*Electronic Supplementary Information*

# Investigations of electronic and nonlinear optical properties of single alkali-metal adsorbed graphene, graphyne and graphdiyne systems by first-principles calculations

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**Contents:**



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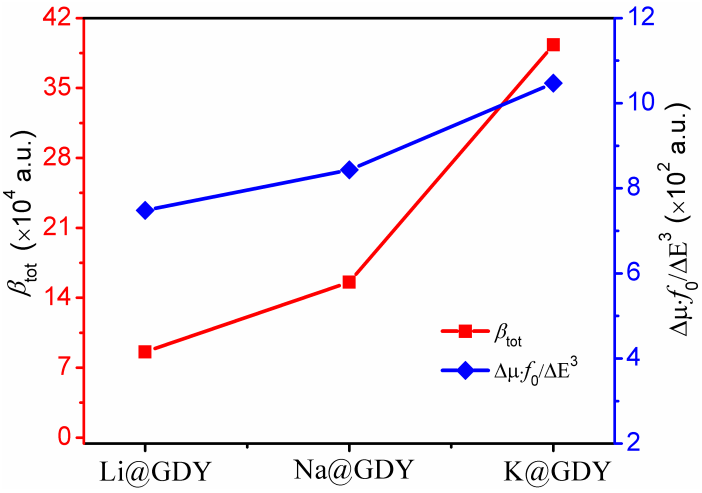


**Figure S1.** Comparison of exactly calculated static first hyperpolarizability ($\beta_{tot}$) and estimated by two-level model ($\Delta\mu\cdot f_0/\Delta E^3$) for these AM@GDY (AM = Li, Na, and K) complexes.

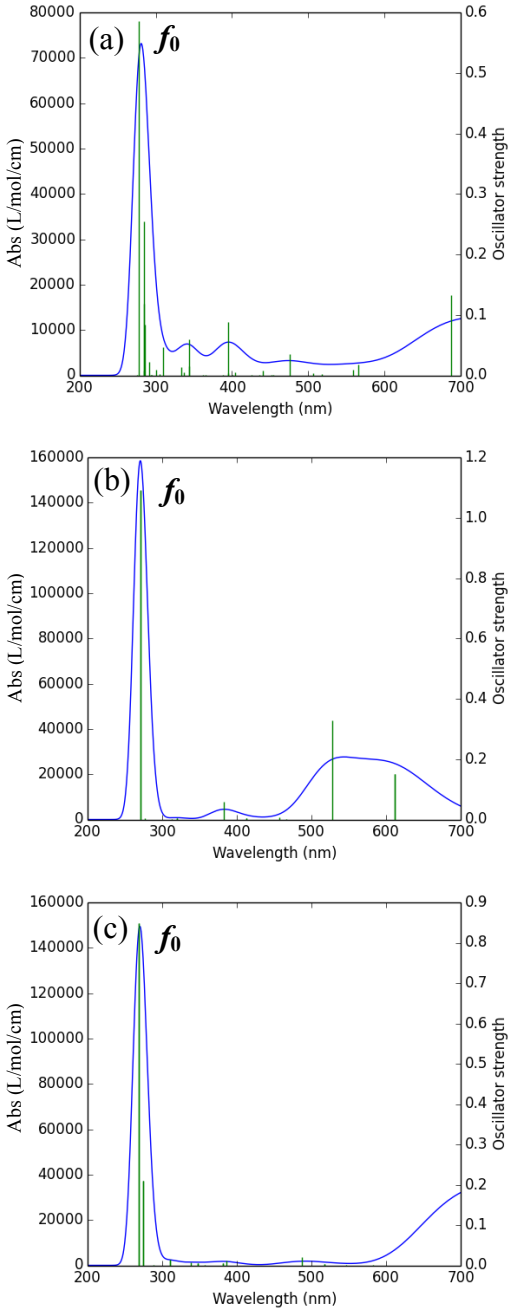


**Figure S2.** Simulated optical absorption spectra of (a) Li@GE, (b) Na@GE, and (c) K@GE complexes using the CAM-B3LYP method. The oscillator strengths of them are inserted.

