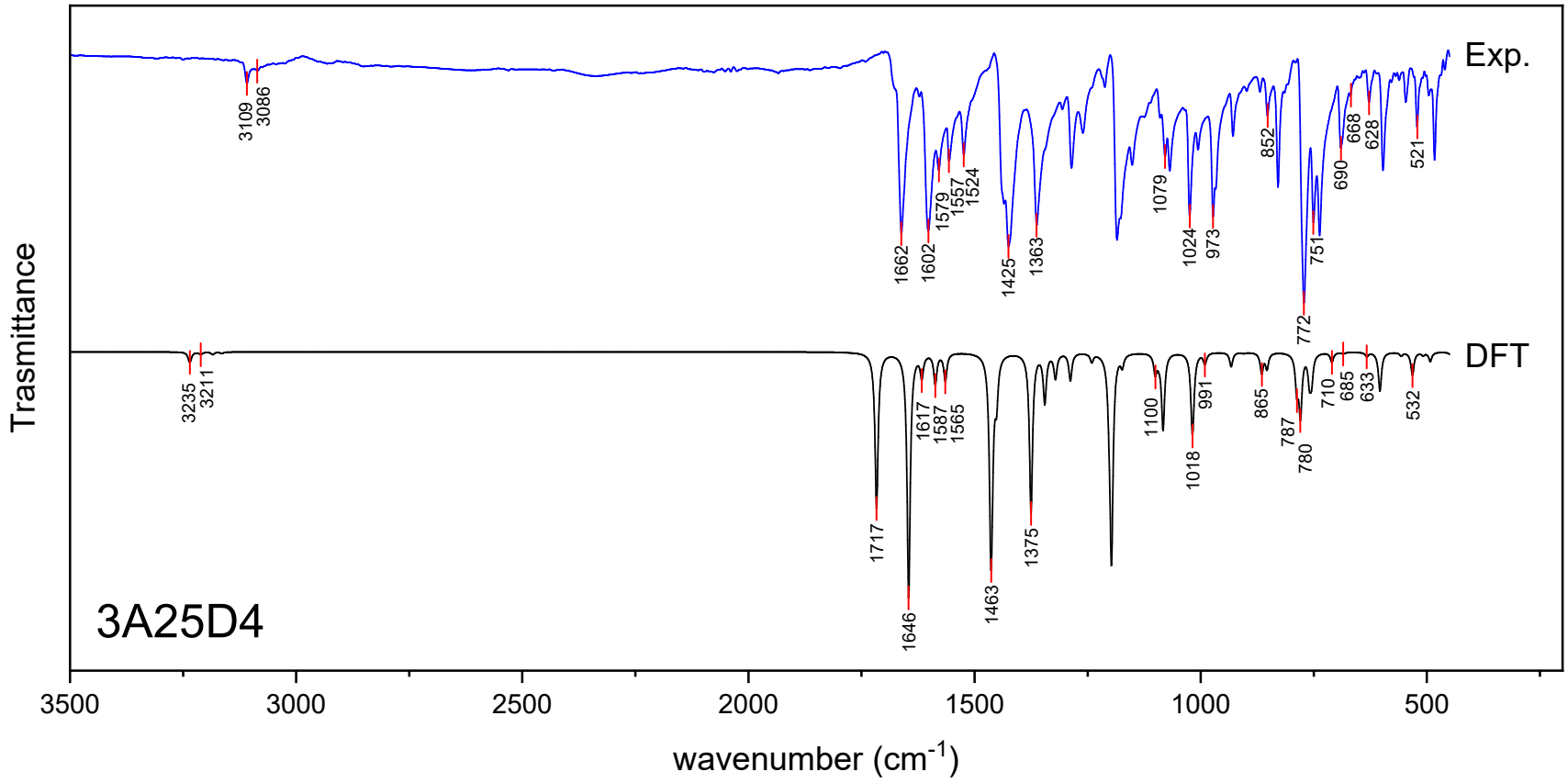


Figure S6 Experimental (top) and calculated (DFT) (below) FT-IR spectra of **3A25D4**.

