Supplementary Material

Figure A: UV-Visible Absorption spectra of Croconate dyes 1 - 10 in DMF.

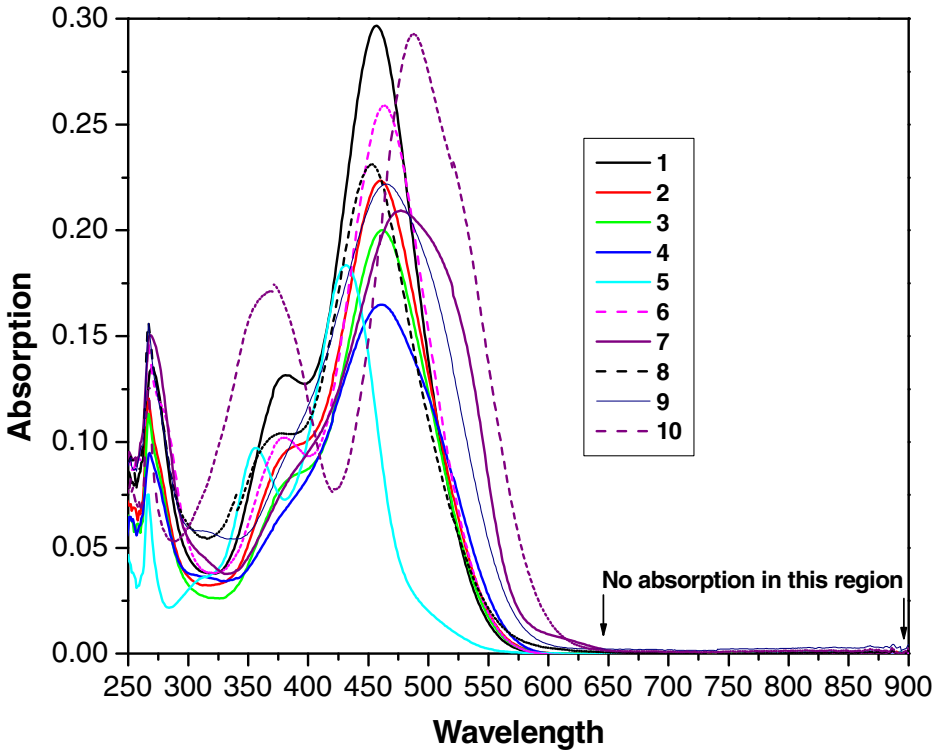
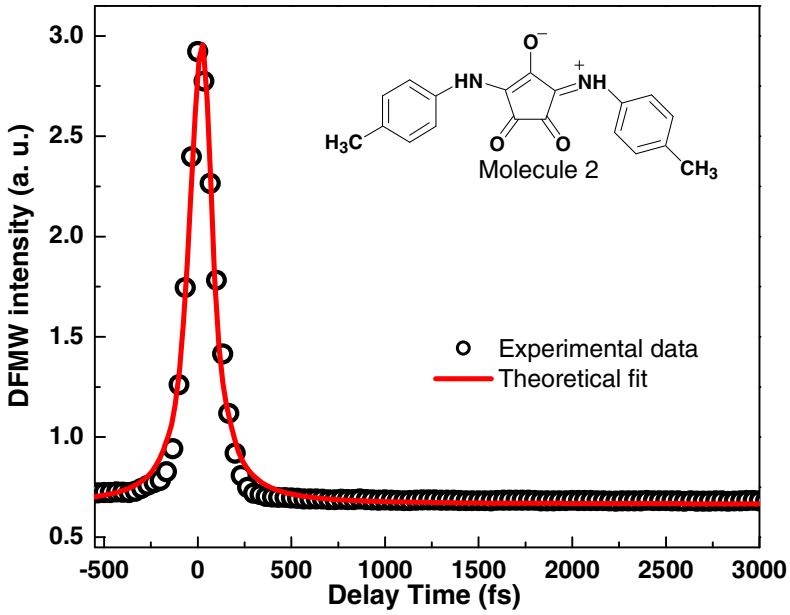
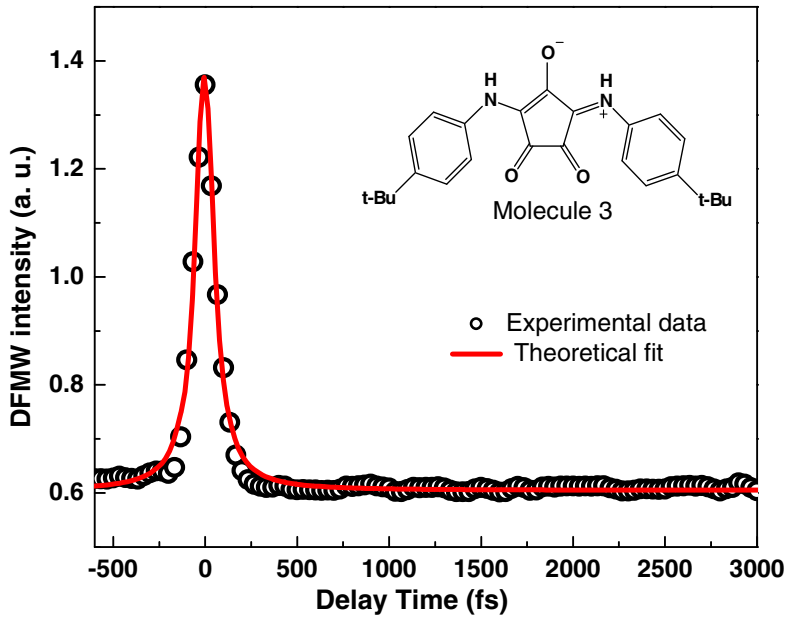
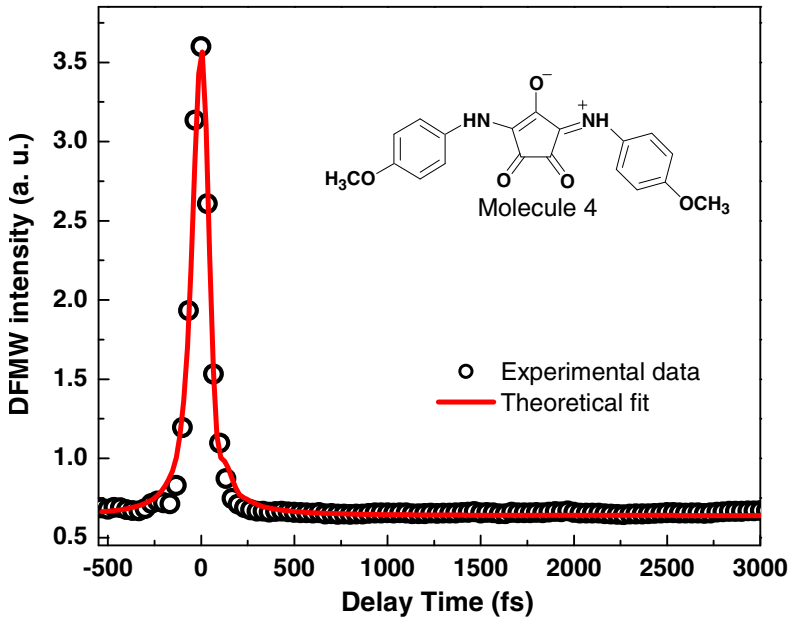
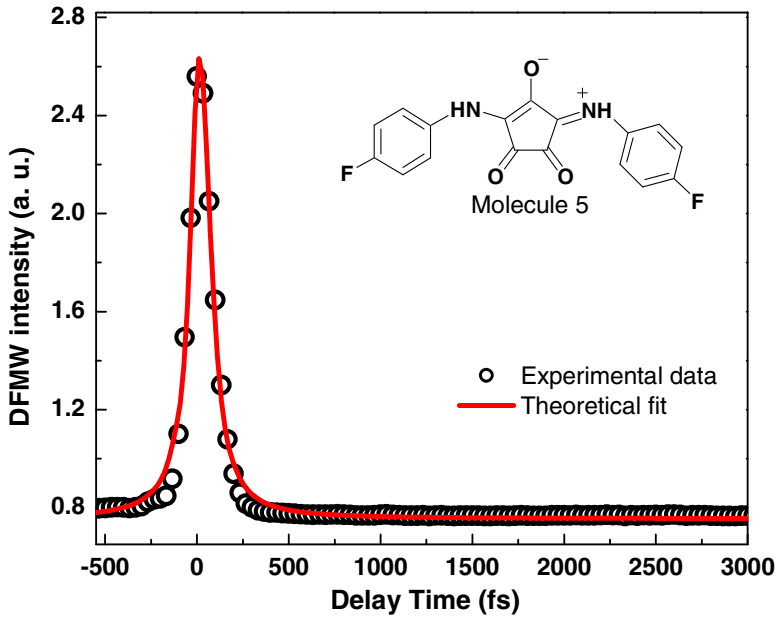


