Supplementary Information

Towards Graphene Semi/Hybrid-Nanogap: A New Architecture for Ultrafast DNA Sequencing

Sneha Mittal, † Biswarup Pathak*, †

†Department of Chemistry, Indian Institute of Technology (IIT) Indore, Indore, Madhya Pradesh, 453552, India
*E-mail: biswarup@iiti.ac.in

CONTENTS

1. Representative in-plane rotation orientations of A-nucleobase inside the graphene semi/hybrid-nanogap:

Scheme S1: Rotation of A-nucleobase inside the graphene semi/hybrid-nanogap: We have considered all possible rotations from 0° to 180° around the x-axis in the yz-plane for all four DNA nucleobases inside the graphene semi/hybrid-nanogap as shown in **Figure S1.**

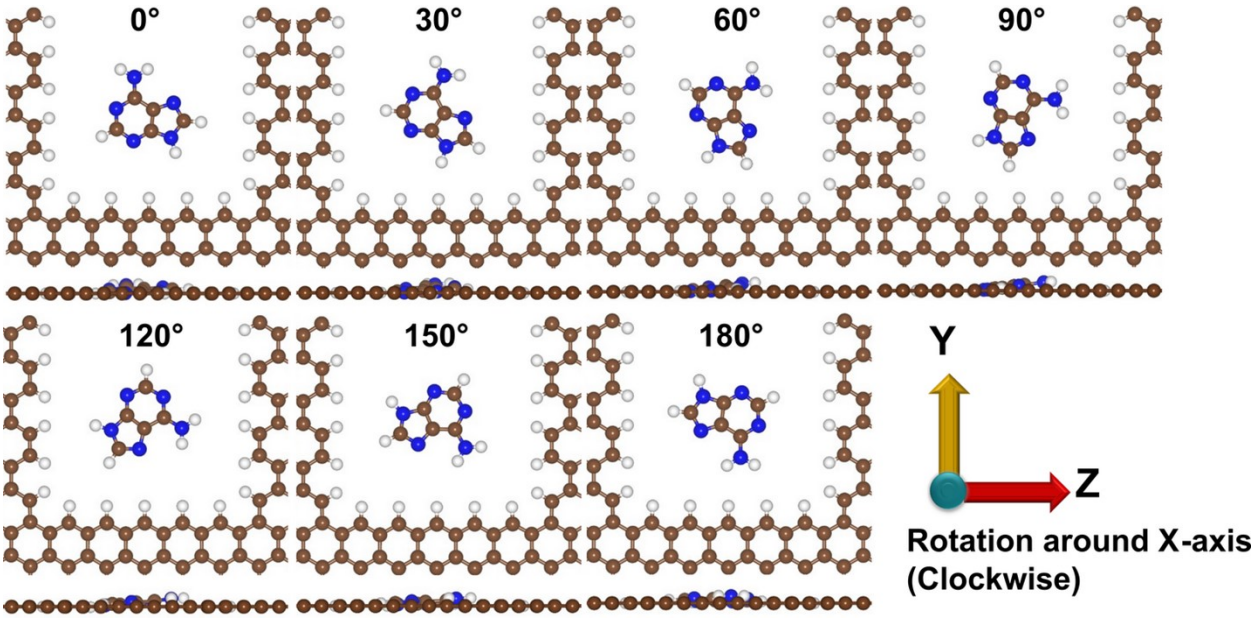

Figure S1: Representative orientations of A-nucleobase inside the graphene semi/hybrid-nanogap are illustrated, corresponding to rotations from 0° to 180° in the steps of 30° around the x-axis in the yz-plane.

Table S1. Relative energies (in eV) of the graphene semi/hybrid-nanogap+nucleobase systems when nucleobases are interacting with graphene semi/hybrid-nanogap edges at different orientations (0° to 180°), as shown in **Figure S1**.

