

Graphene nanoribbons generate a strong third-order nonlinear optical response upon intercalating hexagonal boron nitride

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(Supplementary information)

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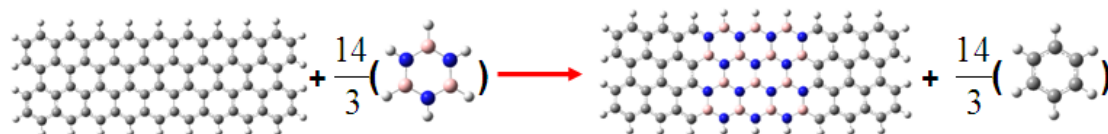
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Formation energy of BCNNRs

Here we design a reactions to simulate the formation of BCNNRs and calculated the formation energy to confirm the stability of the designed BCNNRs. We choose the (3, 9) H-aBCNNRs and (9, 3) H-zBCNNRs as two products on behalf of BCNNRs. The reactants are Graphene nanoribbon and Borazine ($B_3N_3H_6$). We suppose that B and N atoms from Borazine would replace the carbon atoms in graphene nanoribbon, and form the BCNNRs structures.

1. For (3,9) H-aBCNNRs, the reactants are (3,9)graphene nanoribbon (G1) and Borazine(Bor); the products are the (3,9) H-aBCNNRs (BCN1)and benzene(Ben). All the energy of reactants and products were obtained from their optimized structures at DFT-B3LYP-6-31G* level, respectively.

The reaction as follow:



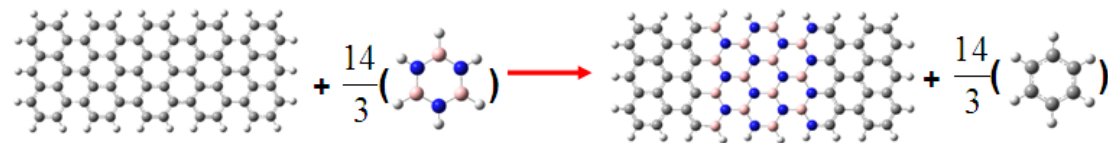
Energy: $E(G1)=-2912.100\text{au}$ $E(Bor)= -242.670\text{au}$

$E(BCN1)=-2960.858\text{au}$ $E(Ben)= -232.249\text{au}$

$$\text{Formation energy(1)}= E(BCN1)+14/3*E(Ben)-E(G1)- 14/3*E(Bor) \\ =-0.126\text{au}=-330.813 \text{ kJ/mol}$$

2. For (9, 3) H-zBCNNRs, the reactants are (9,3)graphene nanoribbon (G2) and Borazine(Bor); the products are the (9,3) H-aBCNNRs (BCN2) and benzene(Ben). All the energy of reactants and products were obtained from their optimized structures at DFT-B3LYP-6-31G* level, respectively.

The reaction as follow:



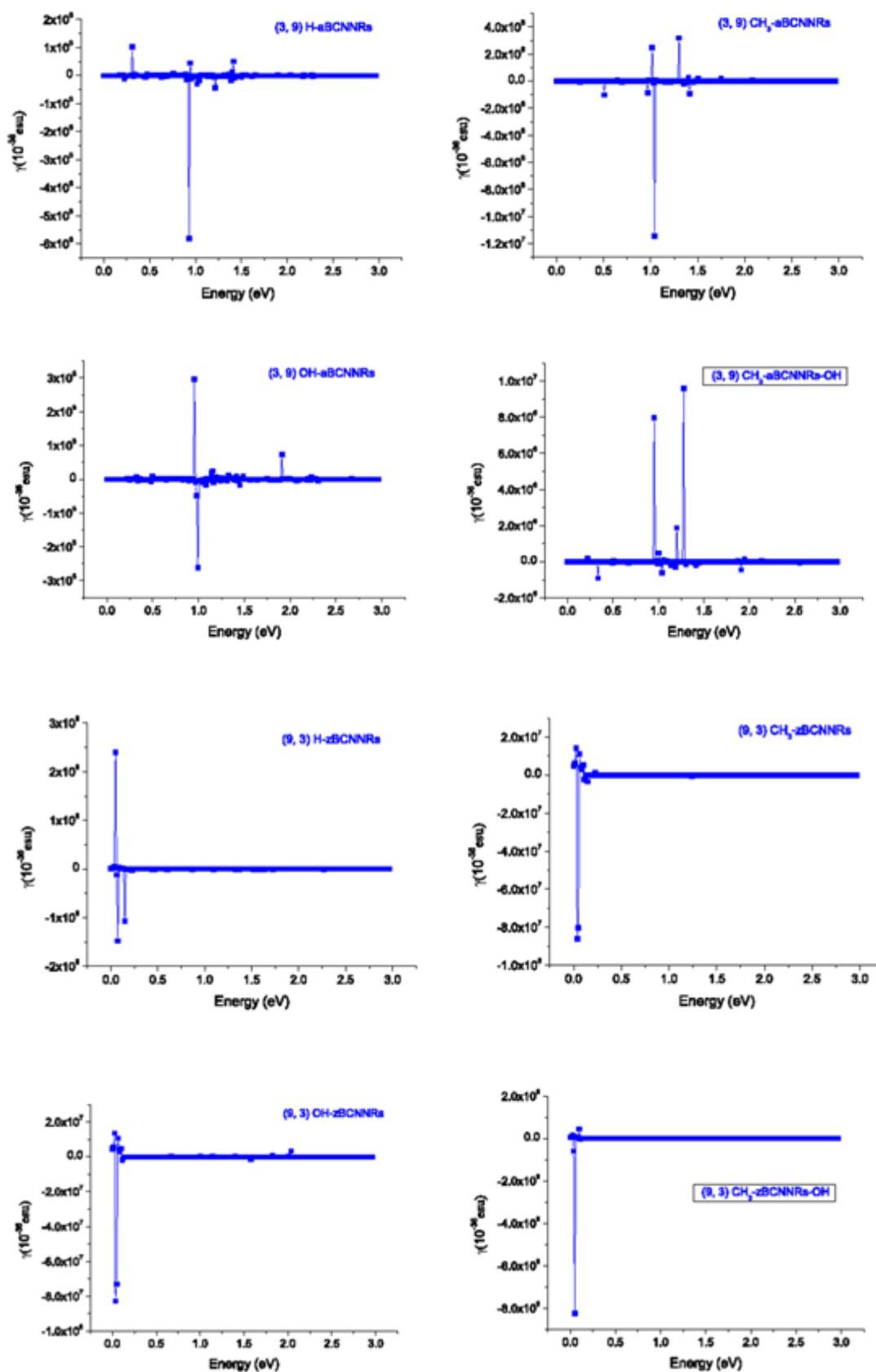
Energy: $E(G2)= -2683.457\text{au}$ $E(Bor)= -242.670\text{au}$