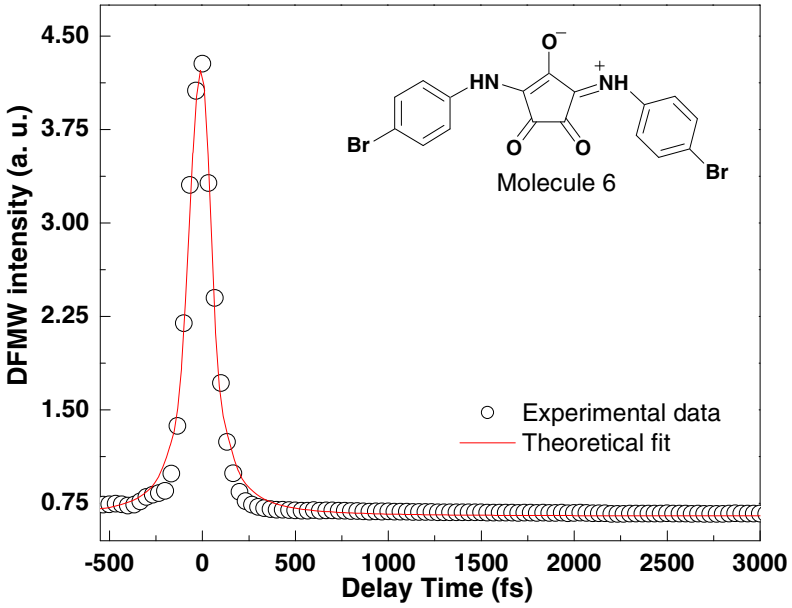
Figure B: Temporal profiles of DFWM signal of the molecules 2-10 in DMF (concentration: 5×10^{-5} M) as a function of beam 3 delay time for the parallel configurations.

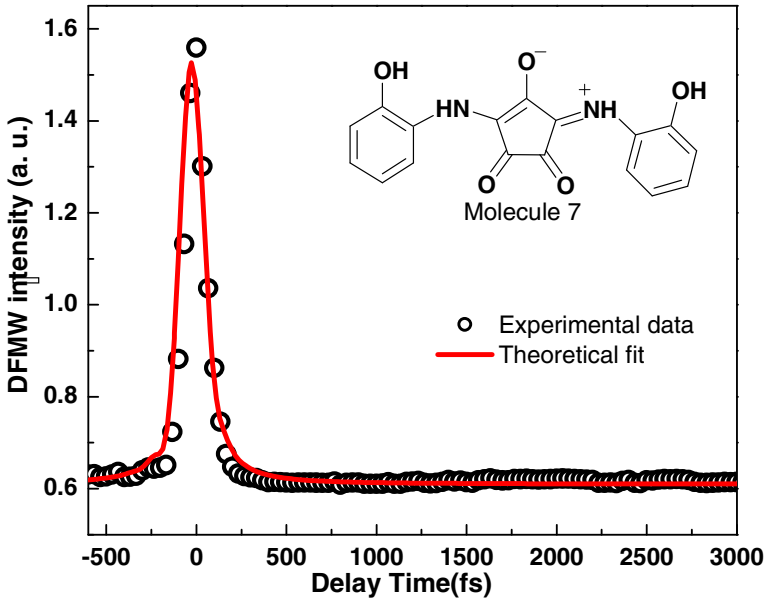


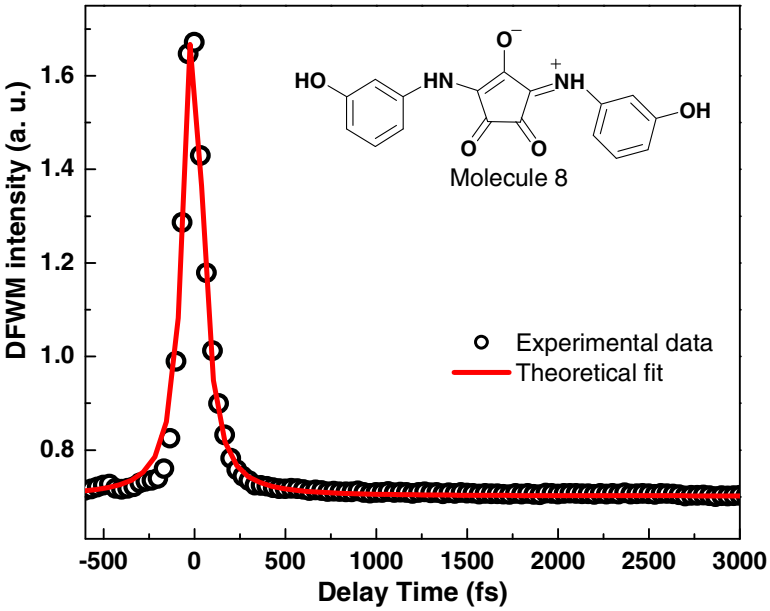

DFMW intensity (a. u.)
1.4
1.2
1.0
0.8
0.6
-500
0
500
1000
1500
2000
2500
3000
Delay Time (fs)
Molecule 3
t-Bu
t-Bu
Experimental data
Theoretical fit