3. Hirshfeld surface analysis and energy framework

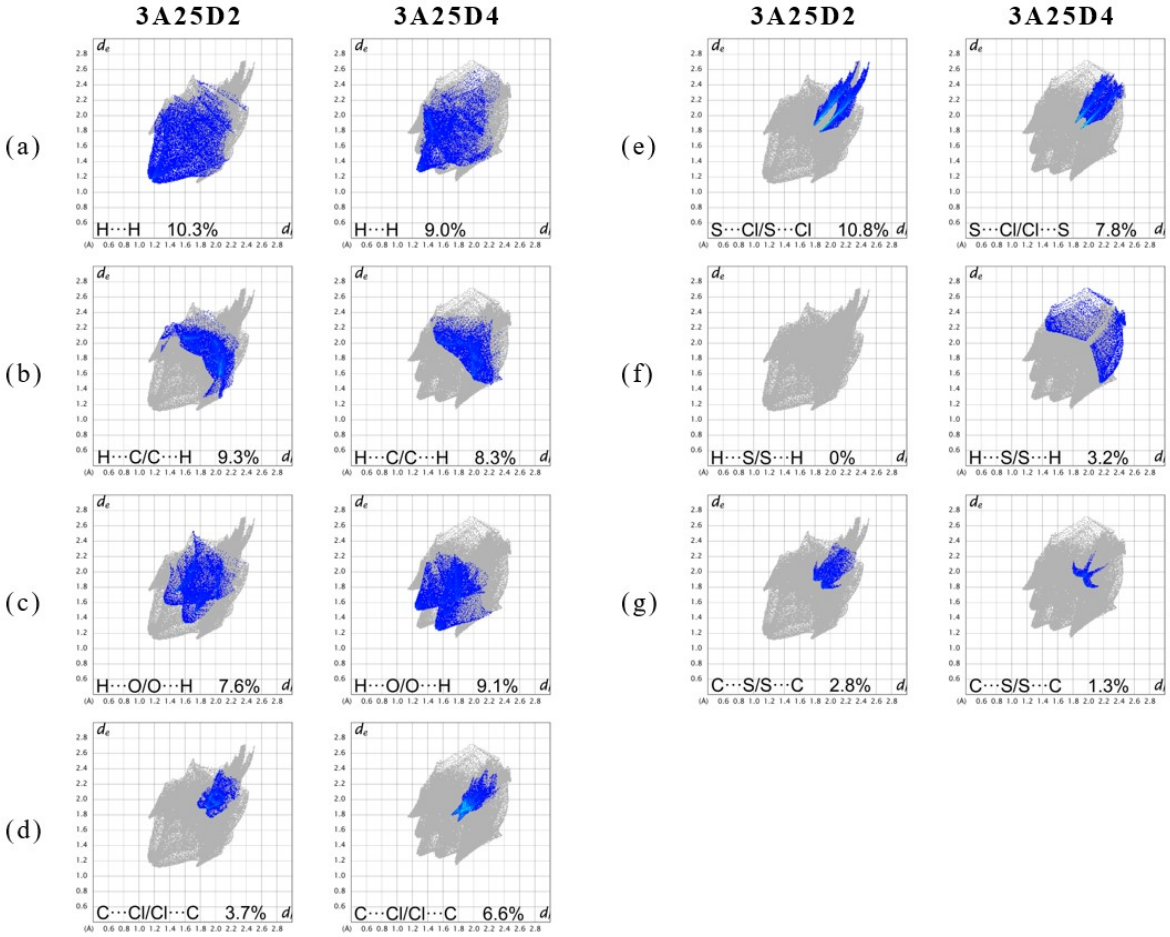


Figure S7 The resolved 2D fingerprint plots of intermolecular contacts: (a) H···H, (b) H···C/C···H, (c) H···O/O···H, (d) C···Cl/Cl···C, (e) S···Cl/Cl···S, (f) H···S/S···H, and (g) C···S/S···C with relative percentage contributions to the Hirshfeld surface area for **3A25D2** and **3A25D4**.

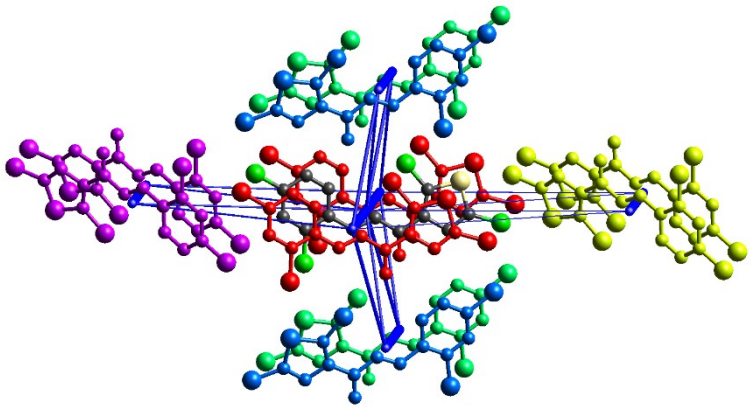


Figure S8 Total energy framework for **3A25D2** with cylinder scale of 50 for energies. H atoms have been omitted for clarity.

Table S1 Energy framework detail of molecular interaction energies with symmetry operations (Symop) for **3A25D2**. (Distances between molecular centroid (*R*) in Å, interaction energies in kJ/mol and number of pair-wise fragments (*N*)).

N	Symop	*R*	E_{ele}	E_{pol}	E_{dis}	E_{rep}	E_{tot}
1	*x*, *y*, *z*	3.99	-6.9	-0.9	-80.0	45.5	-49.5
1	*x*+1/2, -*y*+1/2, -*z*	13.22	-4.3	-0.1	-9.4	12.1	-5.4
0	-*x*, *y*+1/2, -*z*+1/2	6.96	-10.7	-1.8	-24.0	23.3	-19.2
2	-*x*, *y*+1/2, -*z*+1/2	7.09	-5.8	-1.4	-20.9	16.7	-15.1
1	*x*+1/2, -*y*+1/2, -*z*	12.65	-5.3	-0.3	-13.9	13.4	-9.7

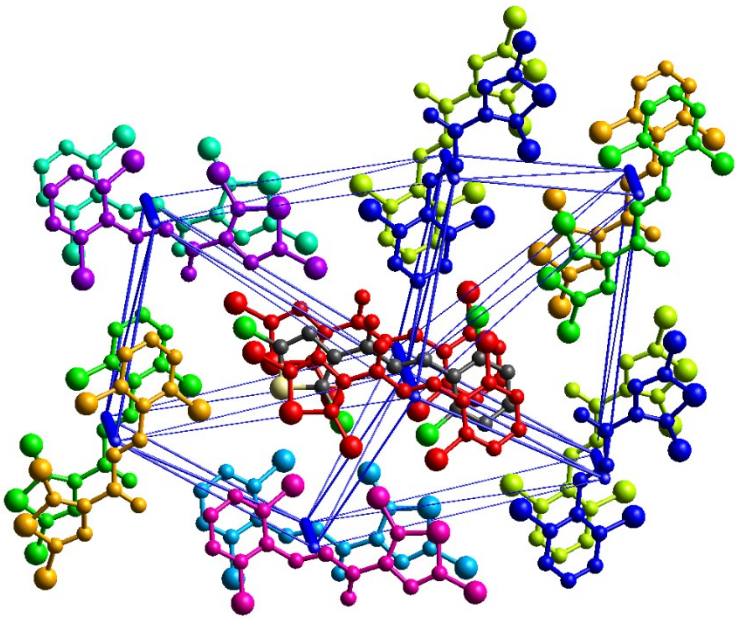


Figure S9 Total energy framework for **3A25D4** with cylinder scale of 50 for energies. H atoms have been omitted for clarity.

Table S2 Energy framework detail of molecular interaction energies with symmetry operations (Symop) for **3A25D4**. (Distances between molecular centroid (*R*) in Å, interaction energies in kJ/mol and number of pair-wise fragments (*N*)).

