Supplementary Information

Orientational DNA binding and directed transport on nanomaterial heterojunctions

Ye Deng[a†], Fuxin Wang[a†], Yang Liu[a], Yanmei Yang[*,b], Yuanyuan Qu[a], Mingwen Zhao[a], Yuguang Mu[*,c], and Weifeng Li[*,a]

a. *School of Physics and State Key Laboratory of Crystal Materials, Shandong University, Jinan, Shandong, 250100, China*

b. *College of Chemistry, Chemical Engineering and Materials Science, Collaborative Innovation Center of Functionalized Probes for Chemical Imaging in Universities of Shandong, Key Laboratory of Molecular and Nano Probes, Ministry of Education, Institute of Molecular and Nano Science, Shandong Normal University, Jinan, 250014, China*

c. *School of Biological Sciences, Nanyang Technological University, 637551, Singapore*

† These authors contribute equally.

* Corresponding authors. yym@sdnu.edu.cn (Y.Y.), ygmu@ntu.edu.sg (Y.M.), *lwf@sdu.edu.cn* (W.L.)

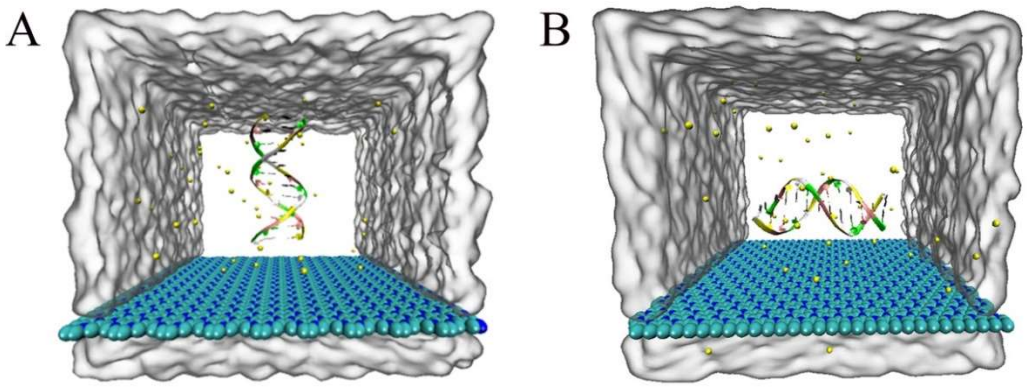

Figure S1. The simulation models of dsDNA with (A) perpendicular and (B) parallel orientations on the C_3N surface. Salt ions are shown as yellow balls. Water solvents are implicitly represented as gray surface.