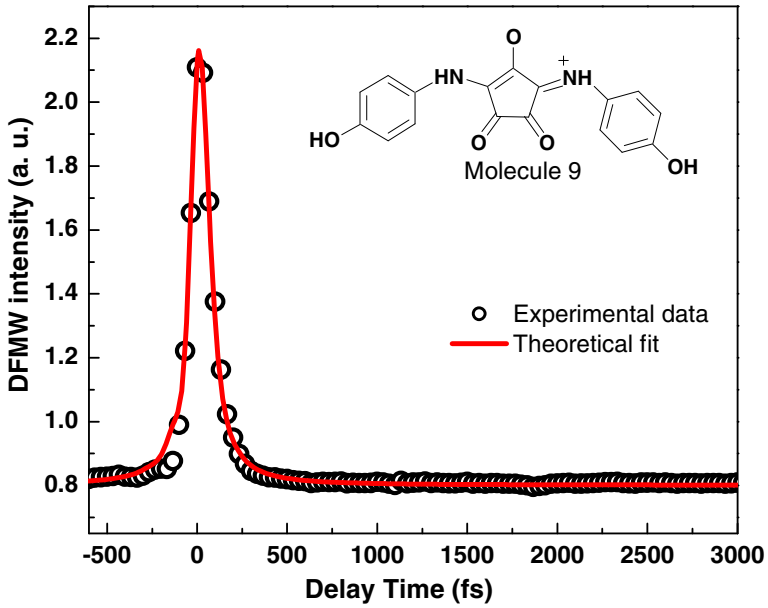

DFMW intensity (a. u.)
3.5
3.0
2.5
2.0
1.5
1.0
0.5
-500
0
500
1000
1500
2000
2500
3000
Delay Time (fs)
Molecule 4
H3CO
OCH3
Experimental data
Theoretical fit


DFMW intensity (a. u.)
Delay Time (fs)
Experimental data
Theoretical fit
Molecule 5
HN
NH
O
F


DFMW intensity (a. u.)
Delay Time (fs)
Experimental data
Theoretical fit
Molecule 6
HN
NH
O
Br


DFWM intensity (a. u.)
1.6
1.4
1.2
1.0
0.8
0.6
-500
0
500
1000
1500
2000
2500
3000
Delay Time(fs)
OH
HN
O⁻
+
NH
OH
O
O
Molecule 7
Experimental data
Theoretical fit


DFWM intensity (a. u.)
1.6
1.4
1.2
1.0
0.8
-500
0
500
1000
1500
2000
2500
3000
Delay Time (fs)
HO
HN
O⁻
+
NH
OH
O
O
Molecule 8
Experimental data
Theoretical fit


DFMW intensity (a. u.)
Delay Time (fs)
Molecule 9
Experimental data
Theoretical fit