N	Symop	*R*	E_{ele}	E_{pol}	E_{dis}	E_{rep}	E_{tot}
2	*x*, *y*, *z*	3.85	-11.5	-0.9	-80.7	52.6	-50.6
2	*x*+1/2, -*y*+1/2, *z*+1/2	12.41	-3.8	-0.3	-8.2	9.2	-5.7
2	-*x*+1/2, *y*+1/2, -*z*+1/2	10.46	-5.6	-1.5	-13.4	7.7	-13.9
2	*x*+1/2, -*y*+1/2, *z*+1/2	12.62	-1.3	-0.2	-7.3	5.0	-4.8
1	-*x*, -*y*, -*z*	11.60	-2.7	-0.1	-8.2	6.4	-6.2
1	-*x*, -*y*, -*z*	6.91	-5.5	-0.2	-19.1	20.8	-9.7
2	-*x*+1/2, *y*+1/2, -*z*+1/2	10.59	-3.6	-1.5	-10.7	5.9	-10.6
1	-*x*, -*y*, -*z*	10.82	-5.4	-0.6	-13.8	13.0	-10.1
1	-*x*, -*y*, -*z*	7.75	-4.2	-0.1	-13.4	14.4	-7.4

4. DFT optimised geometry

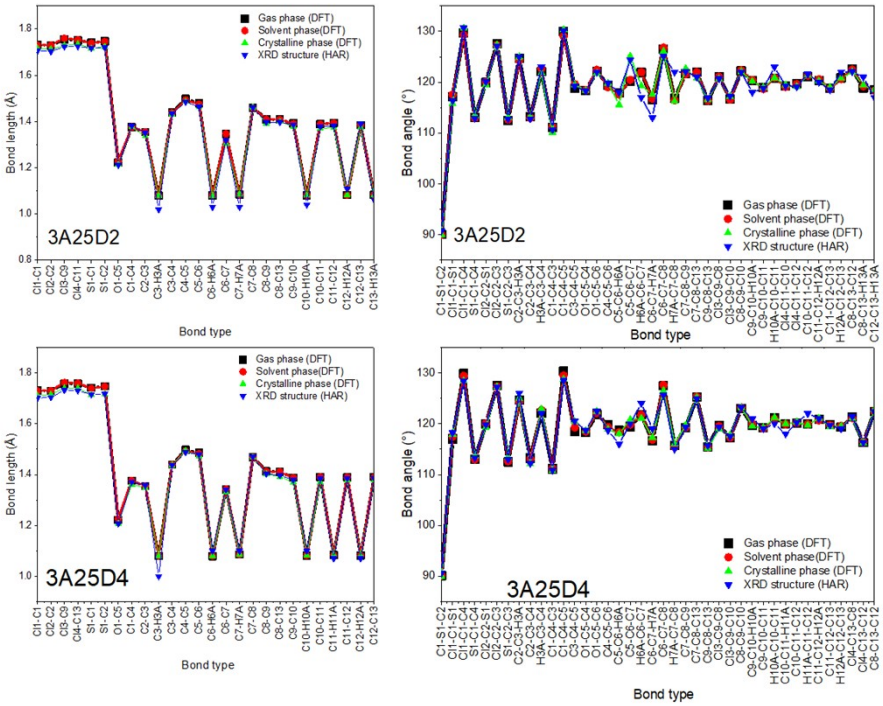


Figure S10 The graph of bond length and bond angle variation for optimized geometries **3A25D2** and **3A25D4** along with HAR-refined structures.

Table S3 The single point energy of distinct phase optimised structure (kJ/mol).

	3A25D2	**3A25D4**
Gas	-7386685	-7386666
Solvent	-7386707	-7386687
Crystalline	-7457710	-7446944

Table S4 Bond length alternation for **3A25D2** and **3A25D4**. (Å)

	3A25D2	**3A25D4**
Gas	-0.12425	-0.13567
Solvent	-0.12002	-0.13219
Crystalline	-0.15765	-0.1401

