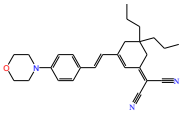
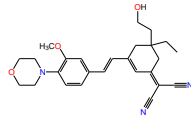
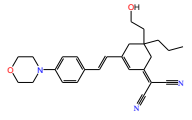
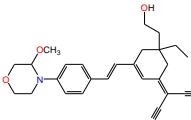
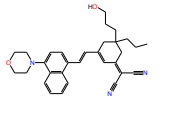
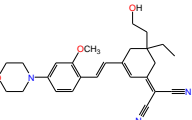
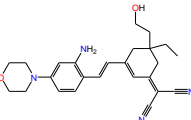
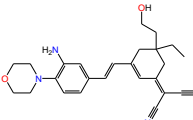
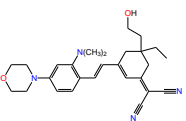
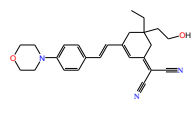
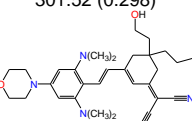
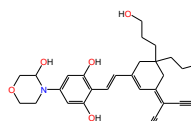
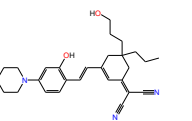
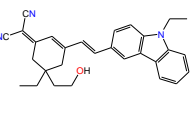
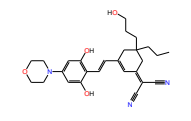
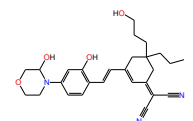
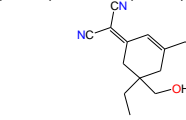
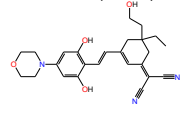
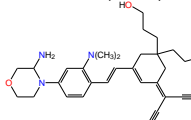
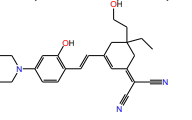
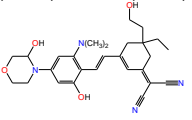
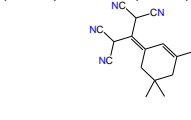
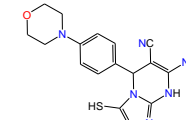
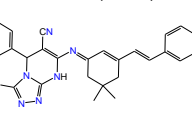
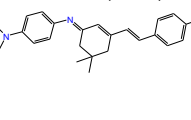
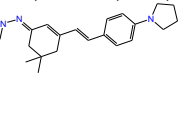
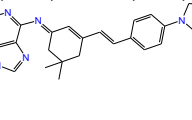
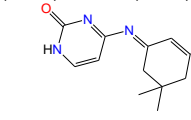
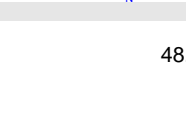
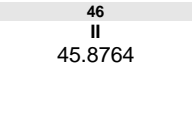
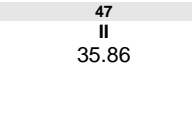
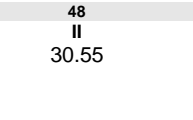
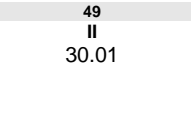
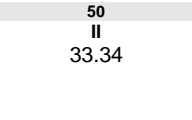
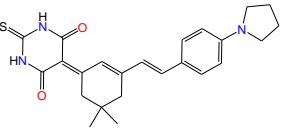
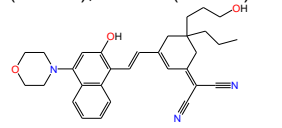
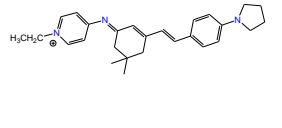
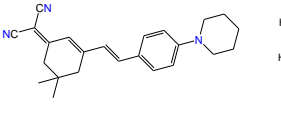
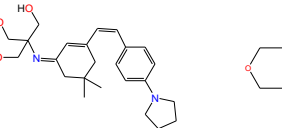
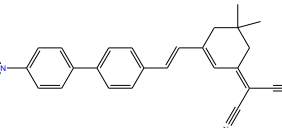
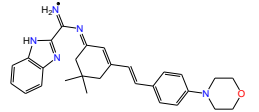
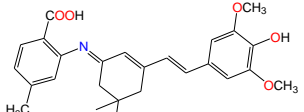
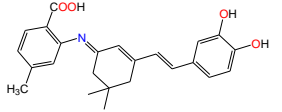
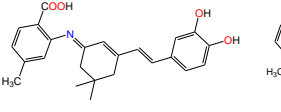
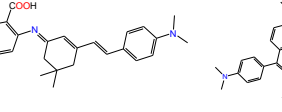
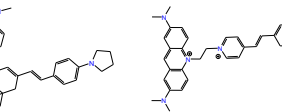
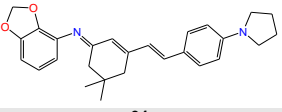
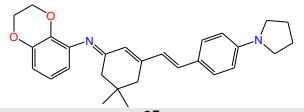
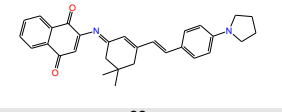
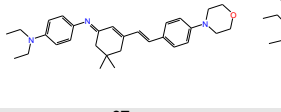
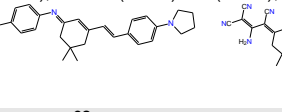
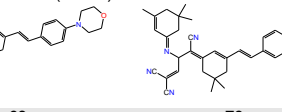
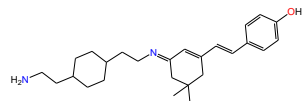


SUPPORTING INFORMATION

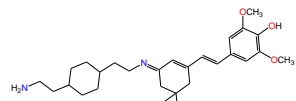
**Table S1.** Theoretical dipole moments ( $\mu_{\text{tot}}$ ), static polarizabilities ( $\alpha_{ij}$ ) and first hyperpolarizabilities ( $\beta_{ijk}$ ) ( $10^{-30} \text{ cm}^5 \text{ esu}^{-1}$ ) as well as electronic absorption data ( $\lambda$ , nm (f- oscillator strength)) depicted dyes and their forms depending the pH at M06-2X theoretical level

												
	<b>23</b>	<b>24</b>	<b>25</b>	<b>26</b>	<b>27</b>	<b>28</b>	<b>29</b>	<b>30</b>				
$\mu_{\text{tot}}$	10.76	9.19	9.9	25.65	11.16	11.56	10.33	11.51	11.54			
$\alpha_{xx}$	-209.56	-219.05	-191.7	-50.07	-199.54	-263.48	-209.24	-201.51	-197.78			
$\alpha_{yy}$	-203.10	-198.37	-200.7	-157.07	-195.21	-230.04	-199.68	-189.15	-189.56			
$\alpha_{zz}$	-190.38	-196.43	-193.9	-151.88	-201.42	-205.35	-195.31	-194.48	-192.74			
$\beta$	<b>822.15</b>	<b>750.69</b>	<b>455.48</b>	<b>1545.07</b>	<b>981.56</b>	<b>931.18</b>	<b>759.10</b>	<b>777.90</b>	<b>833.24</b>			
$\lambda$ , nm (f)	I 457.42 (0.989), 325.45 (0.587), 300.73 (0.001)	480.29 (0.866), 335.44 (0.609), 321.81 (0.123)	460.62 (0.984), 326.46 (0.583), 302.12 (0.003)	385.14 (1.147), 321.40 (0.003), 301.52 (0.298)	450.00 (1.084), 323.02 (0.509), 307.76 (0.001)	508.44 (0.750), 351.03 (0.447), 321.93 (0.138)	456.66 (0.971), 327.77 (0.537), 314.63 (0.046)	484.35 (0.980), 414.44 (0.077), 327.02 (0.505)	569.37 (0.414), 397.05 (0.647), 333.29 (0.486)			
												
