Supporting Information

Design of D-π-A Type Carbon Nanohoop Enhanced Nonlinear Optical Response: A Size-Dependence Effect Study

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Table S1. The components of the first hyperpolarizabilities β_{tot} ($\times 10^{-30}$ esu) of the studied D-CM2PnP-m-A at the CAM-B3LYP/6-31G** level

Compound	β_x	β_y	β_z	β_{tot}
D-CM2P8P-2-A	27.30	1.21	0.26	27.30
D-CM2P10P-3-A	39.22	0.52	1.81	39.30
D-CM2P12P-4-A	48.55	0.35	0.78	48.64

Table S2. The calculated ρ, $\Phi_{J=1}$ and $\Phi_{J=3}$ values of D-CM2PnP-A (n=4, 6, 8, 10, 12) in the three wavelengths at the CAM-B3LYP/6–31G** level

Compound	ρ	$\Phi_{J=1}$ (au)	$\Phi_{J=3}$ (au)
	1064 nm		
D-CM2P4P-A	0.58	0.63	0.37
D-CM2P6P-A	0.51	0.66	0.34
D-CM2P8P-A	0.48	0.68	0.32
D-CM2P10P-A	0.43	0.70	0.30
D-CM2P12P-A	0.43	0.70	0.30
	1460 nm		
D-CM2P4P-A	0.62	0.62	0.38
D-CM2P6P-A	0.53	0.66	0.35
D-CM2P8P-A	0.50	0.67	0.33
D-CM2P10P-A	0.44	0.69	0.31
D-CM2P12P-A	0.45	0.69	0.31
	1907 nm		
D-CM2P4P-A	0.64	0.611	0.39
D-CM2P6P-A	0.54	0.650	0.35
D-CM2P8P-A	0.51	0.662	0.34
D-CM2P10P-A	0.46	0.685	0.32
D-CM2P12P-A	0.46	0.683	0.32

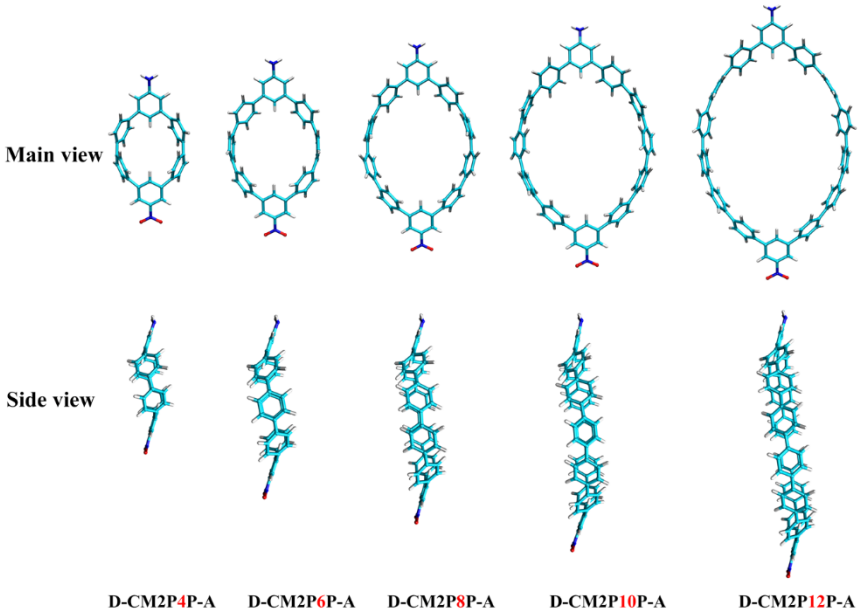

Figure S1. The main view and side view of the D-CM2PnP-A (n=4, 6, 8, 10, 12)

Figure S2. Frontier molecular orbital energy levels and molecular orbital diagram of CM2P4P