5. Heat map of charge transfer

i. Gas

ii. Solvent

iii. Crystalline

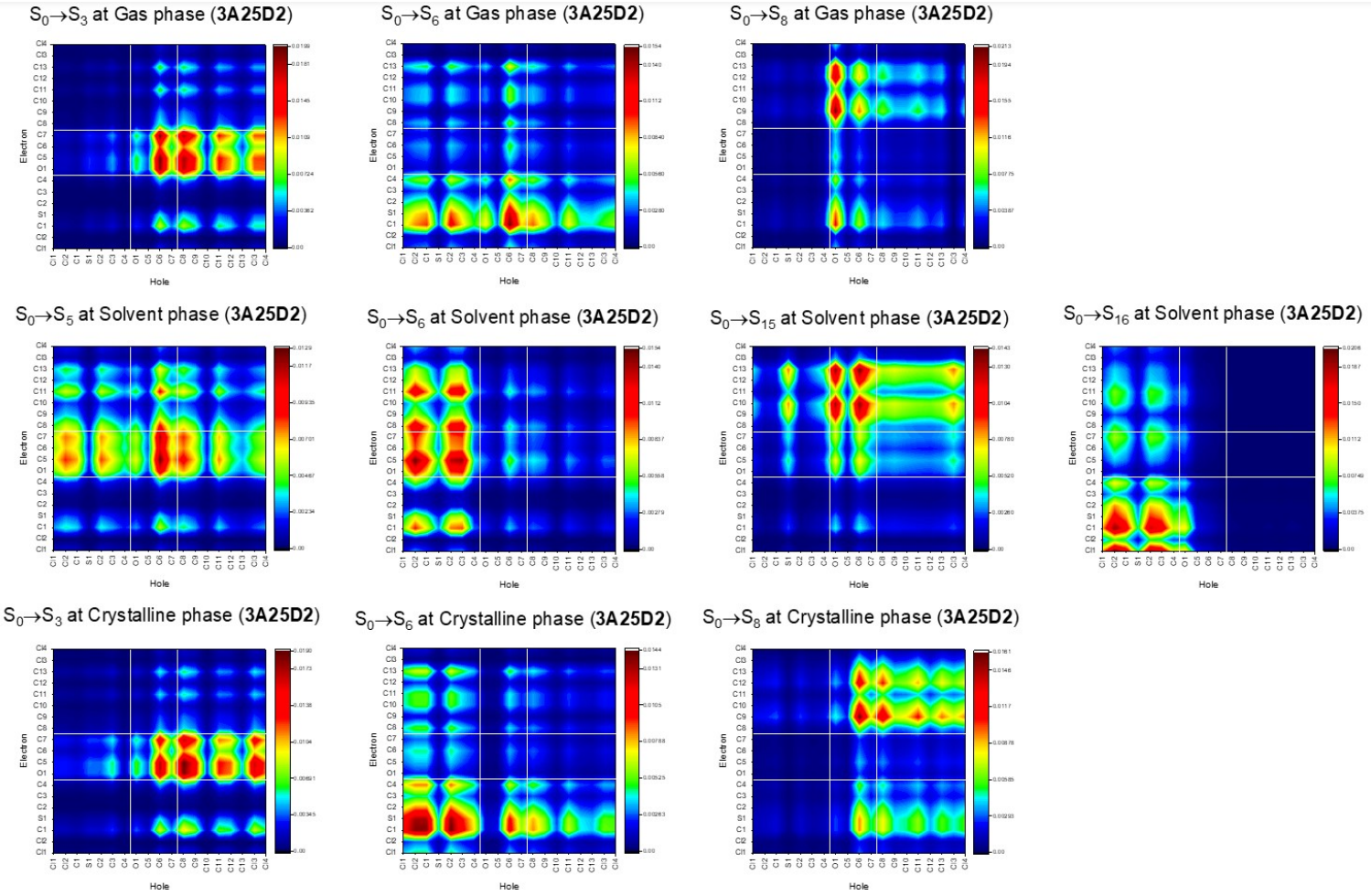
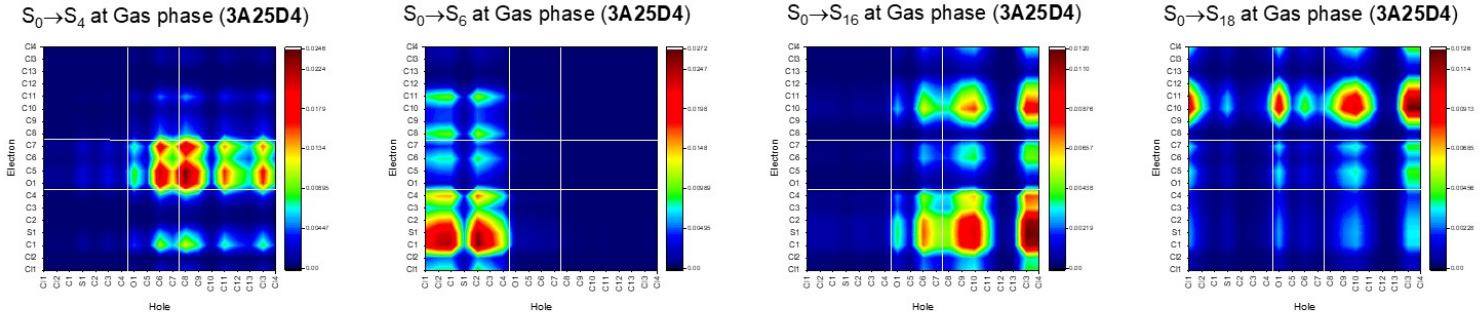
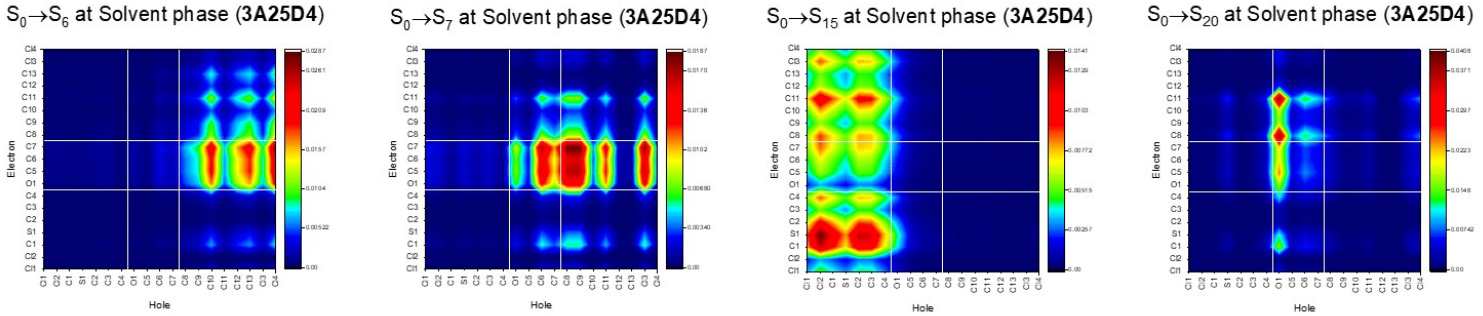


Figure S11 Heat maps of charge transfer for **3A25D2** at distinct phases.

i. Gas



ii. Solvent



iii. Crystalline

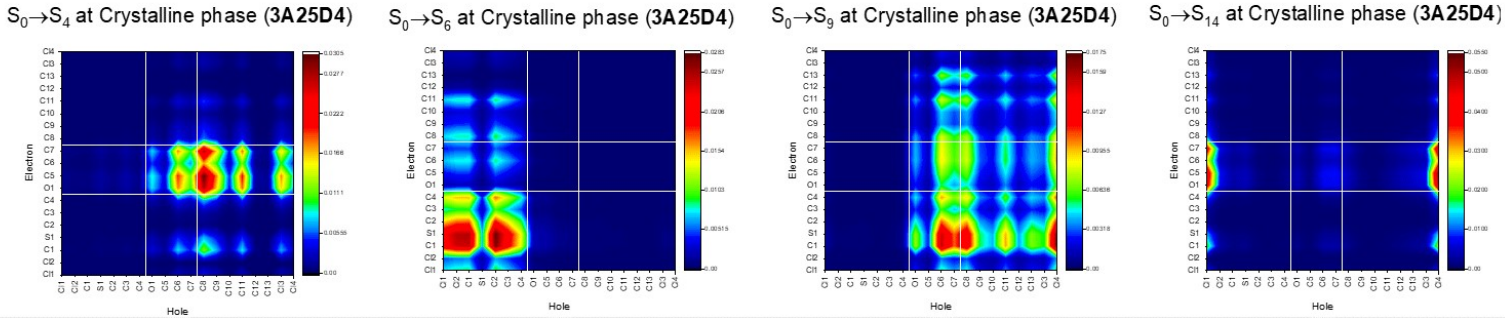


Figure S12 Heat maps of charge transfer for **3A25D4** at distinct phases.