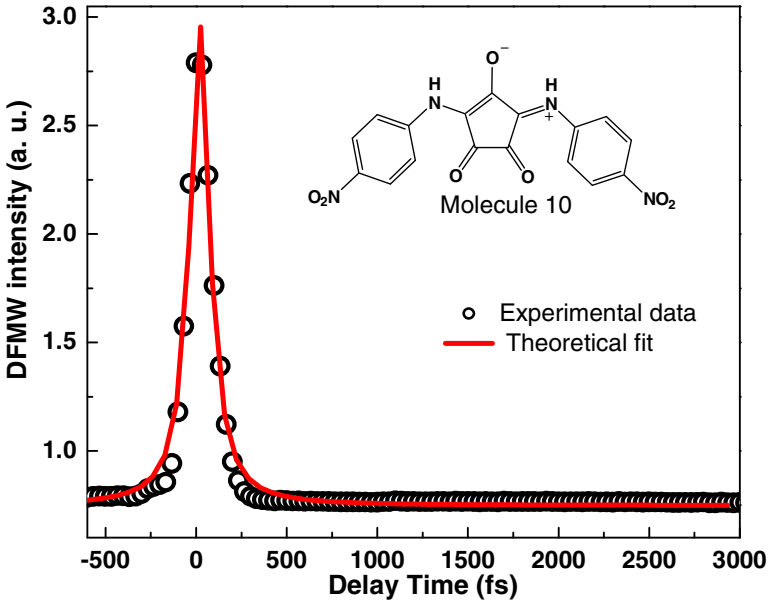
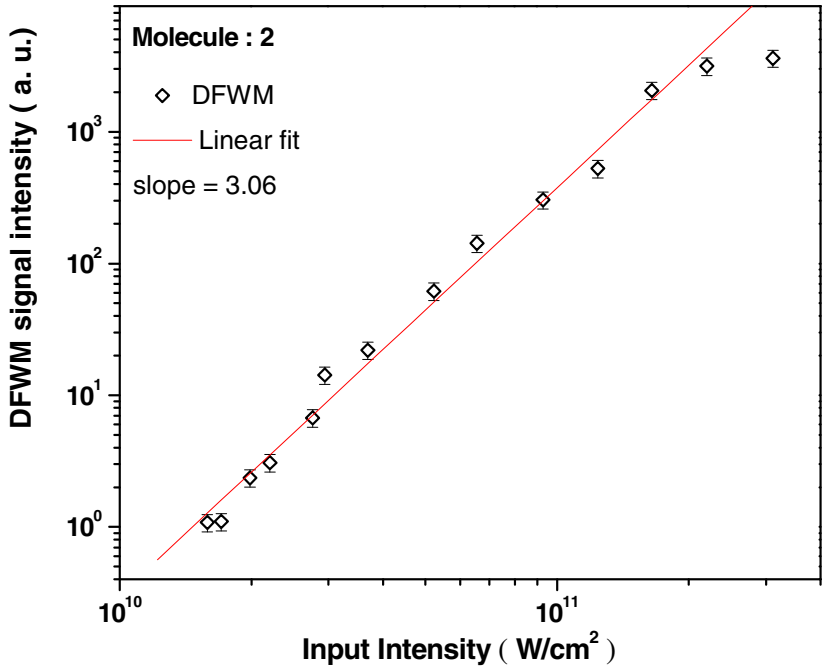
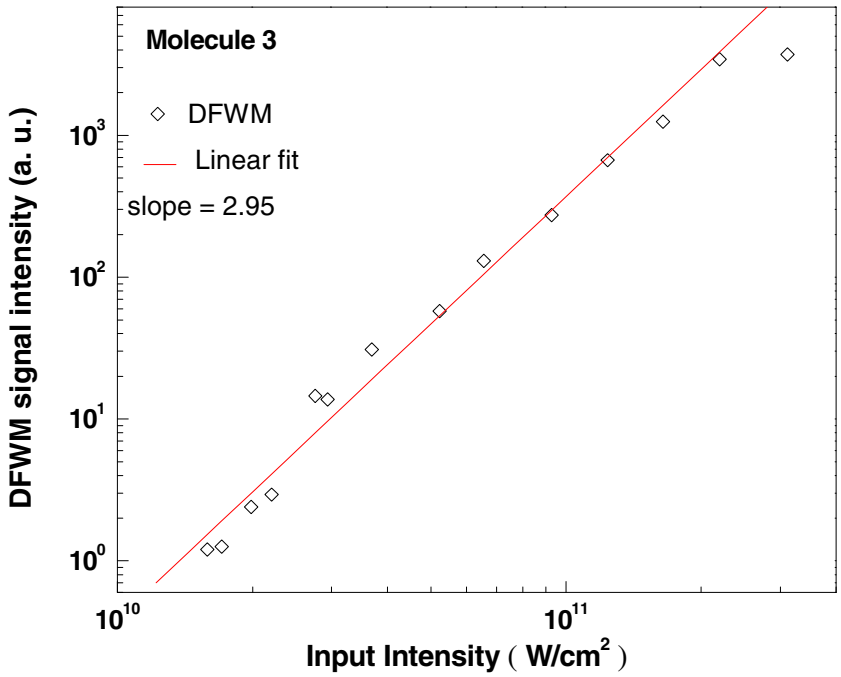
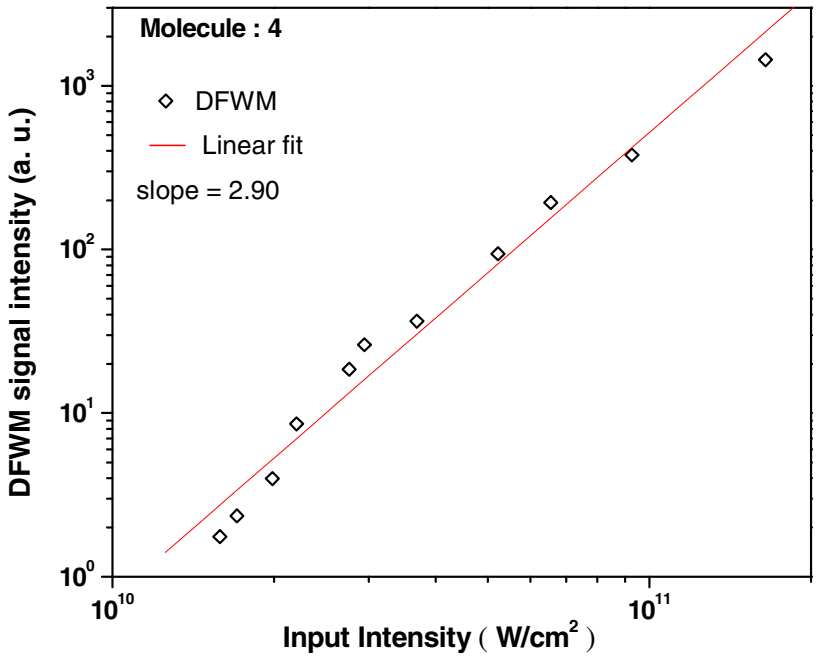
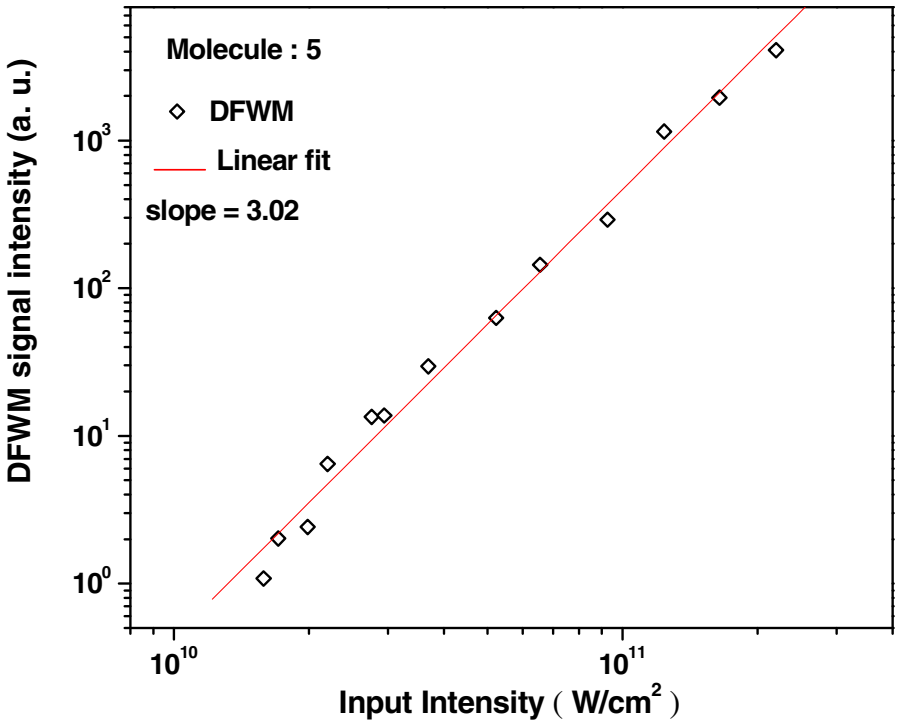

DFMW intensity (a. u.)
Delay Time (fs)
Molecule 10
Experimental data
Theoretical fit

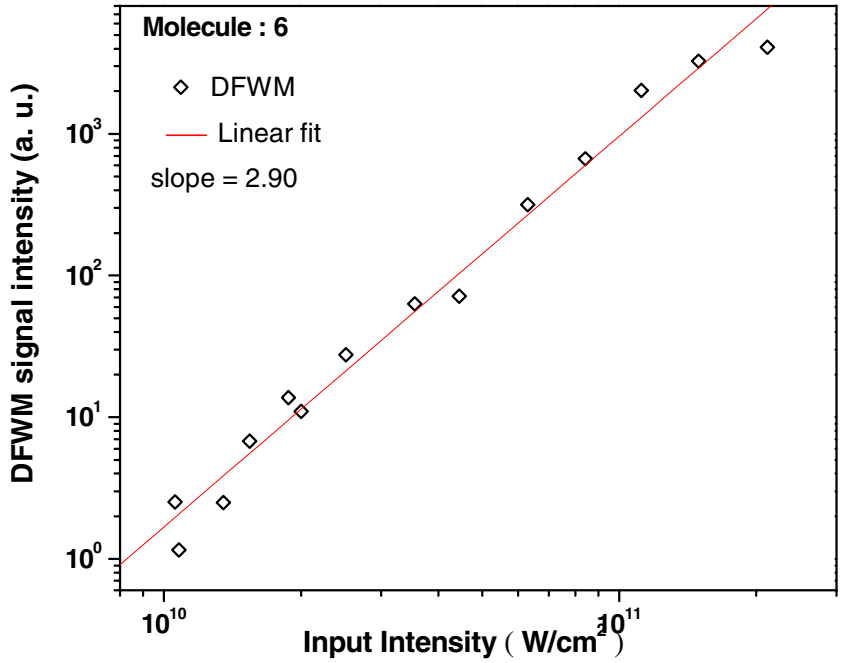
Figure C: Plots showing the cubic dependence of DFWM as a function of input intensity for molecules 2-10.