$E(BCN2)= -2732.123\text{au}$ $E(Ben)= -232.249\text{au}$

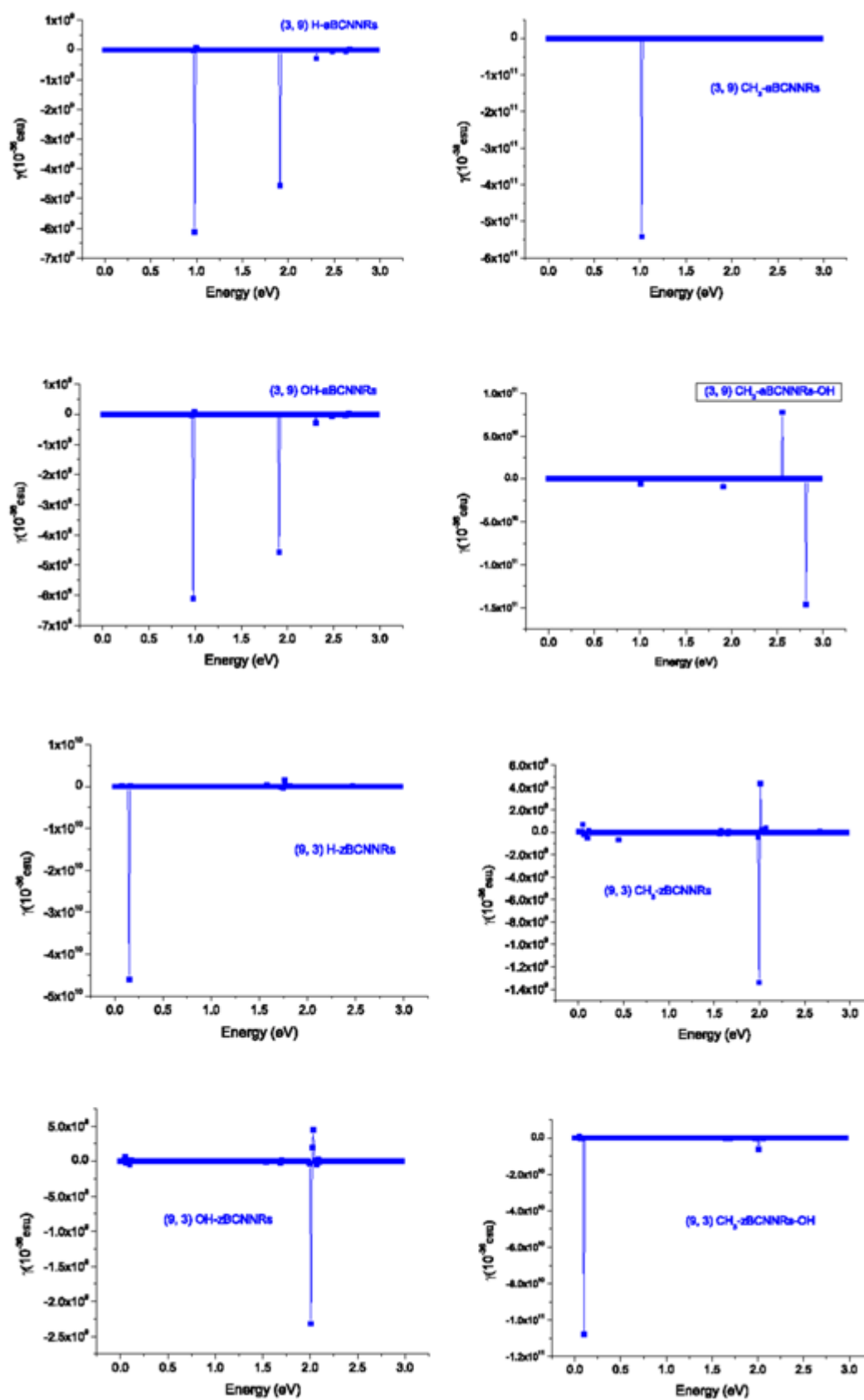
$$\text{Formation energy(2)}= E(BCN2)+14/3*E(Ben)-E(G2)- 14/3*E(Bor) \\ =-0.0347\text{au}= -91.105\text{kJ/mol}$$

The negative formation energy of (3, 9) H-aBCNNRs and (9, 3) H-zBCNNRs indicated the stability of the designed BCNNRs

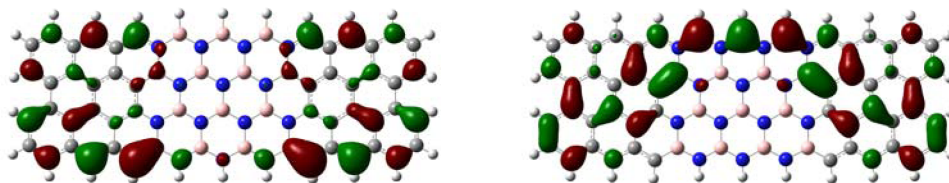
Sfig1. Dynamic third order NLO polarizabilities in THG process.



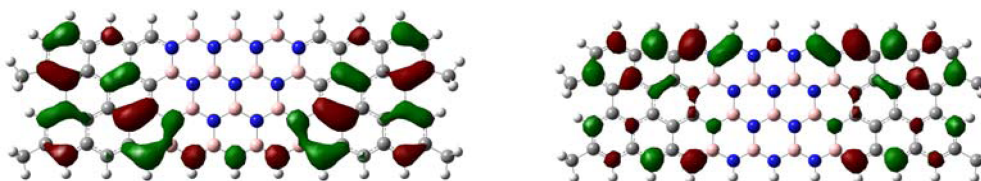
Sfig 2. Dynamic third order NLO polarizabilities in DFWM process.