6. Dipole moment and (hyper)polarizabilities

Table S5 The ground electric dipole moment tensor components and total values. (Debye)

Phase	**3A25D2**				**3A25D4**			
	μ_x	μ_y	μ_z	$\langle\mu\rangle$	μ_x	μ_y	μ_z	$\langle\mu\rangle$
Gas	-0.52	-2.86	-0.60	2.96	-2.59	1.97	-1.81	3.72
Solvent	-0.67	-4.44	-0.91	4.58	-3.31	2.93	-2.79	5.23
Crystalline	0.31	3.83	0.24	3.85	1.10	4.58	1.28	4.88

Table S6 The static and dynamic polarizability tensor components and average values for **3A25D2**. ($\times 10^{-24}$ esu)

λ (nm)	α_{xx}	α_{xy}	α_{yy}	α_{xz}	α_{yz}	α_{zz}	$\langle\alpha\rangle$	$\Delta\alpha$
Gas phase								
0	60.21	2.61	33.42	-1.25	0.25	19.48	37.70	36.21
532	75.91	2.99	35.87	-0.80	0.20	20.26	44.01	50.01
637	69.54	2.88	35.01	-1.02	0.23	20.00	41.51	44.32
655	68.88	2.86	34.91	-1.04	0.24	19.97	41.25	43.74
800	65.50	2.77	34.37	-1.13	0.25	19.80	39.89	40.77
1064	62.98	2.70	33.94	-1.19	0.25	19.66	38.86	38.58
1200	62.35	2.68	33.82	-1.20	0.25	19.62	38.60	38.04
1310	61.99	2.67	33.76	-1.21	0.25	19.60	38.45	37.72
1600	61.38	2.65	33.64	-1.22	0.25	19.56	38.19	37.20
999999	60.21	2.61	33.42	-1.25	0.25	19.48	37.70	36.21
Solvent phase								
0	78.54	3.57	48.45	-1.81	0.28	27.14	51.38	45.26
532	91.71	3.67	42.05	-0.58	0.16	22.86	52.20	61.88
637	81.81	3.48	40.78	-1.01	0.22	22.50	48.37	52.98
655	80.82	3.45	40.64	-1.05	0.22	22.46	47.98	52.10
800	75.88	3.31	39.88	-1.23	0.24	22.24	46.00	47.75
1064	72.32	3.19	39.27	-1.34	0.25	22.06	44.55	44.65
1200	71.44	3.16	39.11	-1.37	0.25	22.01	44.19	43.89
1310	70.93	3.14	39.02	-1.38	0.25	21.98	43.98	43.45
1600	70.10	3.11	38.86	-1.40	0.25	21.93	43.63	42.73
999999	68.50	3.05	38.55	-1.44	0.26	21.84	42.96	41.37
Crystalline phase								
0	24.50	0.59	35.19	15.29	-5.64	50.48	36.72	36.19
532	27.73	0.28	37.96	20.66	-7.08	61.66	42.45	48.37
637	26.45	0.41	36.99	18.48	-6.54	57.20	40.21	43.42
655	26.31	0.42	36.88	18.25	-6.48	56.73	39.97	42.91
800	25.62	0.49	36.27	17.09	-6.16	54.31	38.73	40.28
1064	25.09	0.54	35.78	16.24	-5.92	52.50	37.79	38.33
1200	24.96	0.55	35.65	16.02	-5.86	52.04	37.55	37.84
1310	24.88	0.56	35.58	15.90	-5.82	51.77	37.41	37.56
1600	24.75	0.57	35.45	15.69	-5.76	51.33	37.18	37.09
999999	24.50	0.59	35.19	15.29	-5.64	50.48	36.72	36.19

Table S7 The static and dynamic polarizability tensor components and average values for **3A25D4**. ($\times 10^{-24}$ esu)

λ (nm)	α_{xx}	α_{xy}	α_{yy}	α_{xz}	α_{yz}	α_{zz}	$\langle\alpha\rangle$	$\Delta\alpha$
Gas phase								
0	52.30	-1.06	35.73	1.08	-1.10	18.97	35.67	29.04
532	61.89	-0.88	38.32	1.35	-1.27	19.67	39.96	36.82
637	58.20	-0.98	37.42	1.25	-1.20	19.44	38.35	33.77
655	57.80	-0.99	37.31	1.24	-1.19	19.41	38.18	33.45
800	55.72	-1.02	36.74	1.18	-1.16	19.26	37.24	31.76
1064	54.12	-1.04	36.28	1.13	-1.13	19.13	36.51	30.48
1200	53.71	-1.05	36.16	1.12	-1.12	19.10	36.32	30.15
1310	53.47	-1.05	36.09	1.11	-1.12	19.08	36.21	29.96
1600	53.07	-1.05	35.97	1.10	-1.11	19.04	36.03	29.65
999999	52.30	-1.06	35.73	1.08	-1.10	18.97	35.67	29.04
Solvent phase								
0	67.59	-0.85	52.16	1.31	-2.27	26.94	48.90	35.86
532	72.56	-0.61	45.23	1.65	-1.85	22.29	46.70	43.82
637	67.08	-0.84	43.90	1.44	-1.73	21.97	44.32	39.29
655	66.50	-0.85	43.75	1.42	-1.72	21.94	44.06	38.82
800	63.55	-0.93	42.94	1.31	-1.65	21.73	42.74	36.44
1064	61.34	-0.98	42.30	1.22	-1.60	21.56	41.73	34.68
1200	60.78	-0.99	42.13	1.20	-1.59	21.51	41.47	34.24
1310	60.46	-1.00	42.03	1.19	-1.58	21.49	41.32	33.98
1600	59.92	-1.00	41.86	1.17	-1.57	21.44	41.07	33.56
999999	58.88	-1.02	41.53	1.13	-1.55	21.35	40.59	32.75
Crystalline phase								
0	19.86	-4.55	39.36	0.28	7.67	44.69	34.63	27.42
532	20.61	-5.11	43.42	0.17	10.34	51.15	38.39	34.00
637	20.36	-4.92	41.95	0.21	9.31	48.71	37.01	31.46
655	20.33	-4.89	41.78	0.22	9.20	48.44	36.85	31.19
800	20.17	-4.77	40.90	0.24	8.62	47.04	36.03	29.76
1064	20.03	-4.67	40.19	0.26	8.17	45.94	35.39	28.66
1200	19.99	-4.65	40.00	0.26	8.06	45.66	35.22	28.38
1310	19.97	-4.63	39.90	0.27	7.99	45.50	35.12	28.22
1600	19.93	-4.60	39.72	0.27	7.89	45.22	34.96	27.95
999999	19.86	-4.55	39.36	0.28	7.67	44.69	34.63	27.42