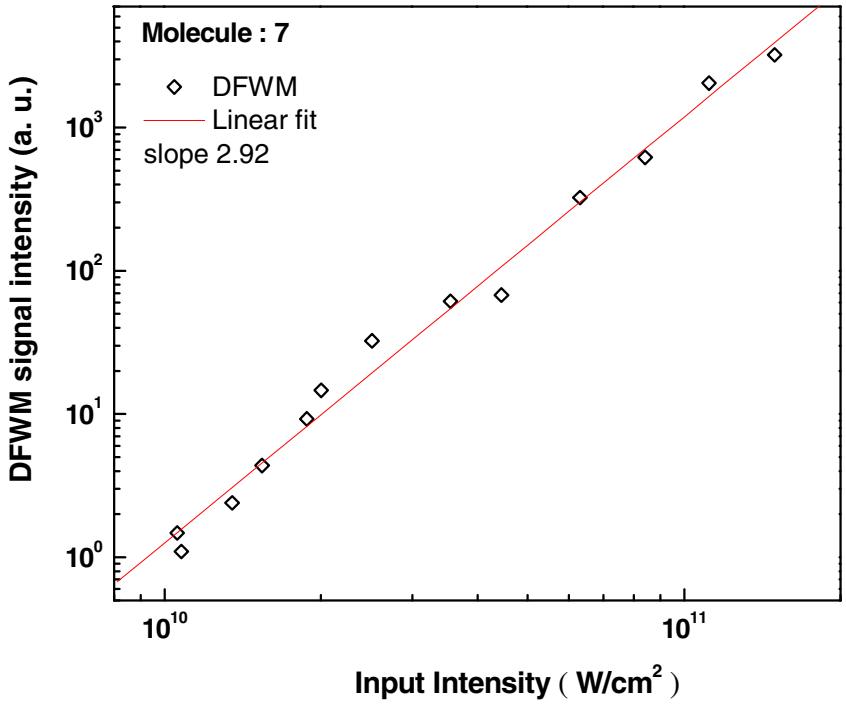


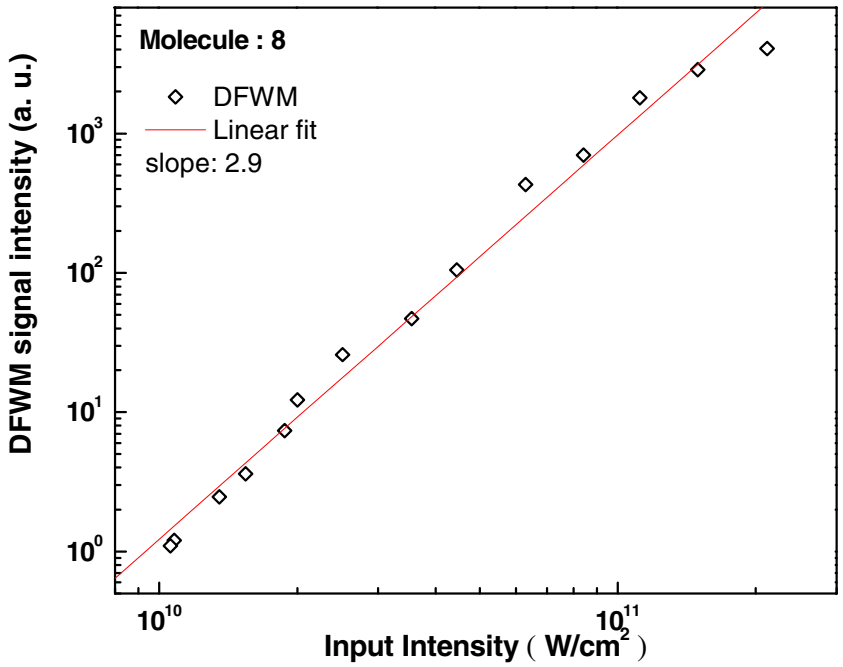


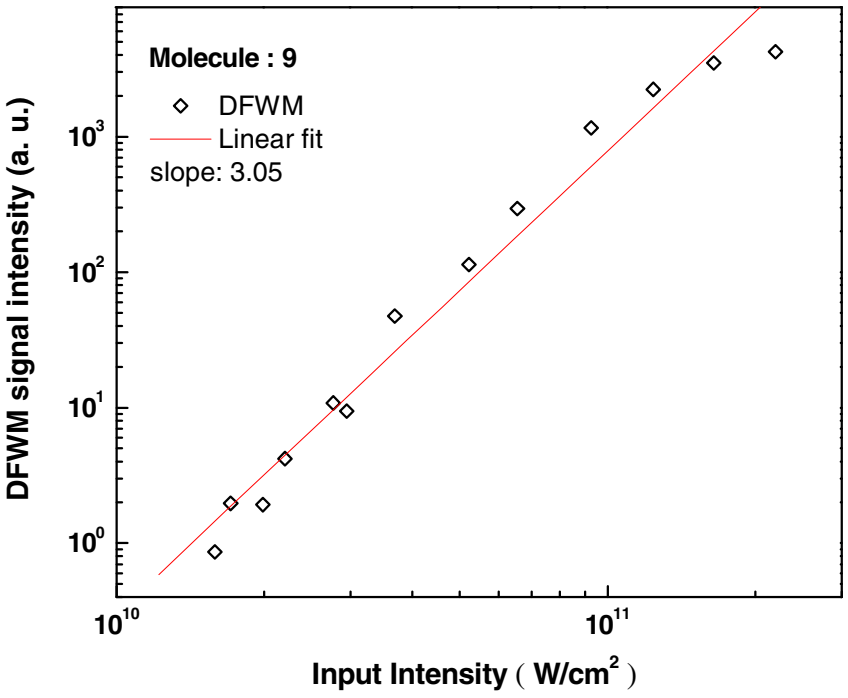

Molecule : 4
DFWM
Linear fit
slope = 2.90
DFWM signal intensity (a. u.)
Input Intensity (W/cm2)


Molecule : 5
DFWM
Linear fit
slope = 3.02
DFWM signal intensity (a. u.)
Input Intensity (W/cm2)


Molecule : 6
DFWM
Linear fit
slope = 2.90
DFWM signal intensity (a. u.)
Input Intensity (W/cm2)


Molecule : 7
DFWM
Linear fit
slope 2.92
DFWM signal intensity (a. u.)
Input Intensity (W/cm2)


Molecule : 8
DFWM
Linear fit
slope: 2.9
DFWM signal intensity (a. u.)
Input Intensity (W/cm2)


Molecule : 9
DFWM
Linear fit
slope: 3.05
DFWM signal intensity (a. u.)
Input Intensity (W/cm2)