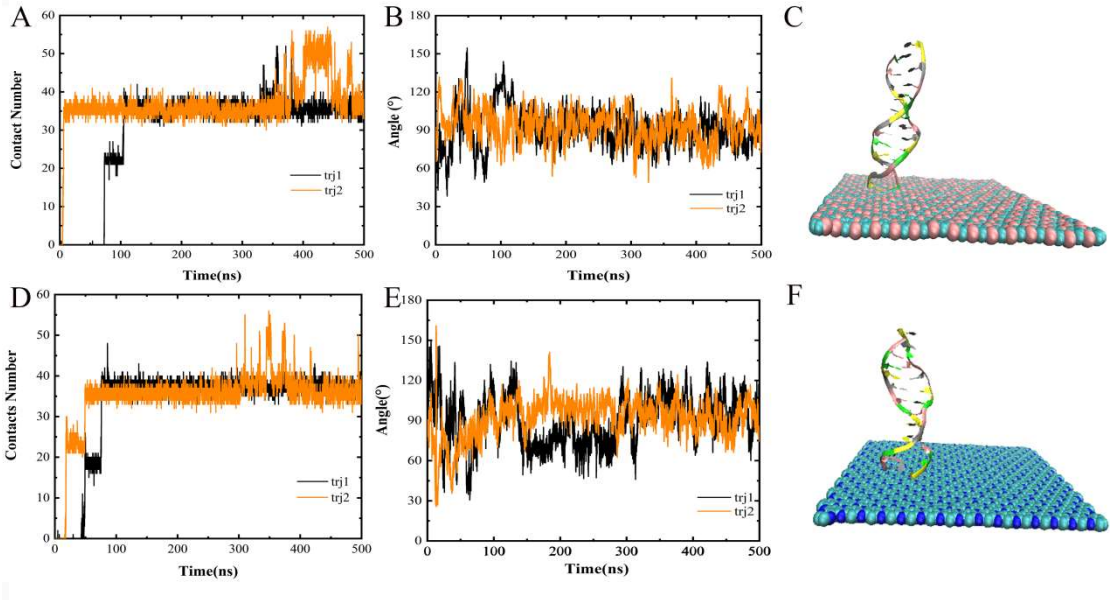

Figure S2. Simulation results of dsDNA with initially perpendicular orientation on BC_3 and C_3N plane. Time evolutions of (A) number of contacts between dsDNA and nanosheets and (B) dsDNA orientation with respect to nanosheet normal direction for BC_3 simulations. Time evolutions of (D) number of contacts between dsDNA and nanosheets and (E) dsDNA orientation with respect to nanosheet normal direction for C_3N simulations. (C) and (F) depict the final conformations of dsDNA binds to the BC_3 and C_3N after 500 ns simulation.

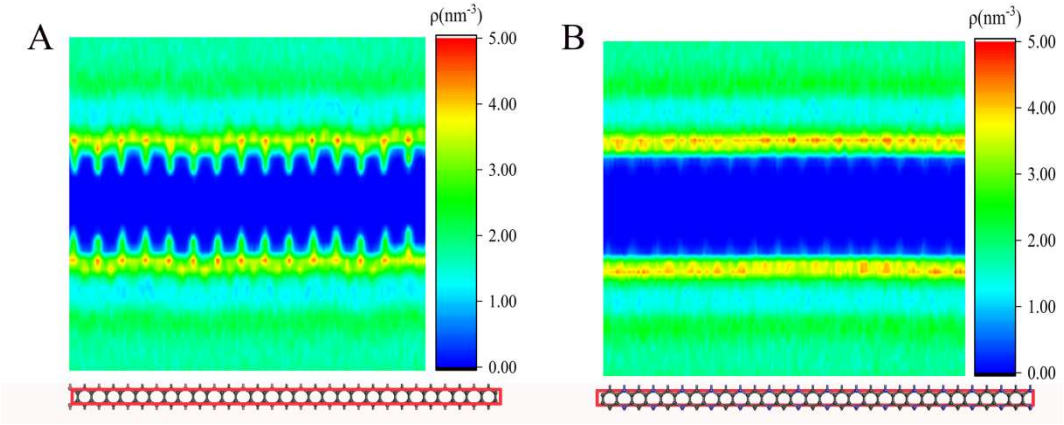

Figure S3. The density map of water along the normal directions of (A) BC_3 and (B) C_3N plane. As depicted in the structure below, the calculations were performed along the zigzag direction. Only water molecules locating above and under the hexagonal rings (as indicated by the red square in the figure below) were counted. The red color regions indicate enhanced water clusters above two nanosheets, especially for the case of BC_3.