	<b>31</b>	<b>32</b>	<b>33</b>	<b>34</b>	<b>35</b>	<b>36</b>	<b>37</b>	<b>38</b>				
$\mu_{\text{tot}}$	11.58	9.92	9.25	7.88	9.82	9.44	8.82	10.19				
$\alpha_{xx}$	-225.22	-190.27	250.09	-220.54	-240.46	-206.82	-248.80	-209.69				
$\alpha_{yy}$	-203.67	-189.17	-216.39	-222.43	-218.51	-198.69	-228.17	-211.32				
$\alpha_{zz}$	-202.76	-187.34	-221.24	-217.07	-188.33	-203.11	-191.28	-214.21				
$\beta$	<b>780.15</b>	<b>684.01</b>	<b>676.76</b>	<b>761.87</b>	<b>834.56</b>	<b>821.30</b>	<b>779.270</b>	<b>830.65</b>				
$\lambda$ , nm (f)	I 498.00 (0.686), 426.02 (0.179), 327.62 (0.620)	460.82 (0.988), 326.52 (0.585), 302.09 (0.003)	596.20 (0.015), 561.31 (0.341), 428.48 (0.005)	489.43 (0.665), 452.75 (0.006), 332.76 (0.518)	476.39 (0.910), 368.29 (0.033), 327.01 (0.583)	449.28 (0.941), 388.18 (0.093), 334.58 (0.533)	513.17 (0.637), 443.99 (0.004), 341.31 (0.533)	453.85 (0.834), 372.96 (0.001), 318.74 (0.627)				
												
	<b>39</b>	<b>40</b>	<b>41</b>	<b>42</b>	<b>43</b>	<b>44</b>	<b>45</b>	<b>46</b>	<b>47</b>	<b>48</b>	<b>49</b>	<b>50</b>
$\mu_{\text{tot}}$	11.54	8.35	10.26	7.64	10.66	4.41	48.5963	45.8764	35.86	30.55	30.01	33.34
$\alpha_{xx}$	-168.33	-209.58	-227.66	-120.36	-223.18	-202.44						
$\alpha_{yy}$	-186.78	-208.06	-225.04	-102.35	-215.26	-199.11						
$\alpha_{zz}$	-161.98	-191.89	-220.56	-117.41	-212.32	-205.00						
$\beta$	<b>976.79</b>	<b>649.83</b>	<b>905.728</b>	<b>420.04</b>	<b>847.39</b>	<b>655.12</b>						
$\lambda$ , nm (f)	I 471.38 (0.976), 327.53 (0.591), 303.17 (0.001)	504.10 (0.734), 440.60 (0.003), 341.50 (0.543)	546.31 (0.246), 466.72 (0.238), 335.17 (0.252)	405.54 (0.751), 310.65 (0.019), 298.43 (0.023)	623.24 (0.104), 541.40 (0.272), 375.49 (0.056)	465.15 (0.003), 418.22 (1.033), 367.22 (0.091)	362.12 (1.001), 315.22 (0.004), 277.13 (0.474)					
												
	<b>45</b>	<b>46</b>	<b>47</b>	<b>48</b>	<b>49</b>	<b>50</b>						
$\mu_{\text{tot}}$	48.5963	45.8764	35.86	30.55	30.01	33.34						

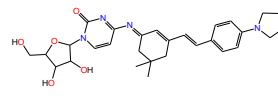
$\alpha_{xx}$	298.1404	261.6945	164.96	31.40	135.18	73.67	
$\alpha_{yy}$	-250.9422	-231.3714	-176.70	-143.85	-159.78	-146.33	
$\alpha_{zz}$	-251.3199	-263.4928	-191.67	-156.47	-178.26	-170.10	
$\beta$			<b>3060.14</b>	<b>2022.62</b>	<b>1945.09</b>	<b>2218.24</b>	
$\lambda$ , nm (f)	II 1117.76 (0.0291), 669.97 (0.0596), 585.29 (0.0004)	1196.28 (0.0349), 675.75 (0.0578), 610.48 (0.0004)	610.59 (0.174), 425.22 (0.002), 412.22 (0.001)	411.16 (0.559), 374.60 (0.022), 342.95 (0.464)	491.96 (0.075), 425.61 (0.110), 359.14 (0.950)	601.04 (0.011), 531.31 (0.003), 428.86 (0.118)	
							
	<b>51</b>	<b>52</b>	<b>53</b>	<b>54</b>	<b>55</b>	<b>56</b>	
$\mu_{tot}$	32.77	10.62	16.93	12.37	29.20	10.57	
$\alpha_{xx}$	49.14	-267.91	121.45	-183.90	80.14	-253.99	
$\alpha_{yy}$	-168.39	-240.17	-168.68	-170.97	-153.92	-198.49	
$\alpha_{zz}$	-180.51	-209.58	-168.40	-164.81	-160.12	-198.34	
$\beta$	<b>2332.29</b>	<b>890.55</b>	<b>1220.05</b>	<b>1078.11</b>	<b>1795.28</b>	<b>1882.42</b>	
$\lambda$ , nm (f)	I	II 549.59 (0.000), 425.14 (0.573), 385.58 (0.046)	549.52 (0.521), 385.90 (0.007), 344.76 (0.496)	847.46 (0.492), 537.53 (0.007), 453.47 (0.883)	470.07 (0.970), 327.90 (0.664), 304.85 (0.001)	487.19 (0.000), 413.89 (0.022), 371.05 (0.103)	521.13 (0.633), 373.99 (1.151), 317.56 (0.259)
							
	<b>57</b>	<b>58</b>	<b>59</b>	<b>60</b>	<b>61</b>	<b>62</b>	
$\mu_{tot}$	36.77	7.37	7.93	7.93	33.88	30.24	
$\alpha_{xx}$	107.28	-149.65	-121.17	-121.17	138.79	142.06	
$\alpha_{yy}$	-176.96	-167.85	-154.93	-154.93	-147.53	-186.12	
$\alpha_{zz}$	-182.52	-178.34	-160.75	-160.75	-160.74	-231.12	
$\beta$	<b>2729.05</b>	<b>539.01</b>	<b>544.75</b>	<b>544.75</b>	<b>2621.30</b>	<b>2821.85</b>	
$\lambda$ , nm (f)	I	II 388.66 (0.231), 357.25 (0.860), 319.75 (0.208)	390.28 (0.207), 351.35 (0.985), 309.10 (0.052)	390.28 (0.207), 351.35 (0.985), 309.10 (0.052)	551.05 (0.081), 391.71 (0.006), 377.74 (0.228)	684.39 (0.112), 599.39 (0.267), 491.38 (0.010)	875.72 (0.011), 815.00 (0.095), 734.29 (0.011)
							