Table S8 The static and dynamic first hyperpolarizability tensor components and the projection of first hyperpolarizability on dipole moment vector μ, βvec for **3A25D2**. ($\times 10^{-30}$ esu)

	(-ω; ω,0)				(-2ω; ω, ω)			
λ (nm)	β_x	β_y	β_z	β_{vec}	β_x	β_y	β_z	β_{vec}
Gas phase								
0	-19.43	-8.00	-0.50	11.20				
532	-56.44	-23.33	-0.61	32.44	738.78	140.93	12.52	-267.05
637	-38.82	-15.74	-0.70	22.07	696.92	26.09	10.84	-148.72
655	-37.18	-15.07	-0.69	21.14	1638.67	95.26	47.40	-386.79
800	-29.41	-11.93	-0.64	16.75	-89.85	-51.81	2.21	65.14
1064	-24.34	-9.91	-0.57	13.91	-41.54	-16.96	-0.70	23.72
1200	-23.15	-9.44	-0.55	13.24	-34.49	-14.00	-0.68	19.64
1310	-22.49	-9.17	-0.54	12.87	-31.11	-12.62	-0.65	17.72
1600	-21.42	-8.75	-0.52	12.27	-26.34	-10.71	-0.60	15.03
999999	-19.45	-7.96	-0.49	11.16	-19.45	-7.96	-0.49	11.16
Solvent phase								
0	-51.16	-26.50	-2.45	33.64				
532	-124.55	-58.91	-4.36	76.14	1206.37	234.24	0.55	-403.22
637	-81.40	-38.10	-3.55	49.51	596.32	-12.98	-3.32	-73.81
655	-77.55	-36.34	-3.44	47.22	870.74	56.14	22.57	-186.00
800	-59.76	-28.32	-2.89	36.74	-176.62	-148.25	3.38	168.78
1064	-48.55	-23.32	-2.49	30.18	-76.09	-34.66	-3.30	45.35
1200	-45.98	-22.17	-2.39	28.67	-61.64	-28.11	-2.91	36.82
1310	-44.55	-21.53	-2.33	27.83	-54.93	-25.17	-2.71	32.95
1600	-42.24	-20.50	-2.24	26.48	-45.69	-21.16	-2.38	27.65
999999	-38.06	-18.62	-2.06	24.02	-32.81	-15.61	-1.87	20.29
Crystalline phase								
0	-7.34	11.32	-15.63	9.68				
532	-21.15	30.10	-45.72	25.34	92.13	-144.57	207.77	-123.22
637	-14.66	21.07	-31.43	17.79	446.70	-146.25	886.58	-53.27
655	-14.05	20.25	-30.10	17.10	5416.74	-1883.26	10683.20	-759.45
800	-11.14	16.34	-23.78	13.85	-33.42	56.82	-75.47	49.05
1064	-9.23	13.78	-19.65	11.72	-15.66	22.51	-33.62	19.00
1200	-8.78	13.18	-18.68	11.22	-13.04	18.91	-27.90	16.00
1310	-8.53	12.84	-18.14	10.94	-11.78	17.21	-25.16	14.57
1600	-8.12	12.29	-17.26	10.48	-9.98	14.80	-21.28	12.57
999999	-7.37	11.28	-15.65	9.63	-7.37	11.28	-15.65	9.63

Table S9 The static and dynamic first hyperpolarizability tensor components and the projection of first hyperpolarizability on dipole moment vector μ, βvec for **3A25D4**. ($\times 10^{-30}$ esu)

	(-ω; ω,0)				(-2ω; ω, ω)			
λ(nm)	β_x	β_y	β_z	β_{vec}	β_x	β_y	β_z	β_{vec}
Gas phase								
0	0.46	2.91	-2.83	2.59				
532	-4.36	9.89	-7.99	12.14	119.21	-67.95	43.68	-140.13
637	-2.13	6.43	-5.53	7.57	-1503.68	-90.51	-178.17	1085.44
655	-1.91	6.12	-5.30	7.14	-463.97	18.25	-38.99	351.59
800	-0.84	4.68	-4.21	5.10	-3.46	21.04	-14.83	20.73
1064	-0.15	3.75	-3.49	3.78	-2.43	6.98	-5.92	8.26
1200	0.01	3.53	-3.32	3.47	-1.52	5.62	-4.93	6.42
1310	0.10	3.41	-3.22	3.30	-1.06	4.99	-4.45	5.54
1600	0.24	3.22	-3.07	3.02	-0.42	4.11	-3.78	4.30
999999	0.49	2.86	-2.78	2.52	0.49	2.86	-2.78	2.52
Solvent phase								
0	-3.75	11.90	-10.52	14.66				
532	-19.01	26.57	-21.60	38.46	234.79	-93.02	58.78	-232.15
637	-11.53	17.10	-14.68	24.72	1047.80	-55.89	119.44	-758.29
655	-10.81	16.28	-14.06	23.48	1073330	-9674.98	138639	-758788
800	-7.40	12.54	-11.15	17.67	-11.94	55.20	-38.37	58.99
1064	-5.22	10.19	-9.27	13.96	-11.26	15.36	-13.30	22.84
1200	-4.72	9.64	-8.83	13.11	-8.61	12.37	-11.01	18.26
1310	-4.44	9.34	-8.58	12.63	-7.32	11.01	-9.94	16.11
1600	-3.99	8.86	-8.19	11.86	-5.50	9.15	-8.44	13.11
999999	-3.19	7.97	-7.45	10.47	-2.95	6.55	-6.28	8.90
Crystalline phase								
0	1.35	4.93	0.02	4.94				
532	2.66	16.44	3.19	16.86	-20.79	-67.78	47.74	-55.72
637	2.09	10.95	1.73	11.20	18.26	625.38	846.47	813.36
655	2.03	10.44	1.59	10.67	-23.40	302.54	321.91	363.16
800	1.75	8.01	0.89	8.14	4.07	28.95	2.76	28.79
1064	1.55	6.42	0.45	6.49	2.19	11.79	1.92	12.06
1200	1.50	6.04	0.34	6.10	1.94	9.60	1.33	9.79
1310	1.47	5.84	0.29	5.88	1.81	8.54	1.04	8.69
1600	1.43	5.50	0.20	5.53	1.63	7.05	0.62	7.14
999999	1.34	4.88	0.03	4.89	1.34	4.88	0.03	4.89

