**Supplementary Information**

**Discriminative modulation of the highest occupied molecular orbital energies of graphene and carbon nanotubes induced by charging**

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**Fig. S1** Models of the (3,5) CNT containing 80 carbon atoms and its corresponding GS. (a) The CNT model with all the edges terminated by 16 hydrogen atoms. (b) The GS model with all the edges terminated by 28 hydrogen atoms. (c) The GS model with 16 hydrogen atoms, where only the edges originally from the CNT model are terminated by hydrogen atoms. (d) The GS model with the edges terminated by 22 hydrogen atoms. The models in (a, b) are the same as those in Fig. 1(a, b) of the main text.

**Fig. S2** Dependences of the HOMO energies of the (3,5) CNT and corresponding unzipped GS with different hydrogen atoms on the amount of the extra electrons, where the vacuum energy is set at 0 eV.