	<b>64</b>	<b>65</b>	<b>66</b>	<b>67</b>	<b>68</b>	<b>69</b>	
$\mu_{tot}$	4.24	31.09	35.42	2.58	3.15	14.73	
$\alpha_{xx}$	-146.24	126.49	158.25	-150.54	195.83	-272.34	
$\alpha_{yy}$	-167.49	-165.61	-189.64	-195.69	-190.45	-198.18	
$\alpha_{zz}$	-189.75	-175.69	-198.79	-210.33	-205.22	-192.88	
$\beta$	<b>457.83</b>	<b>2307.85</b>	<b>2988.98</b>	<b>157.04</b>	<b>227.72</b>	<b>1111.73</b>	
$\lambda$ , nm (f)	I 398.89 (1.545), 326.81 (0.142), 312.59 (0.301)	I 392.00 (1.564), 317.87 (0.321), 305.47 (0.034)	484.06 (0.088), 460.73 (0.015), 429.21 (0.009)	I 464.04 (0.564), 372.84 (1.117), 327.88 (0.020)	I 446.13 (0.906), 380.74 (0.813), 327.79 (0.058)	I 464.48 (1.021), 381.94 (0.043), 337.35 (0.114)	I 414.66 (0.022), 401.96 (1.005), 368.11 (0.040)
	II 600.92 (0.286), 438.41 (0.003), 435.54 (0.000)	II 595.07 (0.286), 458.62 (0.005), 432.92 (0.001)		II 775.81 (0.368), 546.58 (0.005), 445.02 (0.001)	II 979.86 (0.073), 599.08 (0.001), 497.61 (0.000)	II 515.47 (0.061), 378.87 (1.177), 346.50 (0.019)	
	<b>64</b>	<b>65</b>	<b>66</b>	<b>67</b>	<b>68</b>	<b>69</b>	
$\mu_{tot}$	31.09	31.33	35.42	28.56	33.42	34.73	
$\alpha_{xx}$	126.49	161.92	158.25	152.54	195.83	-59.91	
$\alpha_{yy}$	-165.61	-171.02	-189.64	-188.52	-190.99	-189.85	
$\alpha_{zz}$	-175.69	-182.28	-198.79	-196.11	-186.95	-181.14	
$\beta$	<b>2307.85</b>	<b>2365.81</b>	<b>2988.98</b>	<b>2171.73</b>	<b>2983.72</b>	<b>2449.73</b>	
$\lambda$ , nm (f)	I 398.89 (1.545), 326.81 (0.142), 312.59 (0.301)	I 392.00 (1.564), 317.87 (0.321), 305.47 (0.034)	484.06 (0.088), 460.73 (0.015), 429.21 (0.009)	I 464.04 (0.564), 372.84 (1.117), 327.88 (0.020)	I 446.13 (0.906), 380.74 (0.813), 327.79 (0.058)	I 464.48 (1.021), 381.94 (0.043), 337.35 (0.114)	I 414.66 (0.022), 401.96 (1.005), 368.11 (0.040)
	II 600.92 (0.286), 438.41 (0.003), 435.54 (0.000)	II 595.07 (0.286), 458.62 (0.005), 432.92 (0.001)		II 775.81 (0.368), 546.58 (0.005), 445.02 (0.001)	II 979.86 (0.073), 599.08 (0.001), 497.61 (0.000)	II 515.47 (0.061), 378.87 (1.177), 346.50 (0.019)	
	<b>64</b>	<b>65</b>	<b>66</b>	<b>67</b>	<b>68</b>	<b>69</b>	



71



72



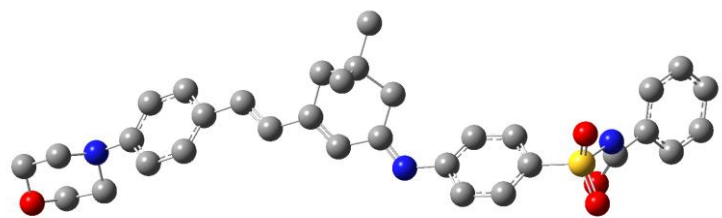
73

	I	I	II
$\mu_{\text{tot}}$	2.83	2.47	41.42
$\alpha_{xx}$	-179.37	-170.66	291.80
$\alpha_{yy}$	-189.68	-185.71	-184.50
$\alpha_{zz}$	-180.38	-192.23	-211.75
$\beta$	118.08	257.48	3885.48
$\lambda$ , nm (f)	I 377.62 (0.027), 341.07 (0.677), 316.53 (0.001)	I 360.09 (0.001), 329.94 (1.321), 302.74 (0.000)	II 496.31 (0.001), 449.13 (0.024), 422.97 (0.015)
	II -	II -	

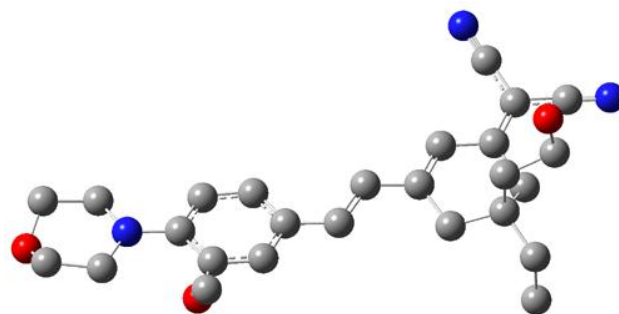
**Table S2.** The first and second hyperpolarizability tensor matrix components  $\beta_{ijk}$  and  $\gamma_{ijkl}$  of the neutral (**I**) and cationic (**II**) forms of selected CLPs