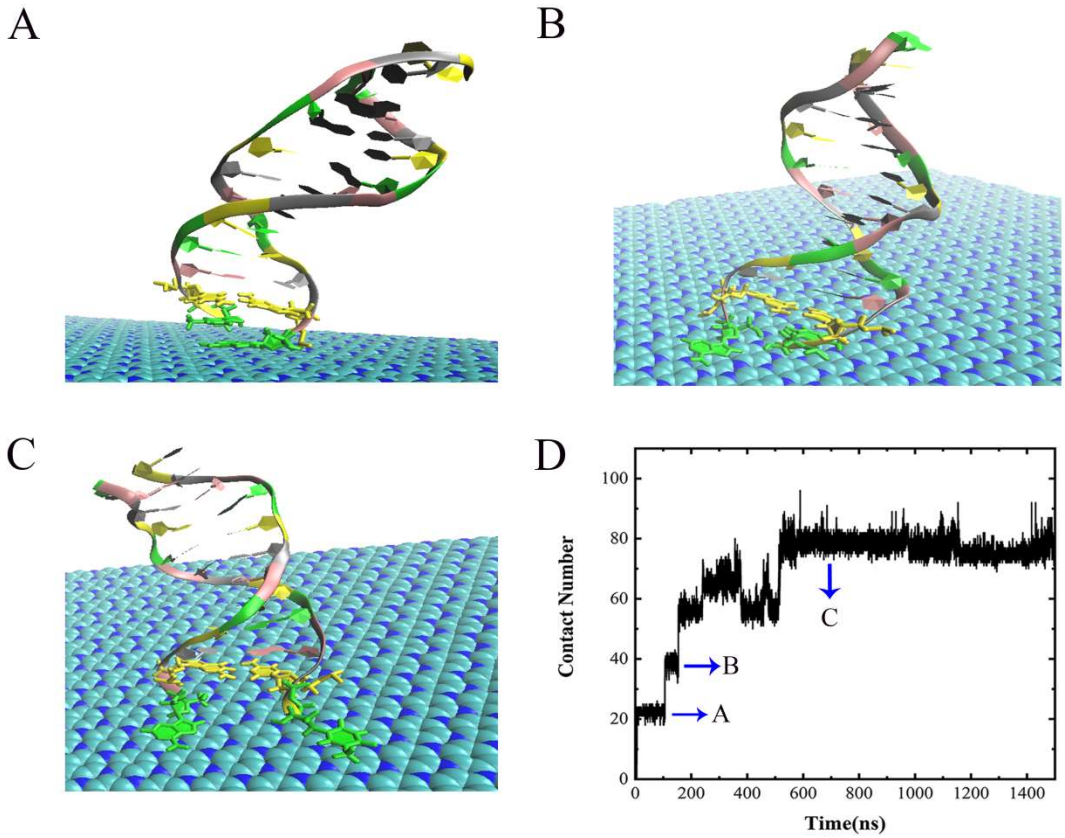

Figure S4. (A-C) The unpairing process of A:T base pair when binding on C_3N. The time evolution of number of contacts between the dsDNA and C_3N during the binding is shown in (D).