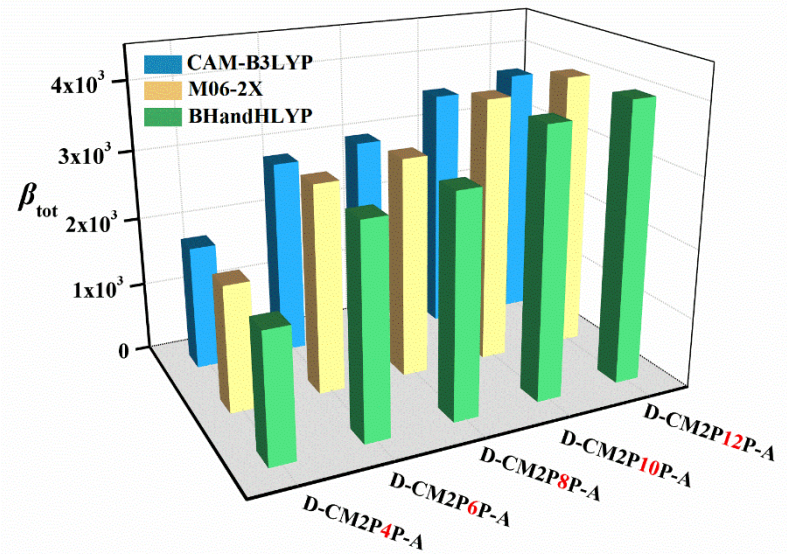

Figure S3. The first hyperpolarizabilities of the D-CM2PnP-A (n=4, 6, 8, 10, 12) calculated by using various functionals with the 6-31G** basis set

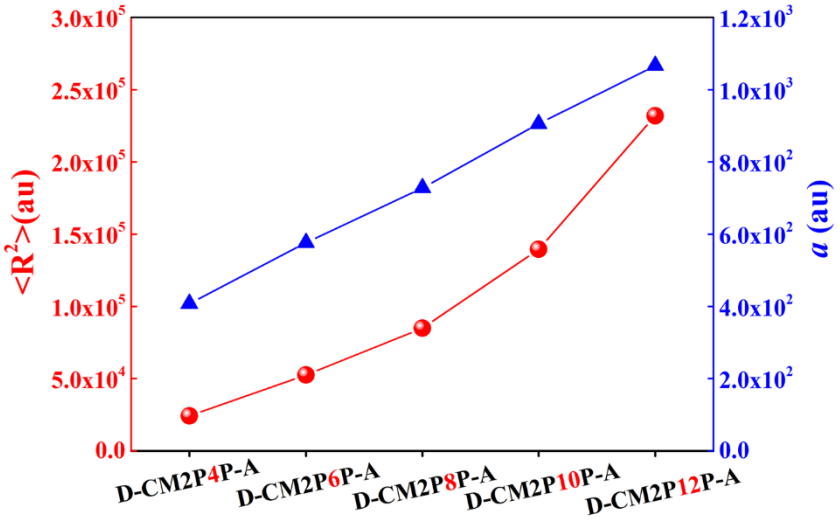

Figure S4. Relationship between the α values and the corresponding $\langle R^2 \rangle$ values of D-CM2PnP-A (n=4, 6, 8, 10, 12)

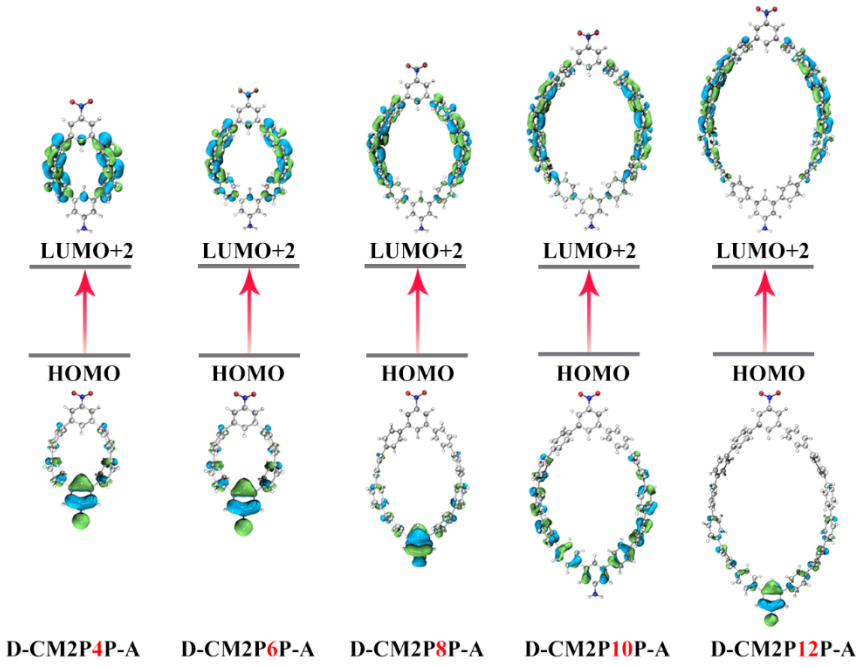

Figure S5. The molecular orbitals of the main excited states of D-CM2PnP-A (n=4, 6, 8, 10, 12)