	<b>MI1</b>	<b>2</b>	<b>5</b>	<b>7</b>	<b>23</b>	<b>24</b>	<b>25</b>	<b>27</b>	<b>44</b>			
	<b>I</b>	<b>II</b>	<b>I</b>	<b>II</b>	<b>II</b>	<b>I</b>	<b>I</b>	<b>I</b>	<b>I</b>	<b>I</b>	<b>II</b>	
$\beta_{xxx}$	54.8052	610.5152	-606.4191	-1393.2530	2847.5402	-2692.8170	-522.7414	-405.3738	-354.5930	-659.8075	-220.6072	-1691.7547
$\beta_{yyy}$	-21.5501	-16.9248	-130.3137	-125.5594	-8.3689	-7.9760	-165.0629	-147.2719	-167.9664	-176.1626	50.0588	64.4043
$\beta_{zzz}$	3.4473	3.7653	-0.1429	-1.2846	-22.0800	4.4738	-2.8261	1.9687	-5.3678	101.2507	-20.6225	-20.8032
$\beta_{xyy}$	118.9248	119.7173	-180.3907	-160.8700	97.2431	-91.5368	-193.7804	-177.2183	-183.2147	-208.4198	-130.1657	-116.4687
$\beta_{xxy}$	-164.1111	-192.6273	-212.3198	-230.6835	-55.0554	-366.0233	-167.1907	-262.2051	-182.2851	-198.3136	29.8889	38.3715
$\beta_{xxz}$	-48.4661	31.5088	126.0726	70.2549	-73.8721	24.2273	168.3371	179.7021	220.7536	83.8460	152.8845	162.1634
$\beta_{xzz}$	5.0666	46.9925	-6.7447	-69.9830	70.7813	-40.9988	-12.5975	-11.3801	-2.0354	37.6839	-95.8314	-149.1928
$\beta_{yzz}$	4.0405	6.7789	-7.7977	-5.7229	-0.3595	-6.8024	-7.1956	-15.1158	-7.1638	-0.6130	51.6173	36.4577
$\beta_{yyz}$	-2.6004	-0.3708	-20.0786	-12.5830	-12.4704	-6.9076	5.0631	-6.9037	9.4926	6.2083	30.0776	31.8072
$\beta_{xyz}$	13.4836	26.6040	-8.1707	-27.5089	-6.5501	29.2778	37.4630	-2.6793	32.4111	29.0889	-10.5399	-29.9431
$\gamma_{xxxx}$	-22016.6398	-15724.4478	-31834.6594	-23085.4094	-33272.4777	-17513.5676	-33089.8670	-34224.8946	-31608.4127	-38606.2263	-36630.5247	-21894.4952
$\gamma_{yyyy}$	-1528.7084	-1374.7328	-4358.0070	-4087.0979	-2354.9643	-2185.7835	-5274.2052	-4636.4519	-5256.7752	-6749.0566	-4230.7067	-4012.4805
$\gamma_{zzzz}$	-438.5484	-572.2250	-621.3510	-766.1068	-1273.4470	-793.2580	-1628.4807	-1380.7587	-1480.9135	-1854.7061	-1303.1650	-1349.5514
$\gamma_{xxxy}$	827.523	405.4522	-1144.3569	-710.3463	-4280.3394	484.0059	-1297.8278	-1482.5752	-1405.2950	-1128.1260	-964.0127	-906.2562
$\gamma_{xxxz}$	28.7644	588.5812	-504.2731	-471.1038	1935.4706	-205.6569	-413.9749	-111.4538	177.4357	-595.5667	1280.3359	1166.0742
$\gamma_{yyyx}$	165.8909	170.5644	-640.1020	-585.0825	-183.4050	153.8756	-614.2226	-727.6594	-631.0601	-872.1555	1280.3359	198.7661
$\gamma_{yyyz}$	-19.3573	-16.5610	-94.1719	-94.9570	8.7387	12.0067	7.6016	-178.2424	1.0085	-71.9704	-105.7614	-94.7230
$\gamma_{zzzx}$	14.7546	5.5371	10.2955	17.1670	-37.8308	-35.3854	16.3239	78.4941	120.8158	514.5319	-31.3773	-70.1779
$\gamma_{zzzy}$	6.3134	1.6829	-1.5740	1.0567	-3.3359	7.2994	12.6003	-19.6017	12.3069	-3.5222	36.4337	38.2781
$\gamma_{xxyy}$	-3627.4634	-3342.9741	-5247.4609	-4929.3981	-8656.4326	-6458.1473	-5732.1123	-5777.1342	-5658.2136	-6640.7148	-6222.4440	-6132.0162
$\gamma_{xxzz}$	-3206.9052	-2856.3654	-4736.4564	-4103.2765	-8564.4426	-6693.3900	-5334.2218	-5104.0659	-5073.0492	-5667.9859	-5597.8677	-4886.6868
$\gamma_{yyzz}$	-324.4393	-306.9136	-757.2377	-728.8024	-600.0478	-503.7060	-1011.1205	-932.7078	-979.4998	-1411.8474	-1037.3146	-980.1495
$\gamma_{xxyz}$	-65.6461	27.8698	81.3301	224.0040	199.5127	116.2721	64.2192	-137.1309	21.1301	160.5276	-460.8434	-312.0022
$\gamma_{yyxz}$	41.4203	59.6334	-60.2952	-101.9698	-53.1406	-43.4614	53.9024	-116.7058	81.5243	53.6440	160.9147	173.4996
$\gamma_{zzxy}$	-6.9534	8.715	-49.2842	-52.5506	-25.7638	37.2673	-52.1163	-59.9825	-62.9689	-95.6225	238.9521	243.2312
	<b>28</b>	<b>36</b>	<b>15</b>	<b>45</b>	<b>46</b>	<b>19</b>	<b>6</b>	<b>47</b>	<b>48</b>	<b>50</b>	<b>21</b>	<b>67</b>
	<b>I</b>	<b>I</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>I</b>
$\beta_{xxx}$	-417.5403	-661.8924	-5573.8837	-5647.5960	-4975.0683	-5143.6295	-6370.2878	-2884.3242	-1897.7623	-2075.4465	3787.8489	-165.5295
$\beta_{yyy}$	-167.6391	-127.5810	129.4475	122.1247	173.4532	93.3795	8.8585	-10.8539	-7.9517	-43.0530	-36.2571	-10.3948
$\beta_{zzz}$	6.1895	6.6831	-80.9435	-76.5707	-64.9466	19.8726	-12.4688	5.2060	5.7798	3.0622	12.1212	3.5926
$\beta_{xyy}$	-185.1198	-144.8026	-103.8172	-251.3199	-61.1699	-139.3190	-500.7378	-93.3337	-82.3144	-19.1820	223.0094	87.6663
$\beta_{xxy}$	-226.6137	-150.1064	1131.3505	1097.8970	867.3061	1125.6378	-665.3174	-277.7148	5.3610	-428.2889	-907.4184	8.3642
$\beta_{xxz}$	192.0227	98.2846	10.5728	132.7237	33.0681	7.4101	196.8042	-46.2561	-20.8440	-157.0485	33.3848	-127.1832
$\beta_{xzz}$	-9.6294	39.5221	-169.2110	-176.3662	-139.9354	-148.5101	-134.6094	-68.1249	-42.5042	-64.2926	99.3123	-28.6906
$\beta_{yzz}$	-13.6486	-2.0276	23.3776	14.4993	-0.5306	13.9580	18.1399	-2.7888	-5.3340	-8.5924	-8.5785	1.3825
$\beta_{yyz}$	-11.2438	-4.9929	60.9210	124.1029	16.9815	-7.0434	96.7461	-11.4139	4.4391	-17.1797	2.8288	10.0068
$\beta_{xyz}$	-6.2682	-6.4295	-162.9971	-67.8813	-168.5815	46.0460	-154.2109	40.1115	-2.3162	5.4592	45.7960	36.5264
$\gamma_{xxxx}$	-32341.1408	-31431.5857	31.6689	1209.7869	-1505.1650	-8308.3755	553.7978	-13592.1005	-17276.4630	-15075.0902	-14709.5827	-51226.8796
$\gamma_{yyyy}$	-4861.6470	-5033.4413	-9733.9648	-11370.4418	-7709.5114	-4777.7565	-12524.3513	-2037.4861	-1868.4967	-1851.5501	-7583.8707	-2609.5031