Nucleobases	**0^0**	**30^0**	**60^0**	**90^0**	**120^0**	**150^0**	**180^0**
A	**0.00**	0.44	0.49	0.55	0.03	0.09	0.04
G	0.13	0.73	0.39	**0.00**	0.50	1.09	1.21
C	0.08	0.27	**0.00**	0.01	0.06	0.73	0.85
T	0.17	0.57	0.63	0.64	0.47	**0.00**	0.43

2. Total density of states (TDOS) plot for pristine graphene semi/hybrid-nanogap device at different k-points:

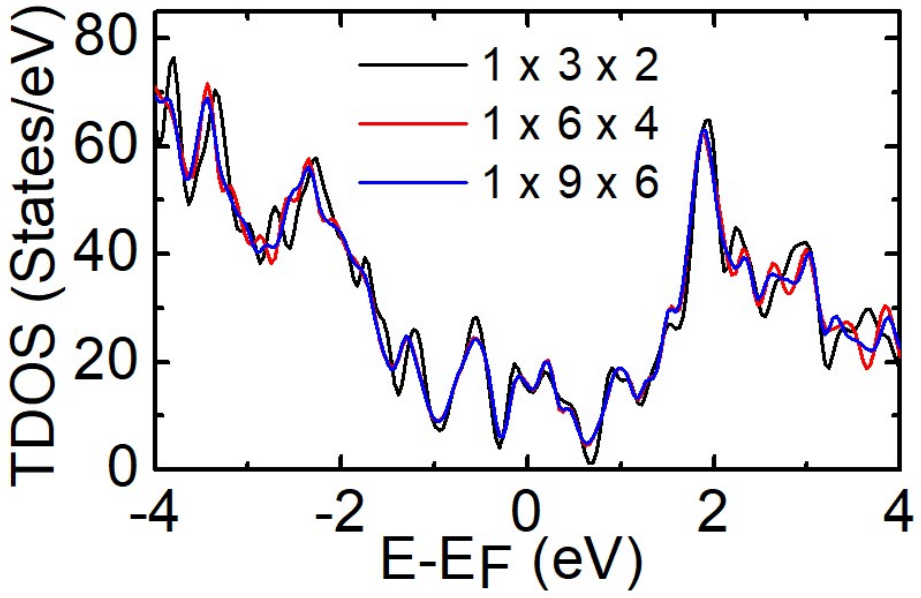

Figure S2: TDOS plot for pristine graphene semi/hybrid-nanogap device at three different k-points: 1×3×2, 1×6×4, and 1×9×6. The Fermi energy level ($E-E_F$) has been aligned to zero.

3. Top and side views of energetically stable structures of graphene semi/hybrid-nanogap+nucleobase-analog (1dzC and 3dzT) systems:

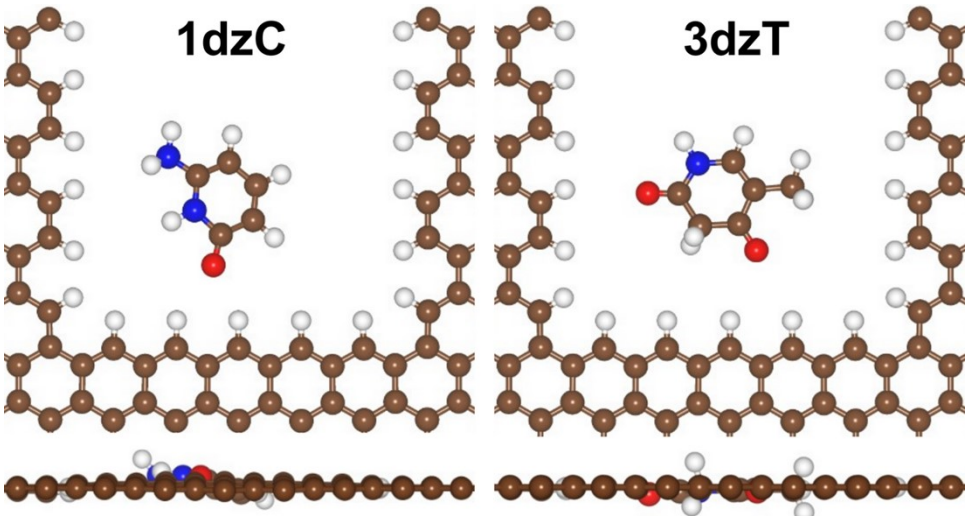

Figure S3. Top and side views of energetically stable structures of graphene semi/hybrid-nanogap+nucleobase-analog (1dzC and 3dzT) systems. Brown, white, blue, and red balls represent C, H, N, and O atoms, respectively.

4. Change in the zero-bias transmission spectra of graphene semi/hybrid nanogap+pyrimidine nucleobase systems due to highly conductive analogs:

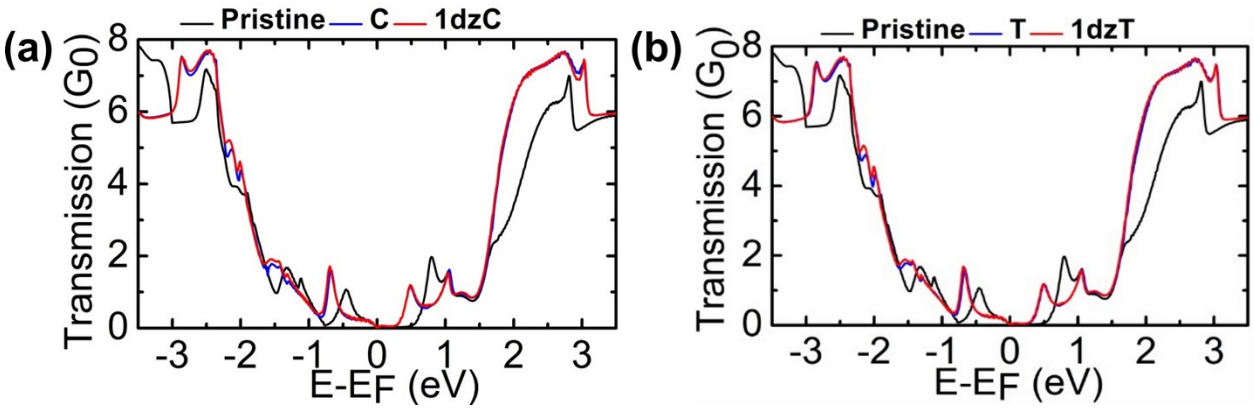

Figure S4. (a) Variation in the zero-bias transmission spectra of C when replaced by the analog 1dzC and **(b)** variation in the zero-bias transmission spectra of T when replaced by the analog 3dzT. The Fermi level has been set to zero.

5. Representative lateral translations of A-nucleobase inside the graphene semi/hybrid-nanogap:

Scheme S2: Lateral translations of A-nucleobase inside the graphene semi/hybrid-nanogap: We have translated all four nucleobases in both forward (+0.5Å) and backward (-0.5Å) directions from the initial position (0.0 Å) inside the graphene semi/hybrid-nanogap as shown in **Figure S5**.