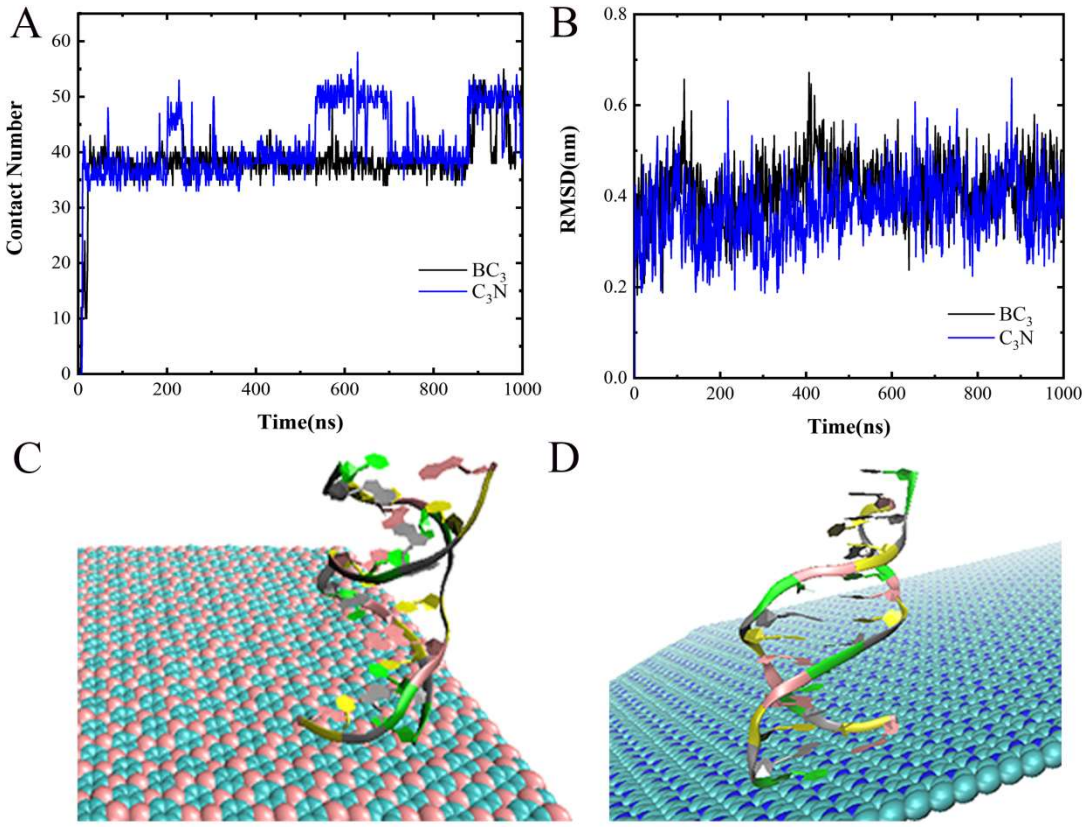

Figure S5. 1000 ns simulations of dsDNA binding on C_3B and C_3N through GC base. Time evolutions of (A) number of contacts between dsDNA and BC_3/C_3N and (B) root mean squared displacement (RMSD) of dsDNA. (C) and (D) depict the final binding conformation of the dsDNA on the BC_3 and C_3N respectively.

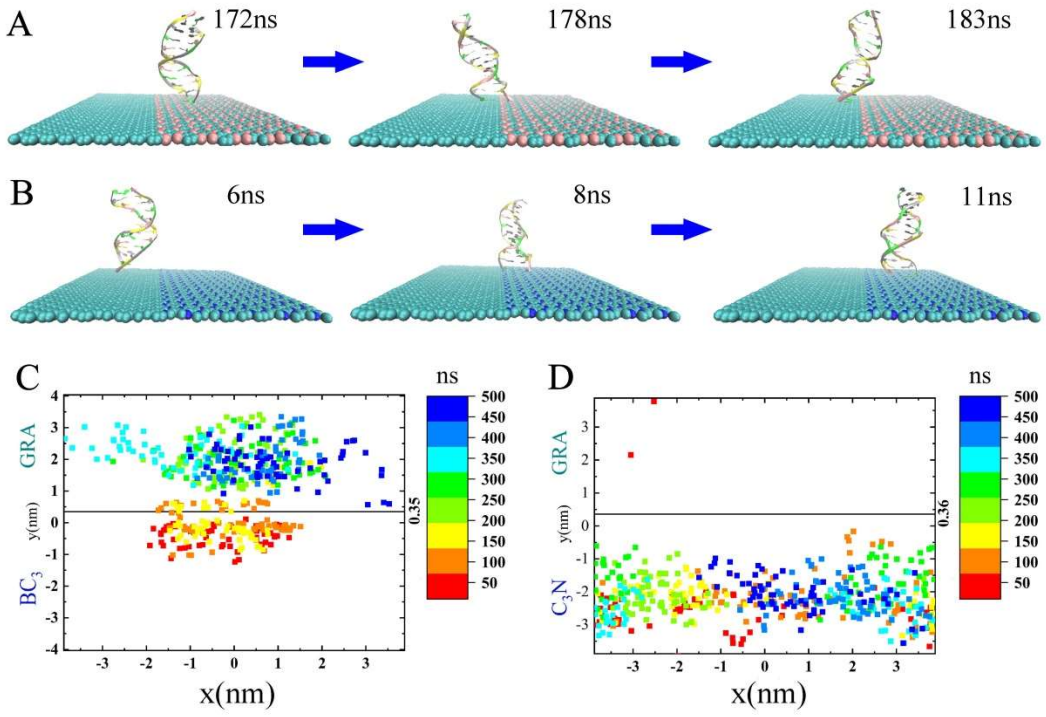

Figure S6. Schematic illustrations of dsDNA migrated across the boundaries of heterojunctions of (A) BC_3/GRA and (B) C_3N/GRA. The migration path of DNA on (A) BC_3/GRA and (B) C_3N/GRA heterojunctions. The black lines in the figure represent the boundaries between the two materials in the heterojunction.