$\gamma_{zzzz}$	-1401.4075	-1378.0339	-2635.6570	-2664.1197	-2315.5959	-3149.3649	-1816.6543	-786.0118	-619.6679	-684.0594	-1486.1237	-760.9730
$\gamma_{xxxY}$	-1481.0943	-2004.3544	-8488.2029	-11507.6480	-5929.2100	-3848.8867	10679.2099	1109.5188	1824.0951	-1490.5356	-3658.8010	377.4022
$\gamma_{xxxZ}$	272.2245	874.8771	-597.5672	-722.6006	-1023.4229	5755.2523	4441.0689	-1439.1997	-548.3824	-1754.9972	987.2501	-1896.9647
$\gamma_{YYXX}$	-760.5627	-712.9163	-81.1265	-699.4031	583.9978	114.3336	1135.0889	95.0344	102.4299	-155.4824	-256.1990	-130.9225
$\gamma_{YYYZ}$	27.8827	-80.9170	-330.0415	55.5503	-271.1889	-16.0358	-95.7855	39.4740	-11.3876	59.2606	-39.9238	-29.4291
$\gamma_{zzzX}$	118.5764	124.6902	-121.0996	-132.6802	-157.5596	396.5098	65.2280	-79.6148	-12.2193	-60.8437	48.3189	42.9175
$\gamma_{zzzY}$	-7.5727	-13.8240	-69.7377	-24.9486	-79.1078	26.3721	-18.5080	-2.3172	-16.8031	-3.3082	10.3435	-17.4894
$\gamma_{XXYY}$	-5512.9760	-5787.1009	-9962.9121	-11093.2153	-9339.9805	-10363.3684	-12578.1357	-7516.6497	-4656.0185	-4837.7891	-8368.3352	-8901.7588
$\gamma_{XXZZ}$	-4759.0966	-5188.9083	-11326.6296	-11738.3894	-10227.7024	-11070.6698	-11940.4503	-7882.1388	-4403.2330	-5331.7769	-7594.4513	-9756.0918
$\gamma_{YYZZ}$	-934.3012	-1013.9682	-2164.9218	-2499.0680	-1770.0948	-1251.1401	-2203.6849	-475.5539	-414.3434	-451.0523	-1540.3604	-559.5206
$\gamma_{XXYZ}$	10.2703	-136.8999	-320.5768	535.4014	-537.5433	-137.6485	-1460.2455	240.5123	3.0273	-46.0451	-124.0322	-116.0684
$\gamma_{YYXZ}$	77.5498	-4.0123	201.8777	686.9506	-8.6074	157.4473	932.5090	-84.6653	22.7609	-135.8039	83.7917	-15.3772
$\gamma_{ZZXY}$	-59.8805	-124.8861	-46.4417	-160.2926	-21.3750	-94.7387	213.9848	38.1265	15.4650	-16.1302	-84.8234	-11.5633
	<b>71</b>	<b>57</b>	<b>72</b>	<b>61</b>	<b>9</b>	<b>73</b>	<b>17</b>					
	<b>I</b>	<b>II</b>	<b>I</b>	<b>II</b>	<b>II</b>	<b>II</b>	<b>II</b>					
$\beta_{xxx}$	41.0031	-3099.8141	-120.8949	-2457.2148	-3100.1232	-3630.5003	-5906.5702					
$\beta_{YYY}$	-31.6055	15.4417	-0.6854	19.5536	26.1410	-65.5234	-148.0418					
$\beta_{zzz}$	-4.0059	34.2781	11.7134	8.3688	17.7926	-61.4698	-29.4259					
$\beta_{XYY}$	-9.8554	-161.4473	-146.8657	-108.3508	-137.9749	-17.7553	-187.0866					
$\beta_{XXY}$	133.5494	-303.7441	93.6441	39.8090	64.6182	-778.6783	-729.2995					
$\beta_{XXZ}$	-37.3657	291.2873	44.4346	-47.4977	-45.3391	-488.5023	-30.9100					
$\beta_{XZZ}$	11.3605	-53.0768	30.9343	-54.5021	-16.1726	-100.5623	-143.6357					
$\beta_{YZZ}$	-1.3907	-0.0657	-11.3882	9.7892	-7.5114	-5.1939	14.7066					
$\beta_{YYZ}$	-3.6234	8.2709	3.4978	-1.9229	9.7005	-17.3158	38.3448					
$\beta_{XYZ}$	52.9372	-8.1783	84.5172	11.2396	1.4080	86.4103	-78.1040					
$\gamma_{xxxx}$	-14101.9263	-17999.1350	-45148.9267	-4572.8058	-31376.6385	-3575.0324	-1636.2118					
$\gamma_{YYYY}$	-8305.3461	-2306.4367	-2440.086	-2413.5276	-3347.5301	-2399.3011	-10090.1017					
$\gamma_{zzzz}$	-788.4627	-890.6357	-915.4168	-525.7496	-1496.1230	-1396.4550	-2124.4106					
$\gamma_{xxxY}$	600.9865	3333.5423	4459.8921	-2689.9689	1769.3357	3047.3541	5750.0872					
$\gamma_{xxxZ}$	-469.5581	226.8476	503.2033	280.9261	1218.0570	1196.5376	2990.5075					
$\gamma_{YYXX}$	-302.6290	407.4961	49.6781	-213.2160	470.3646	-196.9320	-466.7542					
$\gamma_{YYYZ}$	-25.1290	13.6092	-31.4137	-23.1572	2.2450	54.3539	254.4327					
$\gamma_{zzzX}$	1.6243	97.8635	60.1512	22.6827	-5.7082	-123.2605	113.3853					
$\gamma_{zzzY}$	-2.3689	-2.9568	-8.0670	10.6218	35.5044	42.0194	94.6860					
$\gamma_{XXYY}$	-3924.3060	-7892.4932	-8503.867	-4783.9668	-8881.7237	-8270.6295	-10730.4493					
$\gamma_{XXZZ}$	-2475.5813	-7873.7138	-8138.8052	-4501.8010	-8929.1155	-9803.1819	-10897.9630					
$\gamma_{YYZZ}$	-1452.9623	-542.7258	-601.4417	-501.6368	-775.3512	-656.5400	-2135.3823					
$\gamma_{XXYZ}$	191.3883	85.7887	-507.5071	-58.3612	286.9495	614.3745	128.8387					
$\gamma_{YYXZ}$	35.1281	-59.8102	-58.2441	-51.9944	-177.9922	-49.1723	445.9233					
$\gamma_{ZZXY}$	4.4116	153.9106	41.5964	-27.2386	79.9433	60.0349	85.2829					