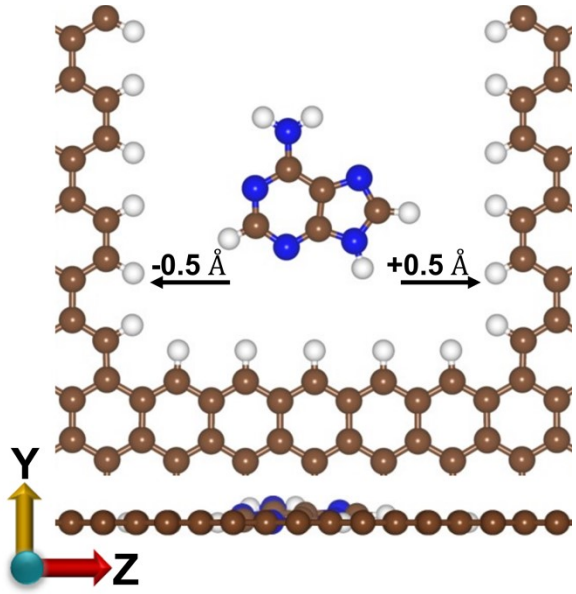

Figure S5: Scheme of A-nucleobase translated inplane along the z-axis in positive and negative directions by ± 0.5Å inside the graphene semi/hybrid-nanogap in the xy-plane.

6. Scheme of A-nucleobase translated out-of-plane along the x-axis in positive and negative directions by ± 1.0 Å inside the graphene semi/hybrid-nanogap:

Scheme S3: Out-of-plane translation of A-nucleobase inside the graphene semi/hybrid-nanogap: We have translated all four DNA nucleobases in both upward (+1.0 Å) and downward (-1.0 Å) directions from the initial position (0.0 Å) along the x-axis in the yz-plane inside the graphene semi/hybrid-nanogap as shown in **Figure S6**.

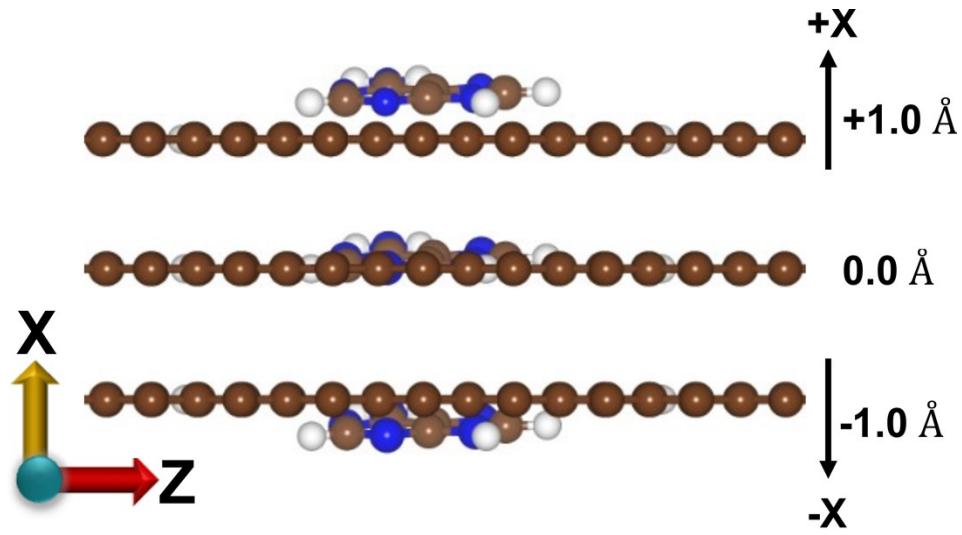

Figure S6: Scheme of A-nucleobase translated out-of-plane inside the graphene semi/hybrid-nanogap along the x-axis in positive and negative directions by ± 1.0 Å.

7. Variation in the transmission Spectra of graphene semi/hybrid-nanogap+nucleobase systems due to in-plane rotations from 0° to 180° in the step of 30º along the *x*-axis in the yz-plane:

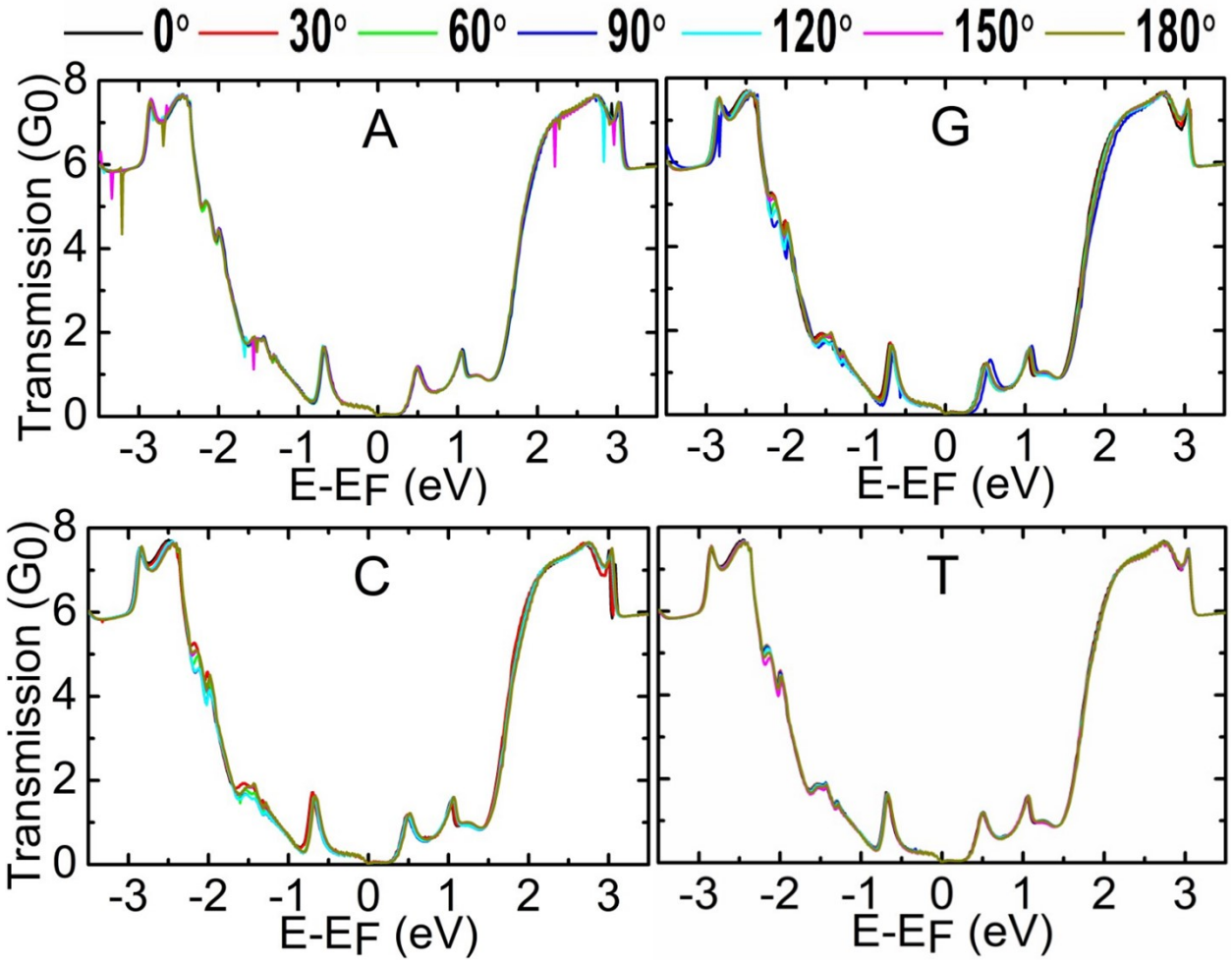

Figure S7. Variation in the transmission spectra due to in-plane rotation from 0° to 180° in steps of 30° along the *x*-axis in the yz-plane for all four nucleobases (A, G, C, T).