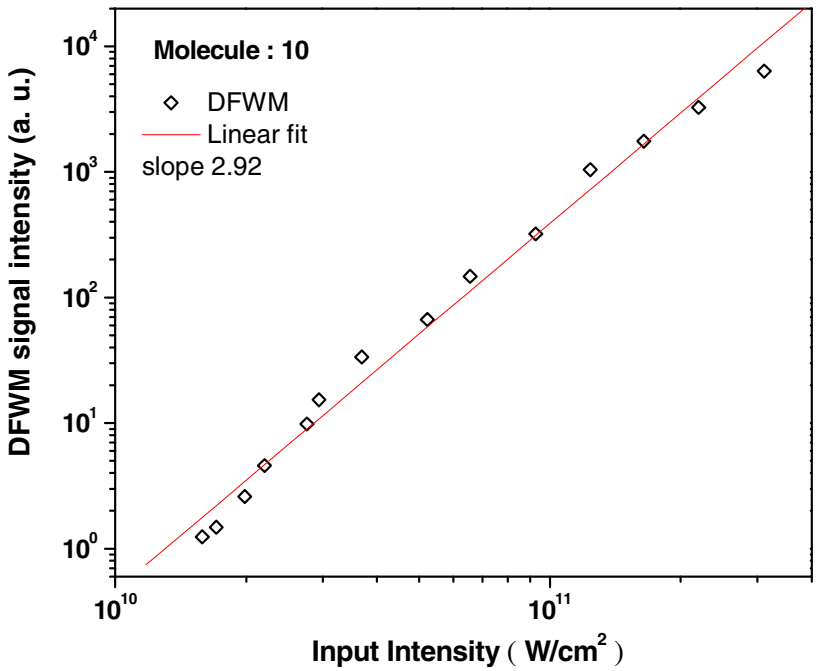
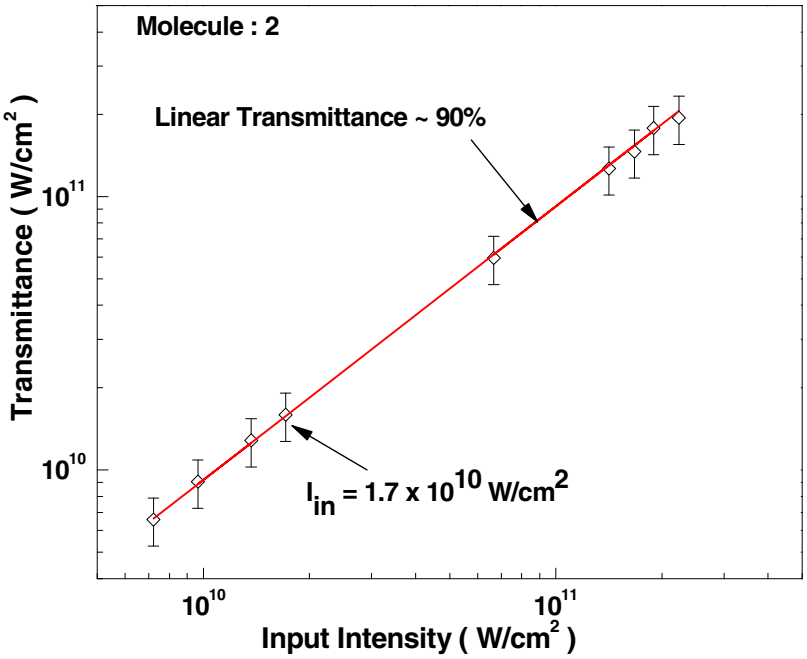
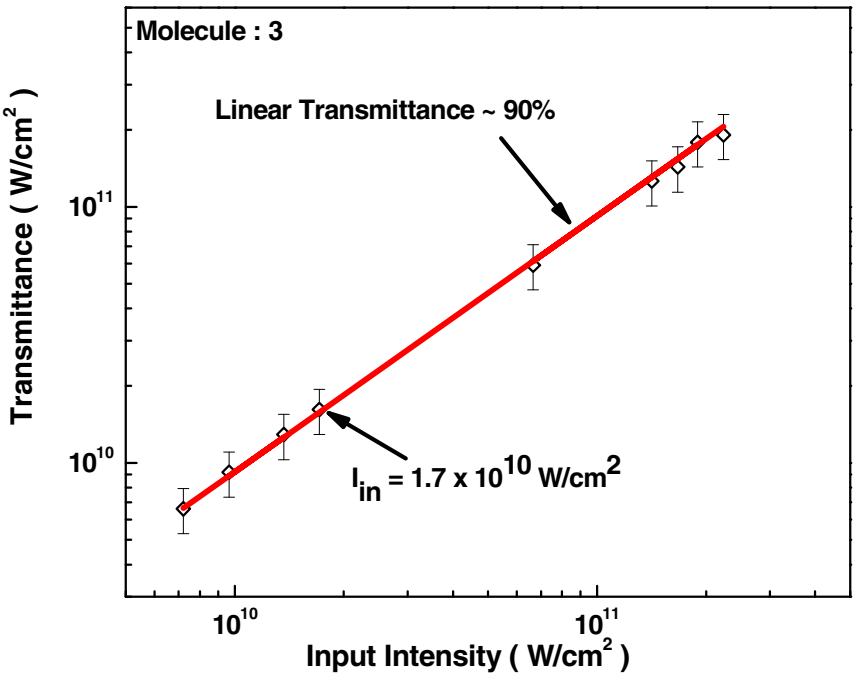
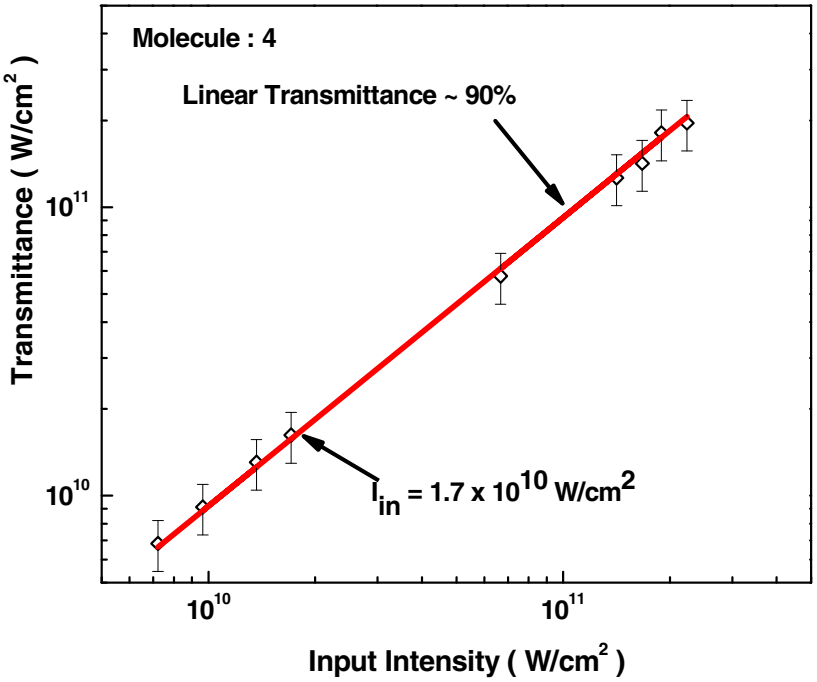
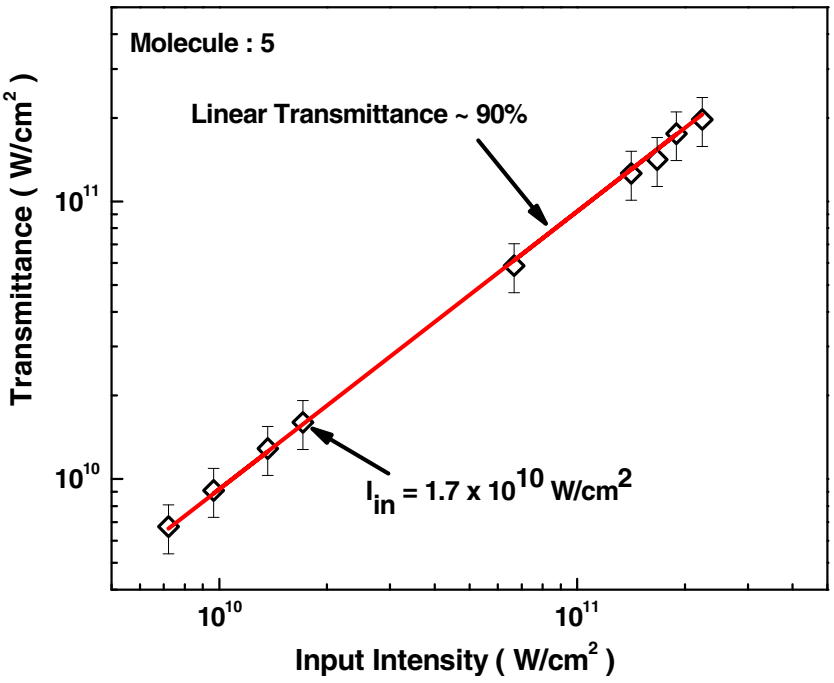