Table S10 The static and dynamic second hyperpolarizability tensor components and average second hyperpolarizability, ⟨γ⟩ for **3A25D2**. ($\times 10^{-34}$ esu)

	(-ω; ω,0,0)							(-2ω; ω, ω,0)						
λ(nm)	γ_{xxxx}	γ_{yyyy}	γ_{zzzz}	γ_{xxyy}	γ_{xxzz}	γ_{yyzz}	⟨γ⟩	γ_{xxxx}	γ_{yyyy}	γ_{zzzz}	γ_{xxyy}	γ_{xxzz}	γ_{yyzz}	⟨γ⟩
Gas phase														
0	4.09	0.18	0.12	0.28	0.09	0.05	1.05							
532	16.55	0.27	0.15	0.89	0.19	0.07	3.93	-241.82	0.40	0.28	0.96	-0.38	0.23	-46.78
637	9.66	0.24	0.14	0.57	0.14	0.07	2.34	120.40	0.35	0.19	-4.69	-0.65	0.09	25.25
655	9.12	0.24	0.14	0.55	0.14	0.07	2.22	781.70	0.40	0.18	-8.60	-0.41	0.10	157.06
800	6.73	0.22	0.13	0.43	0.12	0.06	1.67	55.17	0.41	0.16	2.87	0.34	0.09	12.91
1064	5.33	0.21	0.13	0.36	0.10	0.06	1.34	10.70	0.25	0.14	0.62	0.14	0.07	2.57
1200	5.02	0.21	0.13	0.34	0.10	0.06	1.27	8.28	0.23	0.14	0.50	0.13	0.06	2.02
1310	4.85	0.21	0.13	0.33	0.10	0.06	1.23	7.24	0.23	0.14	0.45	0.12	0.06	1.78
1600	4.58	0.20	0.13	0.32	0.09	0.06	1.17	5.87	0.22	0.13	0.38	0.11	0.06	1.47
999999	4.09	0.20	0.12	0.29	0.09	0.06	1.06	4.09	0.20	0.12	0.29	0.09	0.06	1.06
Solvent Phase														
0	11.85	0.60	0.30	1.09	0.24	0.15	3.14							
532	44.65	0.64	0.28	3.38	0.49	0.15	10.77	-347.13	0.85	0.56	0.97	-1.31	0.54	-68.34
637	23.71	0.54	0.26	1.93	0.33	0.13	5.82	133.01	0.66	0.31	-5.52	-0.62	0.20	47.93
655	22.17	0.53	0.26	1.82	0.32	0.13	5.46	115.80	0.67	0.29	-6.52	-0.60	0.14	26.68
800	15.63	0.48	0.25	1.34	0.26	0.12	3.92	231.77	1.31	0.27	18.99	0.98	0.19	56.59
1064	11.99	0.45	0.24	1.07	0.22	0.11	3.06	23.36	0.46	0.22	1.68	0.29	0.11	5.63
1200	11.21	0.44	0.24	1.01	0.21	0.11	2.87	17.28	0.43	0.22	1.28	0.25	0.11	4.23
1310	10.78	0.44	0.24	0.97	0.21	0.11	2.77	14.78	0.41	0.21	1.12	0.23	0.10	3.64
1600	10.10	0.43	0.23	0.92	0.20	0.11	2.61	11.62	0.39	0.21	0.91	0.20	0.10	2.91
999999	8.91	0.42	0.23	0.83	0.19	0.10	2.32	7.72	0.35	0.19	0.65	0.16	0.09	2.00
Crystalline phase														
0	0.28	0.21	2.46	0.11	0.61	0.31	1.00							
532	0.80	0.30	9.20	0.27	2.52	1.03	3.53	2.90	0.56	48.67	0.22	12.04	4.15	16.15
637	0.52	0.26	5.56	0.19	1.46	0.64	2.17	14.49	0.34	208.96	-0.31	54.41	15.59	70.25
655	0.50	0.26	5.26	0.18	1.38	0.61	2.05	-42913	73.02	-9843	126.84	-2477	-313.43	-45907
800	0.39	0.24	3.95	0.15	1.01	0.47	1.56	2.12	0.36	25.09	0.57	7.34	2.51	9.45
1064	0.33	0.23	3.17	0.13	0.80	0.39	1.27	0.56	0.27	6.11	0.20	1.60	0.66	2.36
1200	0.32	0.22	2.99	0.13	0.75	0.37	1.20	0.46	0.25	4.81	0.17	1.24	0.54	1.87
1310	0.31	0.22	2.90	0.12	0.72	0.36	1.17	0.42	0.25	4.23	0.16	1.08	0.48	1.66
1600	0.30	0.22	2.74	0.12	0.68	0.34	1.11	0.36	0.23	3.47	0.14	0.88	0.41	1.38
999999	0.28	0.21	2.46	0.11	0.61	0.31	1.00	0.28	0.21	2.46	0.11	0.61	0.31	1.00

Table S11 The static and dynamic second hyperpolarizability tensor components and average second hyperpolarizability, ⟨γ⟩ for **3A25D4**. ($\times 10^{-34}$ esu)