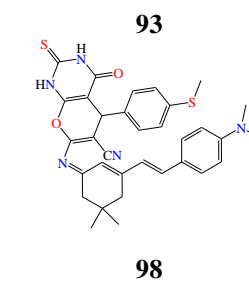
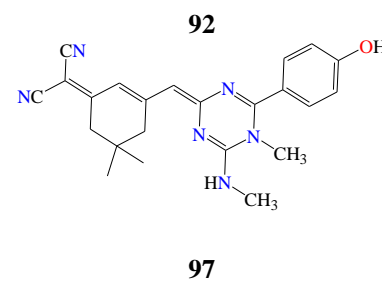
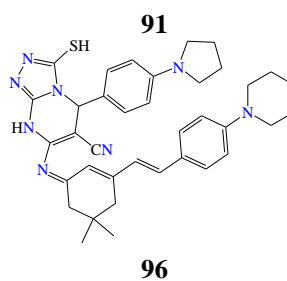
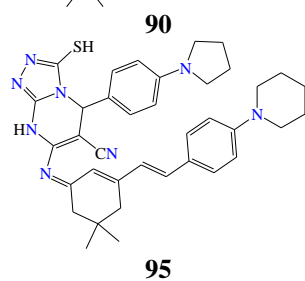
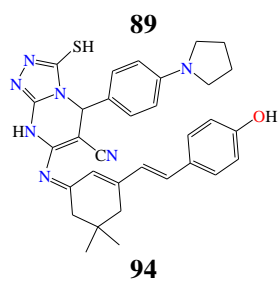
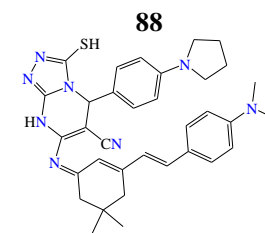
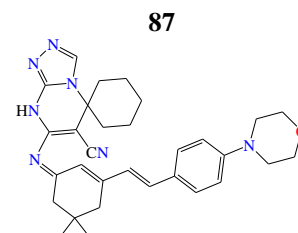
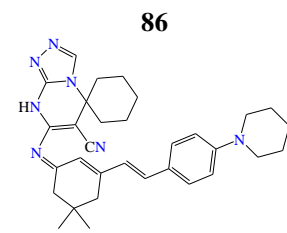
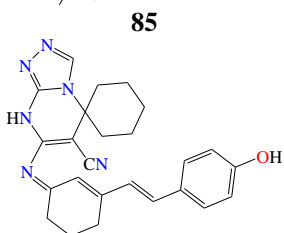
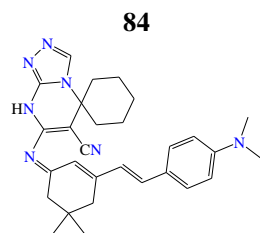
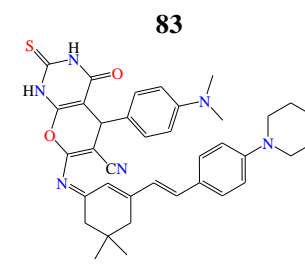
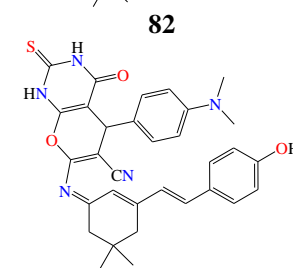
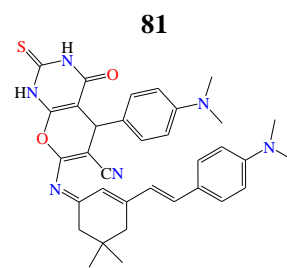
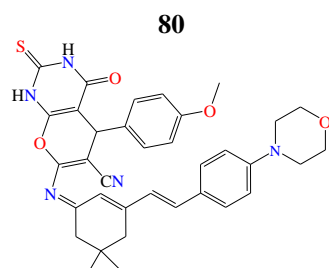
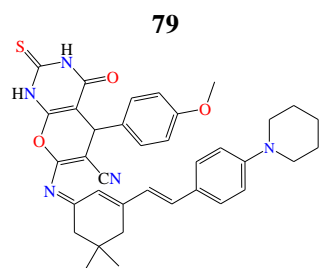
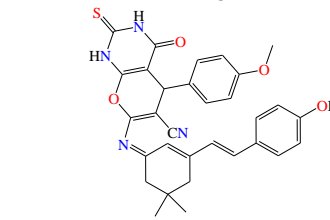
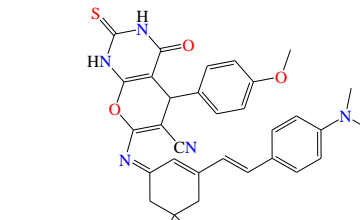
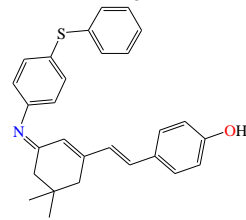
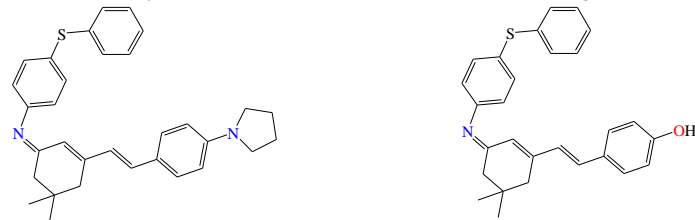
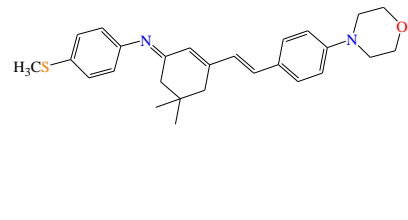
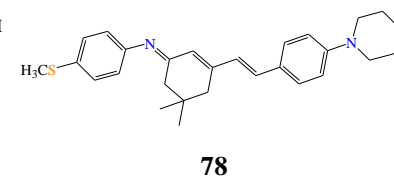
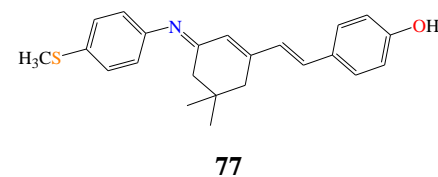
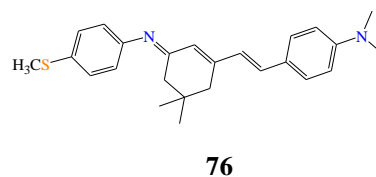
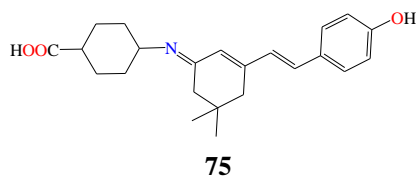
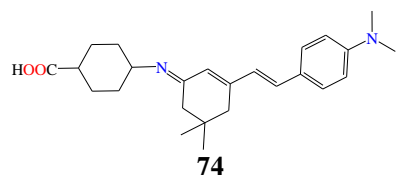


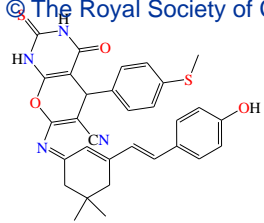
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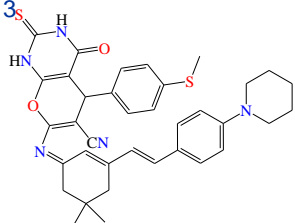
24