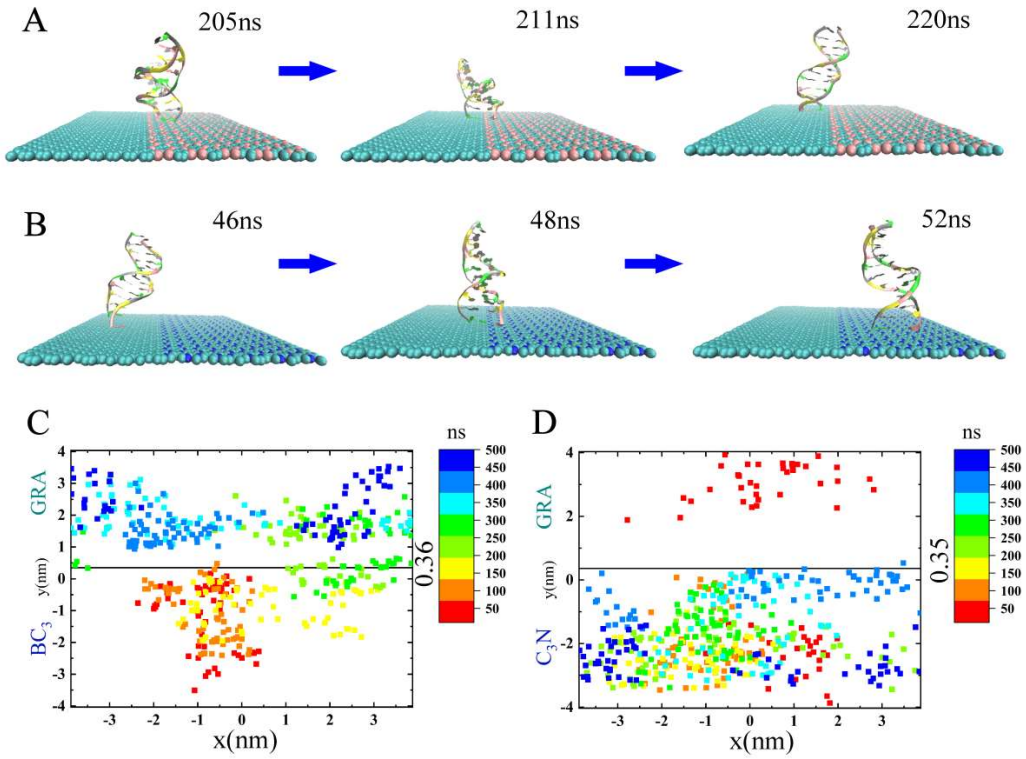

Figure S7. Schematic illustrations of dsDNA migrated across the boundaries of heterojunctions of (A) BC_3/GRA and (B) C_3N/GRA. The migration path of DNA on (A) BC_3/GRA and (B) C_3N/GRA heterojunctions. The black lines in the figure represent the boundaries between the two materials in the heterojunction.