	(-ω; ω,0,0)							(-2ω; ω, ω,0)						
λ(nm)	γ_{xxxx}	γ_{yyyy}	γ_{zzzz}	γ_{xxyy}	γ_{xxzz}	γ_{yyzz}	⟨γ⟩	γ_{xxxx}	γ_{yyyy}	γ_{zzzz}	γ_{xxyy}	γ_{xxzz}	γ_{yyzz}	⟨γ⟩
Gas phase														
0	2.48	0.22	0.12	0.19	0.11	0.05	0.71							
532	8.19	0.30	0.15	0.43	0.24	0.08	2.10	66.89	0.75	0.24	1.28	0.49	0.17	14.87
637	5.20	0.27	0.14	0.31	0.18	0.07	1.37	3171	3.60	0.38	-65.25	18.48	2.96	589.92
655	4.95	0.27	0.14	0.30	0.18	0.07	1.31	164.71	1.62	0.22	9.82	2.13	0.31	43.64
800	3.82	0.25	0.13	0.25	0.15	0.06	1.03	20.70	0.37	0.16	0.97	0.47	0.10	5.12
1064	3.12	0.24	0.13	0.22	0.13	0.06	0.87	5.66	0.28	0.14	0.33	0.19	0.07	1.47
1200	2.97	0.24	0.13	0.21	0.13	0.06	0.83	4.56	0.26	0.14	0.29	0.17	0.06	1.21
1310	2.88	0.23	0.13	0.21	0.13	0.06	0.81	4.06	0.26	0.13	0.26	0.15	0.06	1.09
1600	2.74	0.23	0.13	0.20	0.12	0.06	0.77	3.39	0.25	0.13	0.23	0.14	0.06	0.93
999999	2.49	0.23	0.12	0.19	0.11	0.05	0.71	2.49	0.23	0.12	0.19	0.11	0.05	0.71
Solvent Phase														
0	6.88	0.69	0.32	0.63	0.37	0.15	2.04							
532	21.09	0.72	0.28	1.45	0.80	0.17	5.50	81.08	1.48	0.38	1.94	0.57	0.28	17.42
637	12.35	0.62	0.26	0.94	0.53	0.14	3.29	417.10	1.11	0.43	10.71	7.15	0.36	92.65
655	11.66	0.61	0.26	0.90	0.50	0.13	3.12	-4623	476.34	-70.92	-5247	-4195	-0.16	-2706
800	8.63	0.56	0.25	0.71	0.40	0.12	2.36	66.38	0.86	0.27	3.95	1.92	0.26	16.88
1064	6.85	0.53	0.24	0.60	0.34	0.11	1.92	12.00	0.53	0.22	0.81	0.45	0.12	3.12
1200	6.46	0.52	0.24	0.57	0.32	0.11	1.82	9.29	0.50	0.22	0.66	0.37	0.11	2.46
1310	6.24	0.51	0.24	0.56	0.32	0.11	1.76	8.13	0.48	0.21	0.59	0.34	0.11	2.17
1600	5.90	0.50	0.23	0.53	0.30	0.11	1.68	6.60	0.45	0.21	0.50	0.29	0.10	1.80
999999	5.28	0.49	0.23	0.49	0.28	0.10	1.52	4.62	0.41	0.19	0.39	0.23	0.09	1.32
Crystalline phase														
0	0.13	0.55	1.24	0.07	0.08	0.44	0.62							
532	0.15	1.26	4.04	0.10	0.13	1.19	1.72	0.33	2.81	84.32	0.70	2.27	5.88	23.21
637	0.14	0.92	2.56	0.09	0.11	0.82	1.15	0.19	229.31	531.61	3.72	3.61	387.00	284.72
655	0.14	0.88	2.43	0.09	0.11	0.79	1.11	0.29	465.32	63.87	39.74	53.63	198.60	174.67
800	0.13	0.74	1.88	0.08	0.10	0.63	0.89	0.16	2.26	10.46	0.13	0.16	2.18	3.74
1064	0.13	0.64	1.55	0.08	0.09	0.53	0.75	0.14	0.97	2.79	0.09	0.11	0.87	1.23
1200	0.13	0.62	1.47	0.07	0.09	0.51	0.72	0.14	0.83	2.24	0.08	0.10	0.73	1.02
1310	0.13	0.61	1.43	0.07	0.09	0.50	0.70	0.14	0.77	2.00	0.08	0.10	0.67	0.93
1600	0.13	0.59	1.36	0.07	0.09	0.48	0.67	0.13	0.68	1.68	0.08	0.09	0.57	0.80
999999	0.12	0.55	1.24	0.07	0.08	0.44	0.62	0.12	0.55	1.24	0.07	0.08	0.44	0.62

Table S12 The third harmonic generation $\gamma(-3\omega;\omega,\omega,\omega)$ and intensity-dependent refractive index $\gamma(-\omega;\omega,\omega,-\omega)$ values. ($\times 10^{-34}$ esu)

	3A25D2			**3A25D4**		
λ(nm)	Gas	Solvent	Crystalline	Gas	Solvent	Crystalline
$(-3\omega; \omega, \omega, \omega)$						
532	18.37	48.94	16.16	9.07	22.83	7.22
637	8.83	19.25	7.98	4.69	9.57	3.83
655	8.08	17.06	7.32	4.33	8.53	3.54
800	4.77	7.80	4.36	2.68	3.98	2.22
1064	2.83	2.64	2.60	1.67	1.32	1.40
1200	2.40	1.52	2.20	1.44	0.72	1.21
1310	2.16	0.91	1.99	1.31	0.40	1.11
1600	1.79	-0.06	1.64	1.10	-0.12	0.94
999999	1.11	-1.76	1.01	0.73	-1.05	0.63
$(-\omega; \omega, \omega, -\omega)$						
532	6.82	18.41	6.05	3.50	8.97	2.82
637	3.64	8.51	3.33	2.03	4.55	1.69
655	3.39	7.78	3.11	1.91	4.20	1.59
800	2.29	4.69	2.12	1.36	2.68	1.15
1064	1.64	2.97	1.53	1.03	1.80	0.88
1200	1.50	2.60	1.40	0.95	1.60	0.82
1310	1.42	2.40	1.33	0.91	1.49	0.78
1600	1.29	2.07	1.21	0.84	1.32	0.73
999999	1.07	1.51	1.01	0.71	1.01	0.62