Electronic Supplementary Information

Design of Novel Graphdiyne-based Materials with Large Second-Order Nonlinear Optical Properties

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Contents:

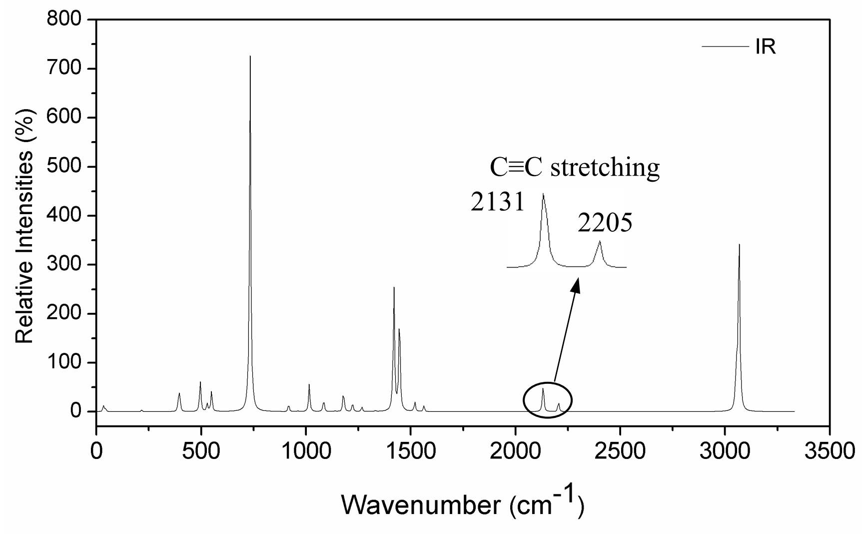

Fig. S1. Simulated infrared spectrum of the GDY cluster, obtained at the B3LYP/6-31+G(d) level of theory. The scaling factor of 0.953 was applied to correct all calculated vibrational frequencies. The vibrations for the C≡C stretching modes are specially labeled.

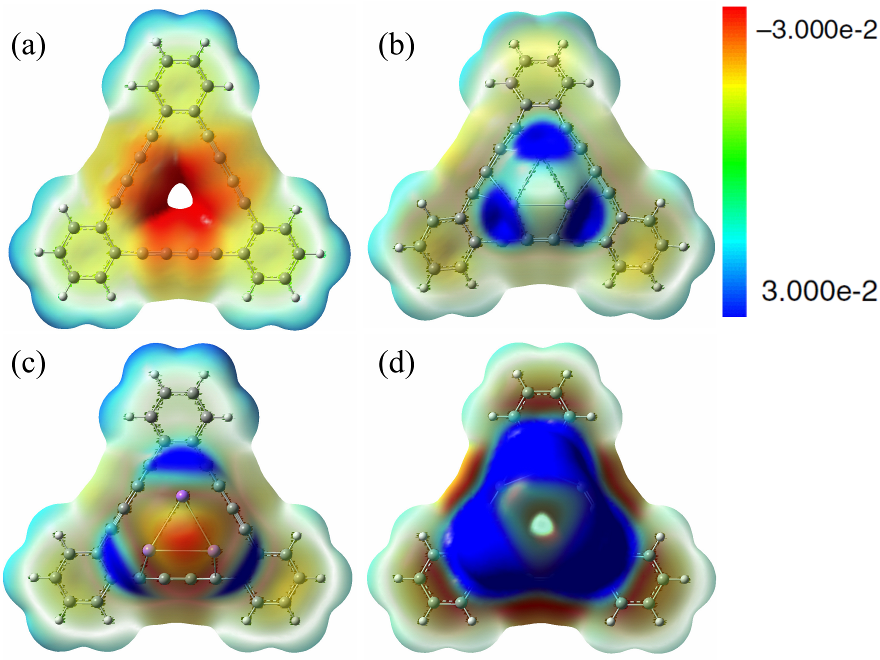

Fig. S2. Electrostatic potential maps of the (a) GDY, (b) Li_3@GDY, (c) Na_3@GDY, and (d) K_3@GDY clusters.

Table S1 Mean dipole moment (μ_0, in a.u.), static polarizability (α_0, in a.u.), the static first hyperpolarizability (β_{tot}, in a.u.), transition energy (ΔE, in eV), maximum oscillator strength (f_0, in a.u.), the change in dipole moment ($\Delta\mu$, in a.u.), and crucial transitions of crucial excited energies for the AM_3@GDY (AM = Li, Na, K) clusters.

Clusters	μ_0	α_0	β_{tot}	ΔE	f_0	$\Delta\mu$	Crucial Transitions*
Li_3@GDY	0.84	671.19	9208.88	3.32	0. 178	2.186	β(H → L+16) (35%), β(H → L+20) (19%)
Na_3@GDY	1.62	786.44	69788.24	2.75	0.232	3.444	α(H → L+10) (26%), α(H → L+5) (11%)
K_3@GDY	3.32	1065.49	161201.31	I: 2.98	0. 777	10.650	β(H-2 → L) (14%), β(H-1→ L+1) (13%)
				II: 1.91	0. 315	6.735	α(H → L+4) (34%), β(H → L+2) (22%)

*H = HOMO, L = LUMO.