8. Change in the transmission function spectra of graphene semi/hybrid-nanogap+nucleobase systems due to in-plane lateral translations:

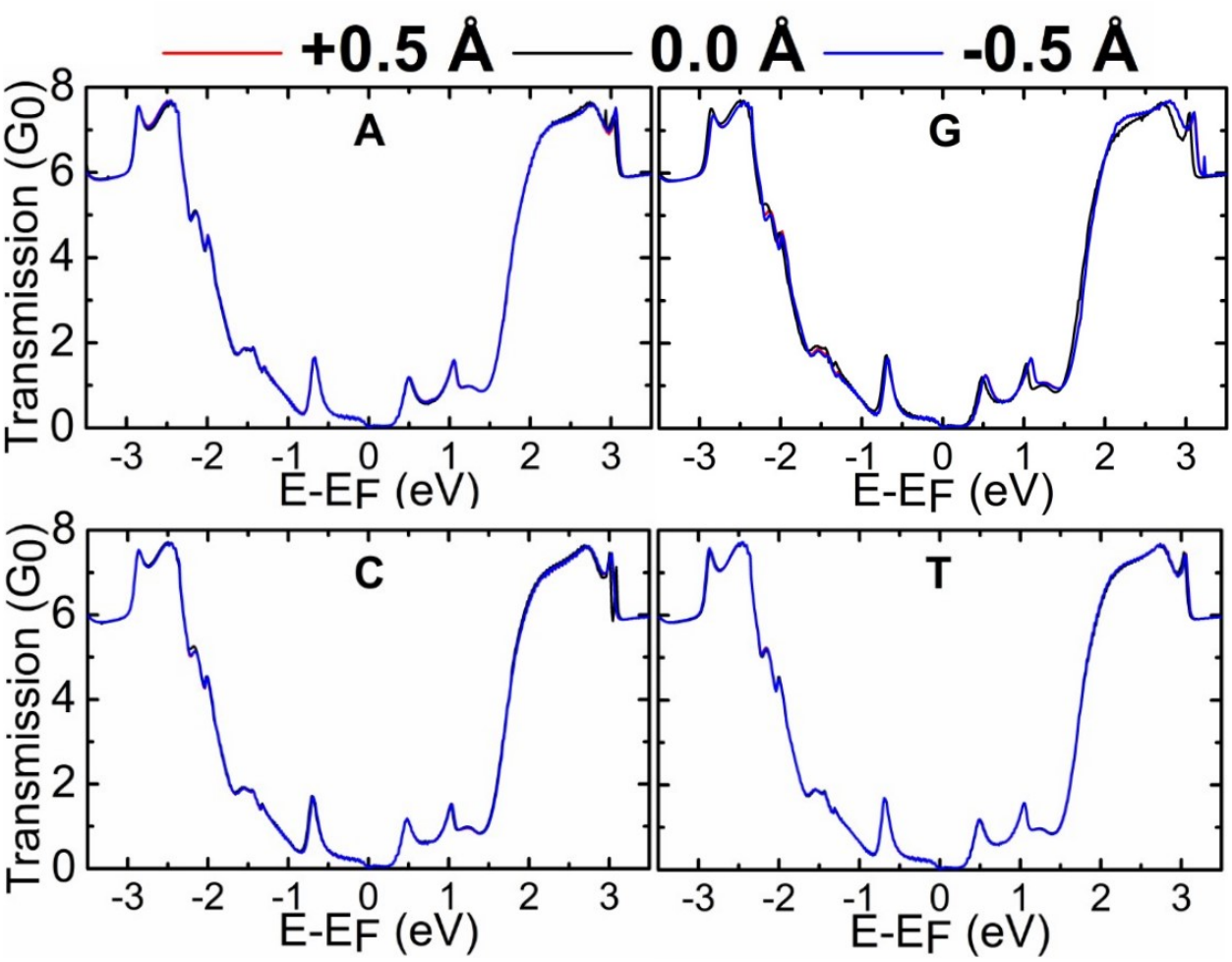

Figure S8. The change in the transmission function for each targeted nucleobase due to lateral in-plane translations (±0.5 Å).