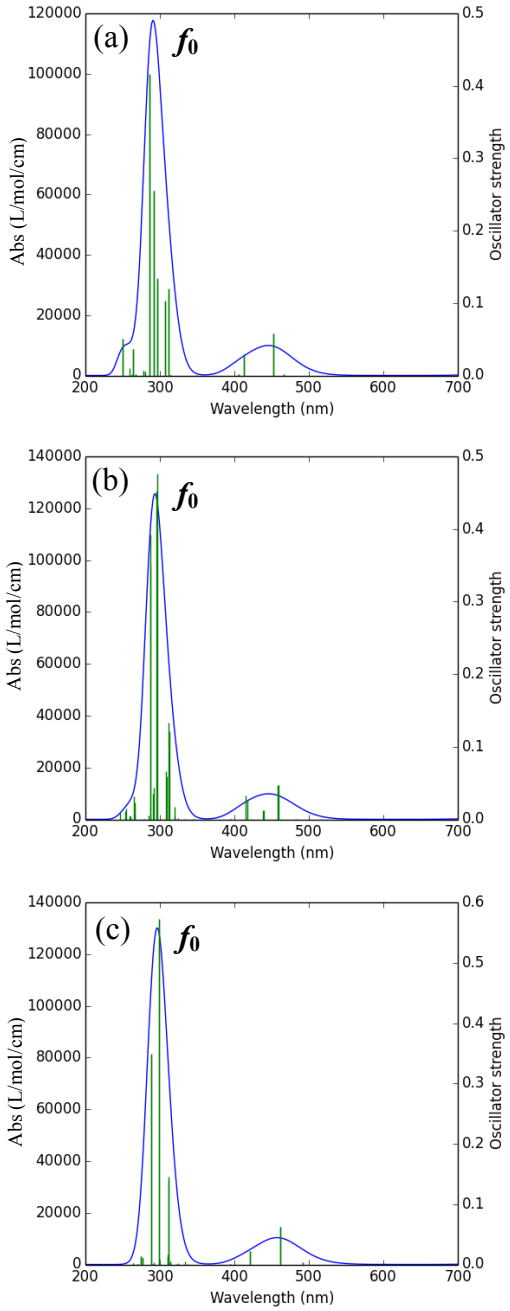


**Figure S3.** Simulated optical absorption spectra of (a) Li@GY, (b) Na@GY, and (c) K@GY complexes using the CAM-B3LYP method. The oscillator strengths of them are inserted.

**Table S1.** Maximum oscillator strength ($f_0$, in a.u.), the change in dipole moments ($\Delta\mu$, in a.u.) between ground and crucial excited states, and main transitions of crucial excited states for AM@GE, AM@GY, and AM@GDY (AM = Li, Na, K) complexes, obtained at the TD-CAM-B3LYP/6-31+G(d) level of theory.

| Complexes | $f_0$ | $\Delta\mu$ | Crucial Transitions[a] |
|---|---|---|---|
| Li@GE | 0.5859 | 5.357 | αH-2 → αL (16%) |
| | | | αH-1 → αL+1 (17%) |
| | | | βH-1 → βL (16%) |
| | | | βH →βL+1 (17%) |
| Na@GE | 1.0916 | 9.719 | αH-2 → αL (24%) |
| | | | αH-1 → αL+1 (24%) |
| | | | βH-1 → βL (24%) |
| | | | βH → βL+1 (24%) |
| K@GE | 0.8477 | 7.5046 | αH-2 → αL (11%) |
| | | | αH-1 → αL+1 (11%) |
| | | | βH-1 → βL (12%) |
| | | | βH → βL+1 (12%) |
| Li@GY | 0.4167 | 3.920 | βH → βL+2 (22%) |
| | | | αH-1 → αL+1 (10%) |
| Na@GY | 0.4767 | 4.646 | αH-1 → αL+1 (25%) |
| | | | βH → βL+2 (29%) |
| K@GY | 0.5727 | 5.629 | αH-1 → αL+1 (30%) |
| | | | βH → βL+5 (30%) |
| Li@GDY | 0.5314 | 3.228 | αH-2 → αL (43%) |
| | | | βH-1 → βL (23%) |
| Na@GDY | 0.4349 | 4.374 | αH-2 → αL (24%) |
| | | | βH-1 → βL (19%) |
| K@GDY | 0.3344 | 7.044 | αH-2 → αL (41%) |
| | | | βH-1 → βL (13%) |

[a]H = HOMO, L = LUMO.