**Scheme S1.** Optimized molecular geometries of selected CLPs; Hydrogens are omitted.

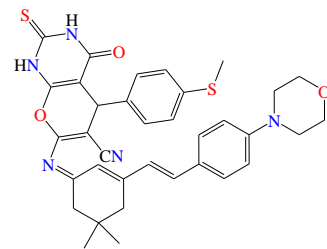




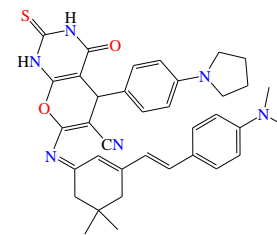
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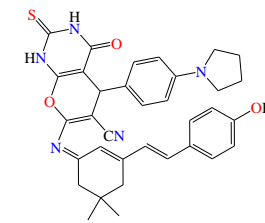
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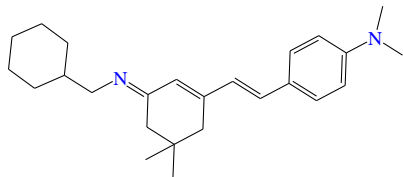
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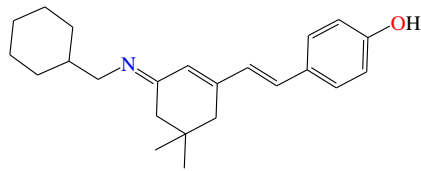
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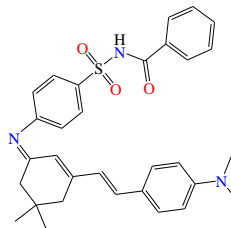
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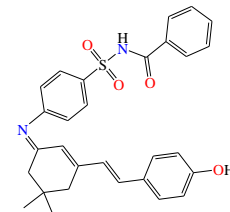
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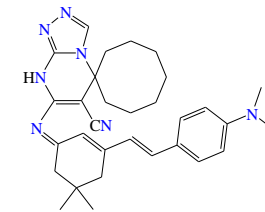
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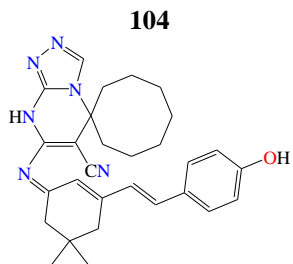
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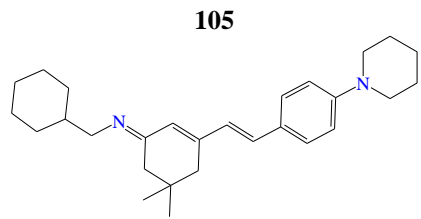
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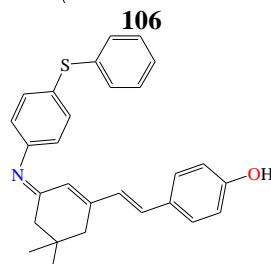
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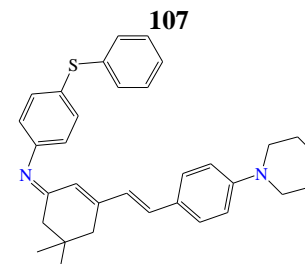
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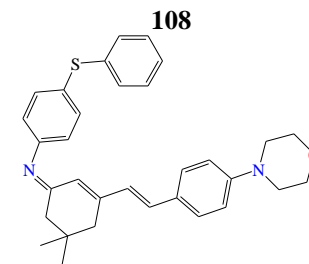
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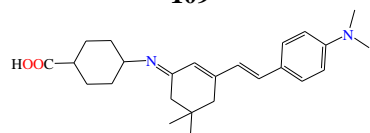
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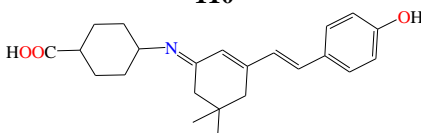
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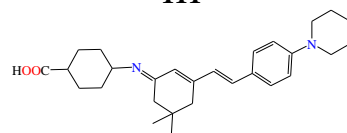
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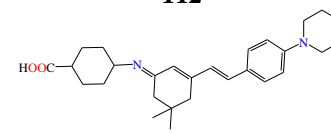
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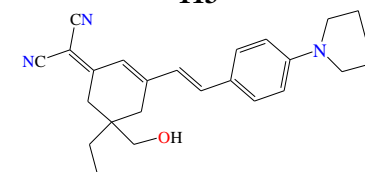
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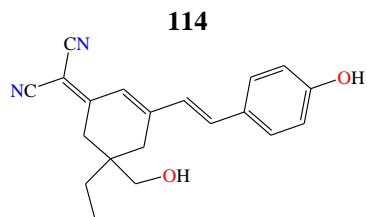
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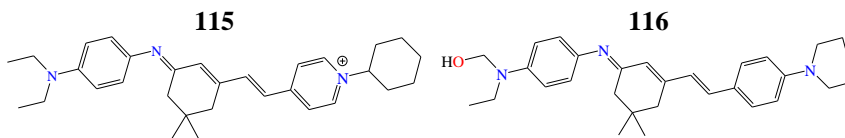
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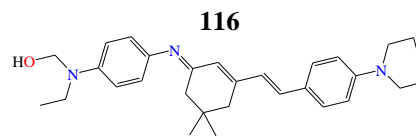
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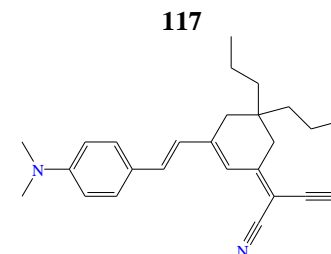
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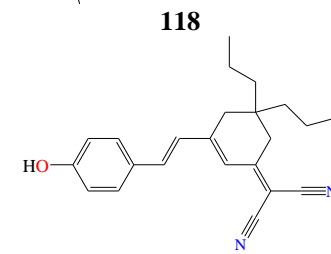
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121