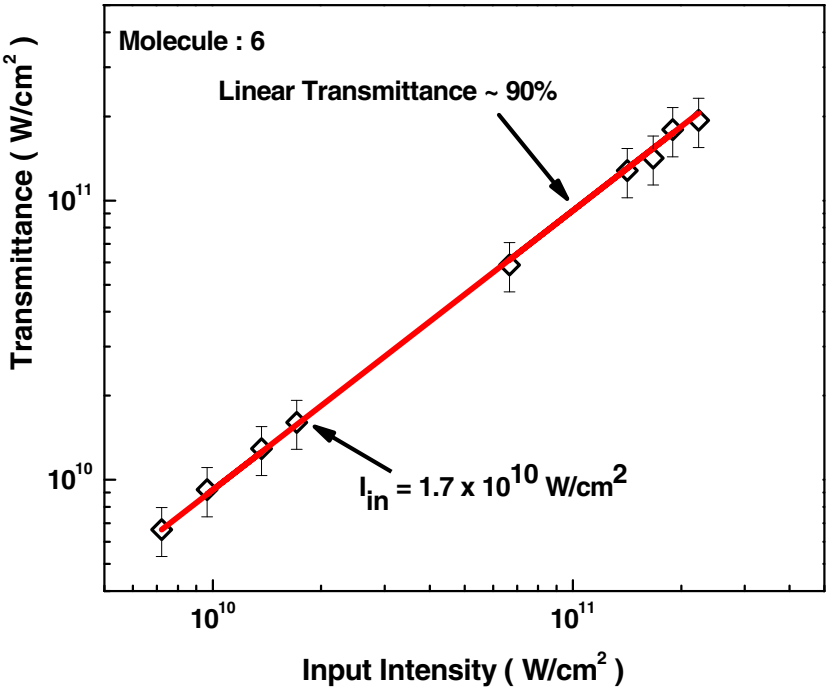
Figure D: Plot of output transmittance vs input power for the molecules 2- 10.