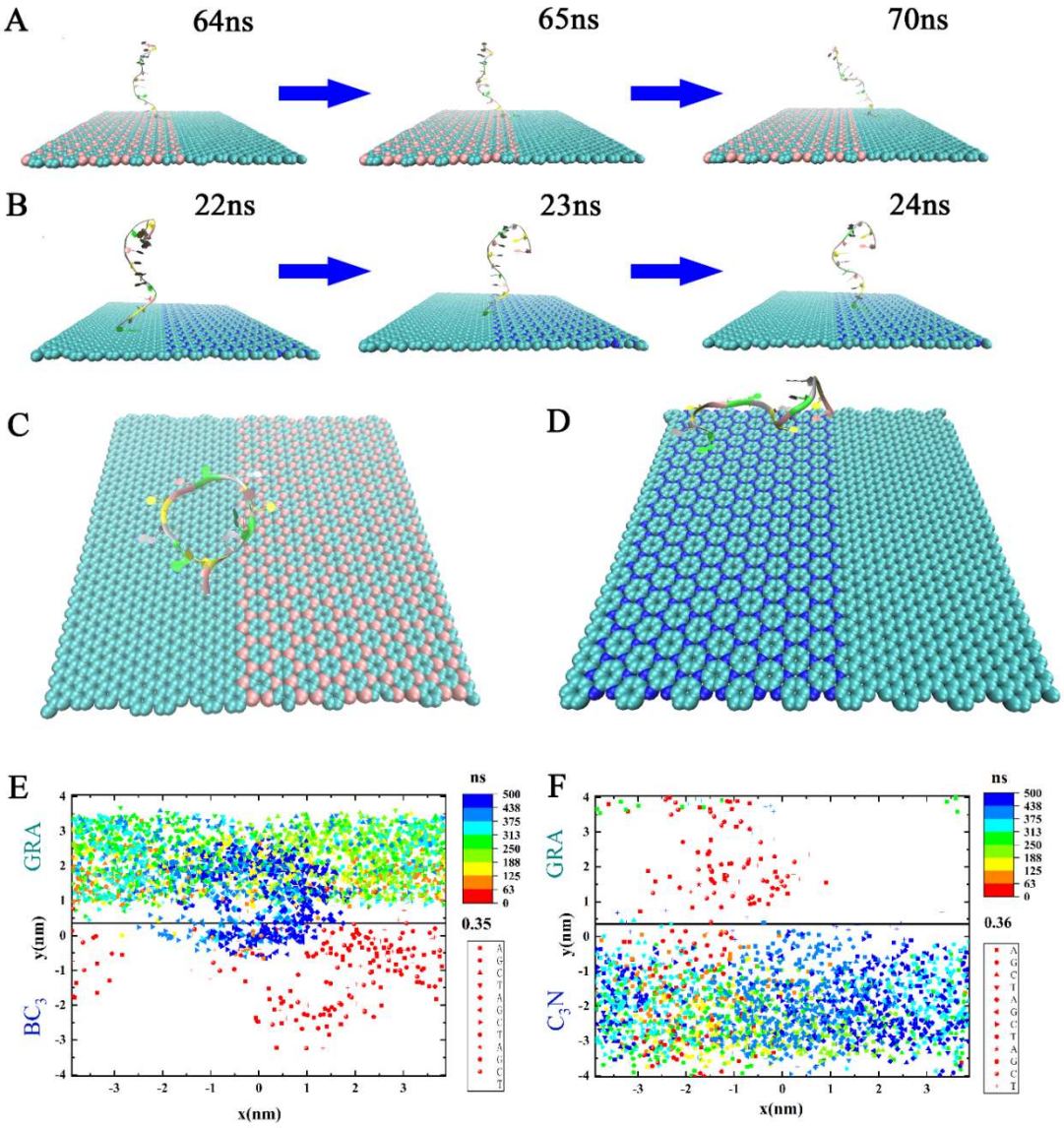

Figure S8. Simulation results of single strand DNA (ssDNA). Migrating processes of ssDNA across the boundaries of heterojunctions of (A) BC_3/GRA and (B) C_3N/GRA. The final binding conformations of the ssDNA on (C) BC_3 and (D) C_3N. The migration path of each nucleotide base of ssDNA on (E) BC_3/GRA and (F) C_3N/GRA heterojunctions. The black lines near y = 0 in (E) and (F) represent the boundaries between the two materials in the heterojunction.

Table S1. The force field parameters.

	ε (kj/mol)	σ (nm)	Charge (e)
C (BC_3)	0.360	0.340	-0.126
B (BC_3)	0.399	0.345	0.378
C (C_3N)	0.360	0.340	0.056
N (C_3N)	0.711	0.325	-0.168

The atomic charges of BC_3 and C_3N were calculated by Quantum Mechanics calculations using Gaussian 09 at the HF/6-31G* level and parameterized using the RESP method. The parameters of the Gaussian's input file are set to #HF/ 6-31g * SCF=tight Test Pop=MK iop(6/33=2, 6/42=6, 6/50=1) opt.