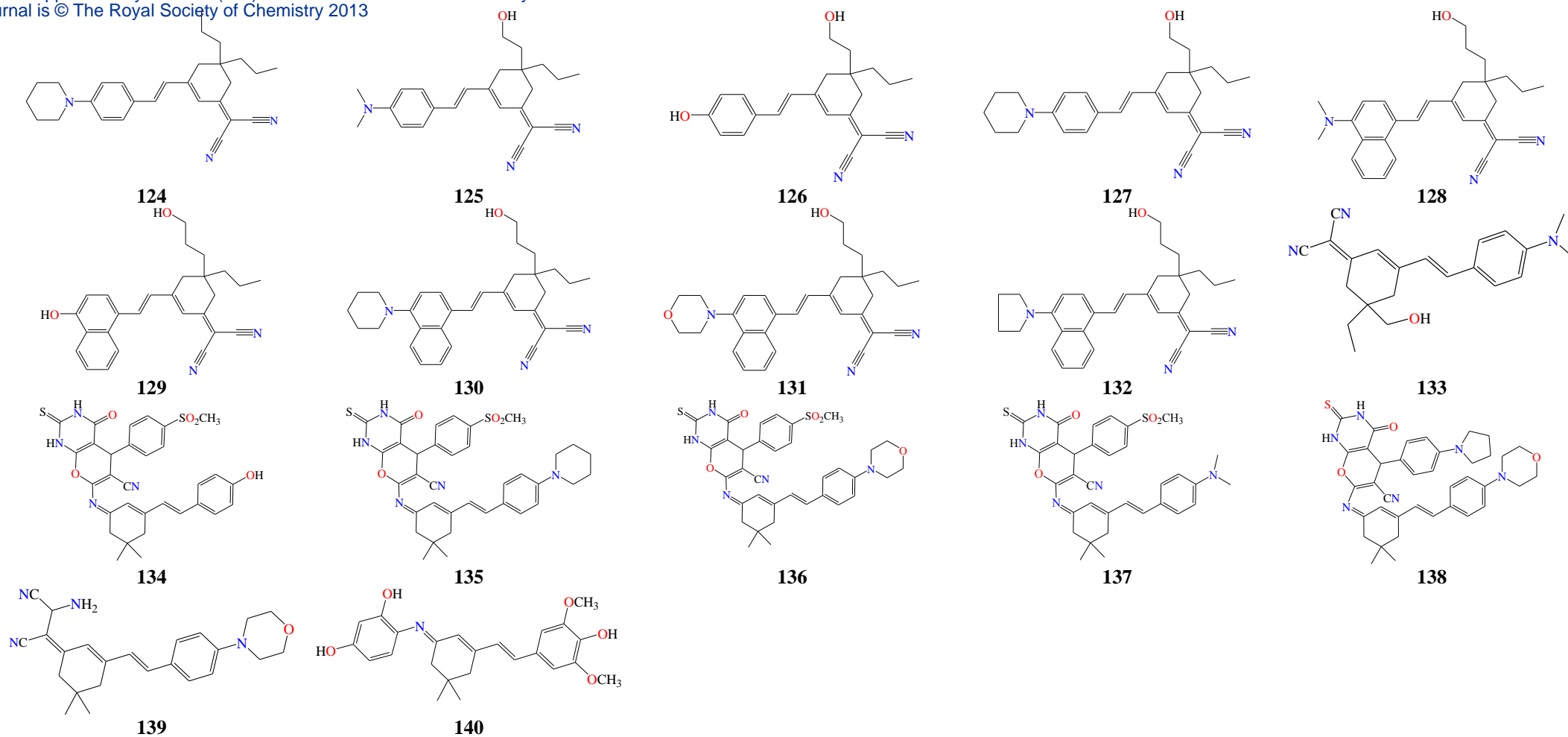


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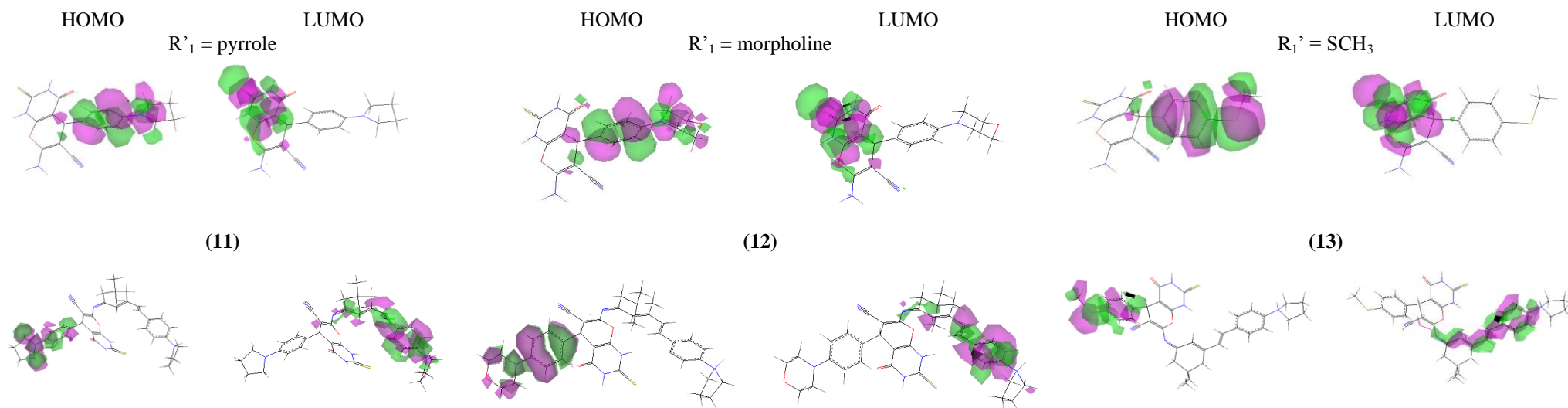


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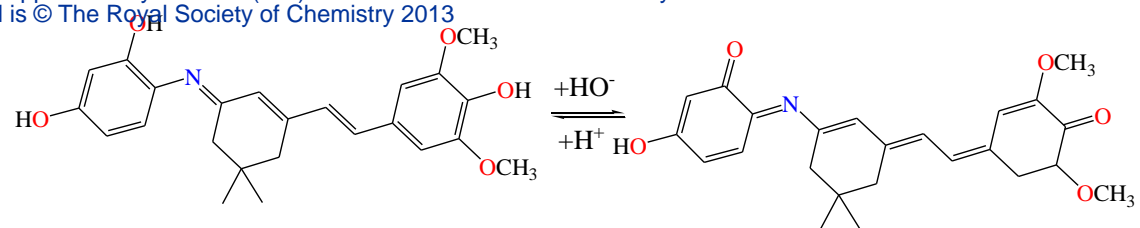




**Scheme S2.** Chemical diagram of the completed list of 74–140 described structural modifications of studied CPLs

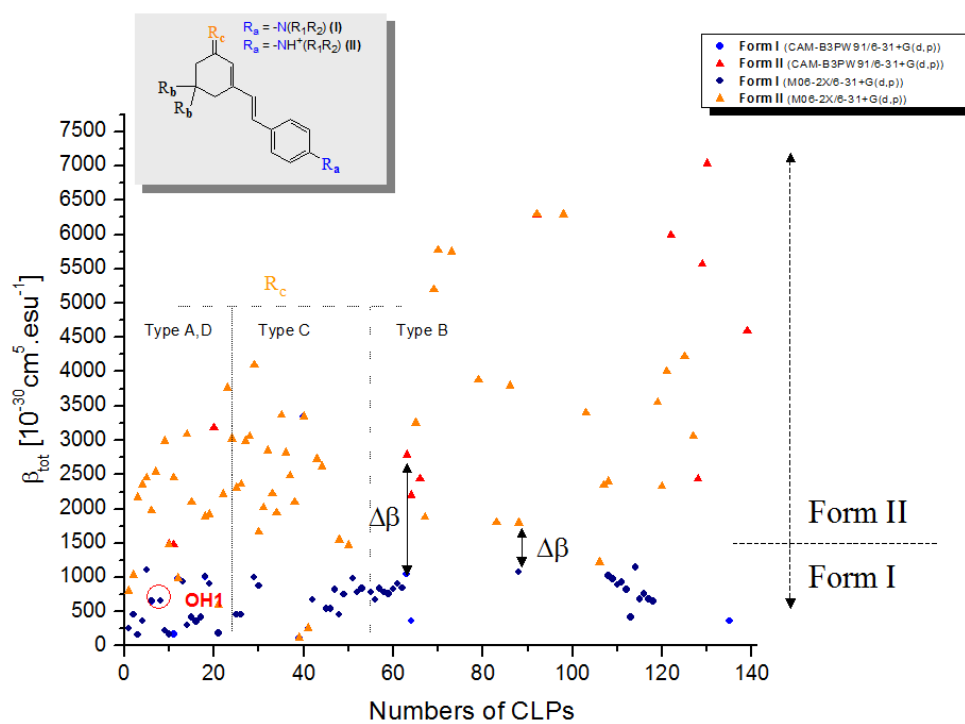


**Scheme S3.** MOs of CLPs **10**, **11** and **19** from subclass **VII** at CAM-B3PW91/6-31+(d,p) level of theory.



**Scheme S4.** Tautomerism of **140** depending of the pH.





**Figure S1.** First hyperpolarizability tensor matrix components  $\beta_{ijk}$ , ( $10^{-30} \text{ cm}^5 \cdot \text{esu}^{-1}$ )