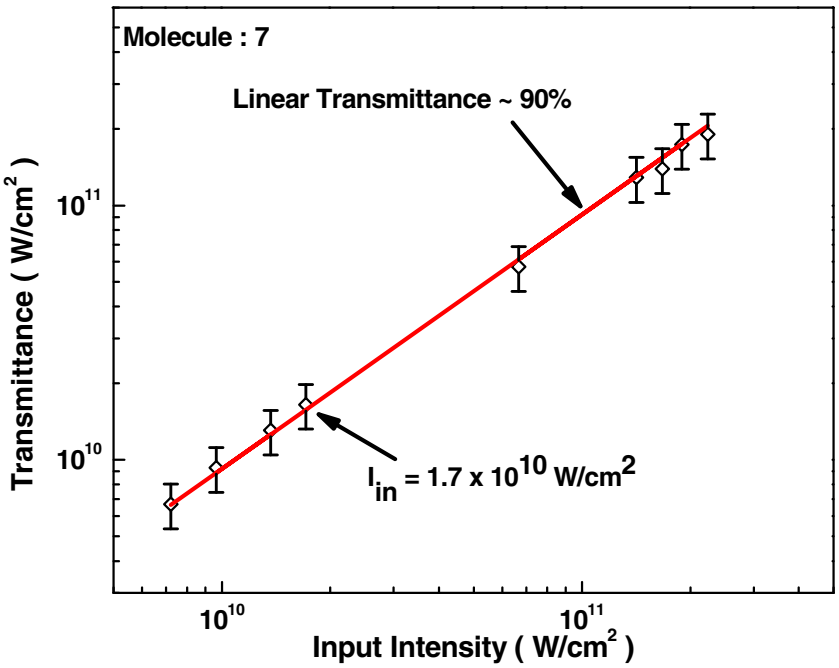


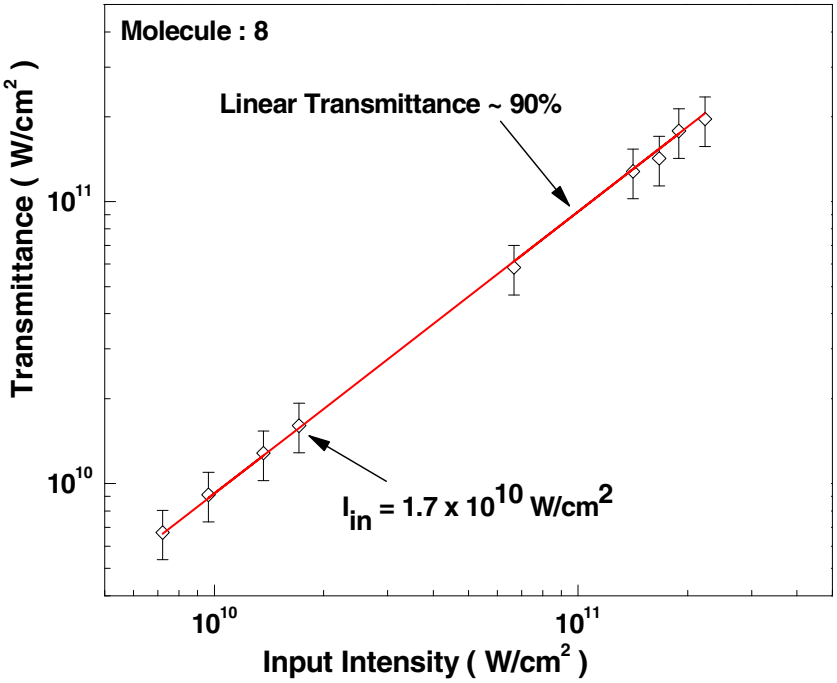

Molecule : 3
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}

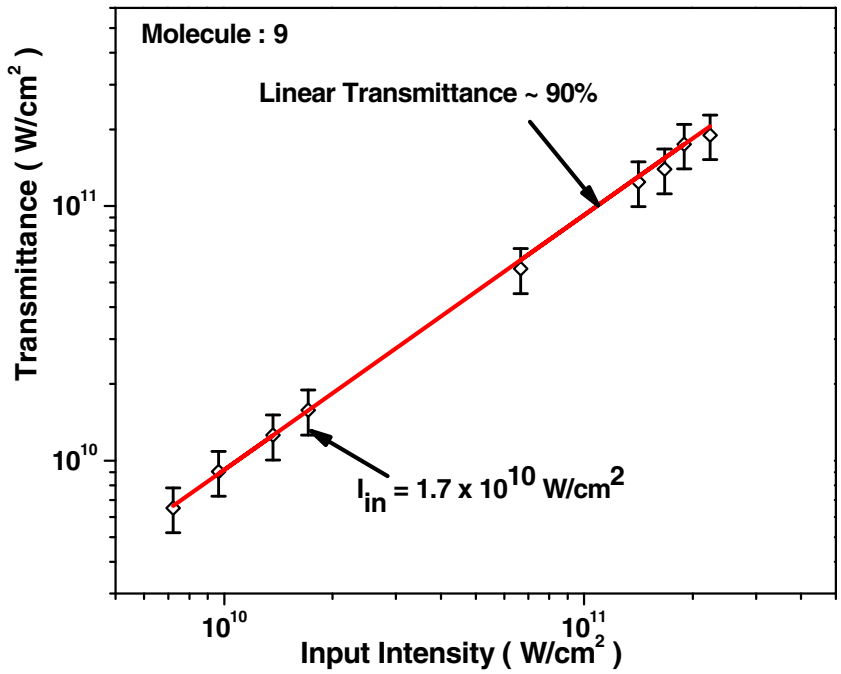

Molecule : 4
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}


Molecule : 5
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}


Molecule : 6
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}


Molecule : 7
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}


Molecule : 8
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}


Molecule : 9
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}

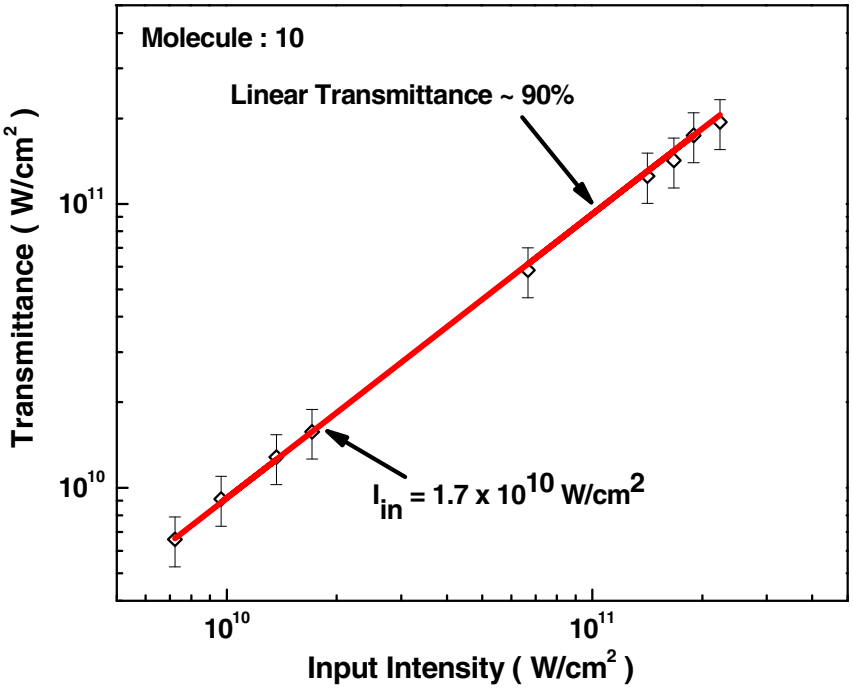
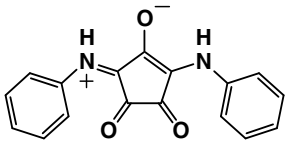
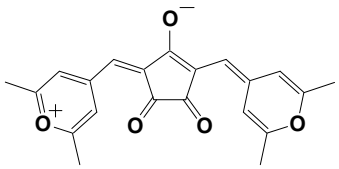

Molecule : 10
Linear Transmittance ~ 90%
I_{in} = 1.7 x 10^{10} W/cm^2
Transmittance (W/cm^2)
Input Intensity (W/cm^2)
10^{10}
10^{11}

Table A: Calculated absorption energies (ΔE_{cal} in eV), Oscillator strength (*f*),transition dipole moments(μ_{ge} in Debye) of all Molecules with Cs Symmetry in different media obtained at B3LYP/6-31G(d,p) level.

Molecule	$CHCl_3$				DMF			
	ΔE_{cal}	*f*	μ_{ge}^{x}	μ_{ge}^{y}	ΔE_{cal}	*f*	μ_{ge}^{x}	μ_{ge}^{y}
1	2.42	1.042	-8.00	7.05	2.46	1.028	-7.84	6.99
2	2.36	1.153	-5.10	10.13	2.40	1.141	-4.97	10.03
3	2.36	1.239	-0.91	4.54	2.39	1.234	-0.87	4.50
4	2.25	1.200	-0.70	11.84	2.28	1.177	-0.60	11.64
5	2.39	1.057	-5.31	9.39	2.44	1.041	-5.17	9.27
6	2.33	1.240	5.56	10.45	2.38	1.224	5.51	10.27
7	2.31	1.055	-8.89	6.42	2.34	1.026	-8.71	6.32
8	2.37	1.012	0.20	10.60	2.41	0.978	0.14	10.35
9	2.27	1.156	-5.28	10.32	2.30	1.127	-5.11	10.17
10	2.22	1.225	2.05	11.88	2.24	1.218	2.09	11.79

Table B: Details of the SAC/ SAC- CI calculations carried out using the B3LYP/ 6-31G (d, p) optimized geometries, Excitation energy (ΔE in eV), oscillator strength (*f*) and ground to excited state transition dipole moment (μ_{ge} in Debye) of Croconates absorbing in Visible and NIR Regions.

Structure[a]	ΔE	*f*	μ_{ge}[b]	Active Space (M, N)[c]	SAC		SAC-CI	
					No. of Config.[d]	Linked operators[e]	No. of Config.[d]	Linked operators[e]
	2.48	1.15	11.08	(41, 120)	24	3143061/ 44531	18	3140511/ 86447
	1.58	1.67	16.73	(41, 120)	16	3047451/ 30952	18	3044979/ 75599

[a] Both molecules are in C_{2v} Symmetry.
[b] Transition dipole moment is aligned along the y-axis.
[c] M= number of occupied orbitals, N= number of virtual orbitals.
[d] Number of configurations that have CI Coefficients larger than 0.03.
[e] Linked operators total generated/ selected.