9. Change in the transmission spectra of graphene semi/hybrid-nanogap+nucleobase systems due to out-of-plane translations:

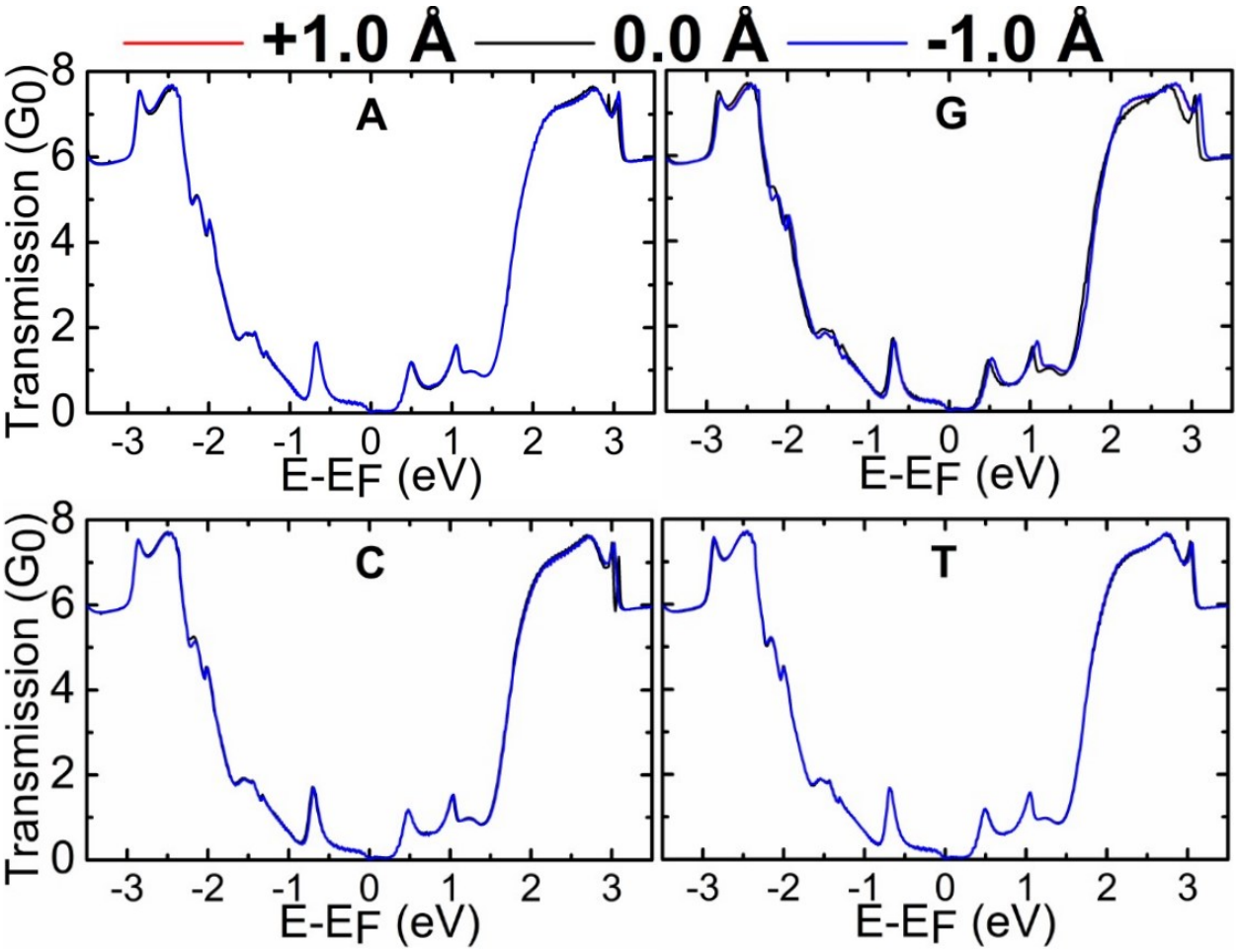

Figure S9. The change in the transmission function for each targeted nucleobase due to out-of-plane translations (±1.0 Å).