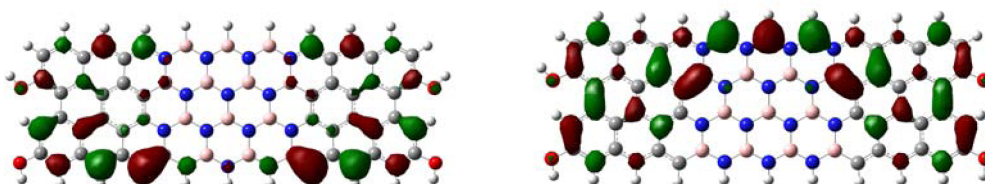
Sfig 3. Isosurfaces of the frontier orbitals which participate in the relevant TPA excitations for the studied conformers



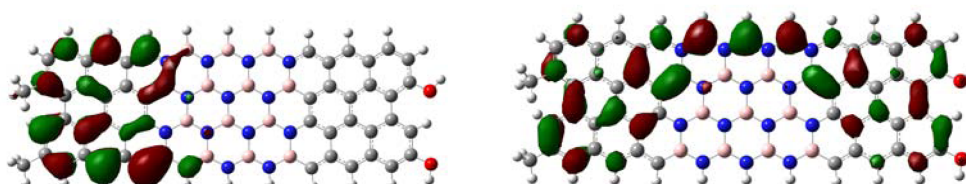
(3, 9) H-ABCNNRs: HOMO->LUMO+2



(3, 9) CH₃-ABCNNRs: HOMO-2->LUMO

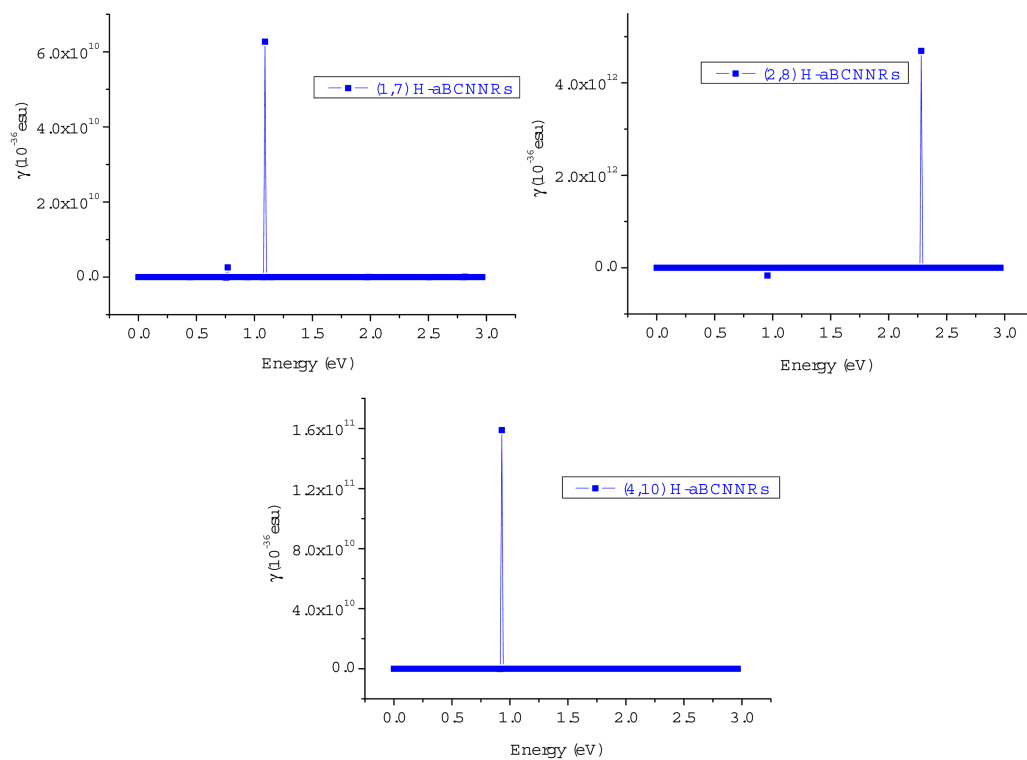


(3, 9) OH-ABCNNRs: HOMO->LUMO+2



(3, 9) CH₃OH-ABCNNRs: HOMO-1->LUMO+2

Sfig 4. Third order NLO polarizability of (1,7) H-aBCNNRs, (2,8) H-aBCNNRs, (4,10) H-aBCNNRs in DFWM process, respectively.



Sfig 5. Two-photon absorption spectra for (1,7) OH-aBCNNRs, (2, 8) OH-aBCNNRs, (4,10) OH-aBCNNRs, respectively.

