

atom name	charge	Leonard-Jones parameters	
		$\sigma$ (nm)	$\epsilon$ (kJ/mol)
C2,C4,C6	0.4498	0.34	0.360
C1,C3,C5	0.6246		
N2,N3,N5,N6,N8,N9	-0.5402	0.325	0.711
N1,N4,N7	-0.2328		
N10	0.2508		

Figure S1. Force field parameters of  $C_3N_4$ .

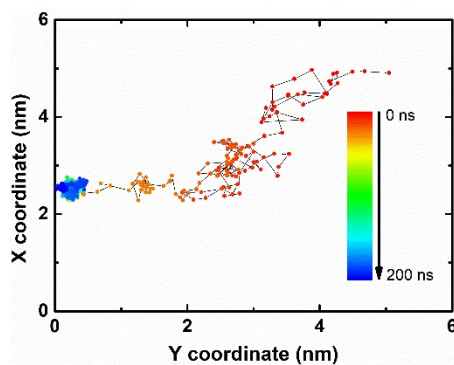


Figure S2. CoM track of the  $\lambda$ -repressor protein when bound to a non-charged  $C_3N_4$  nanosheet.

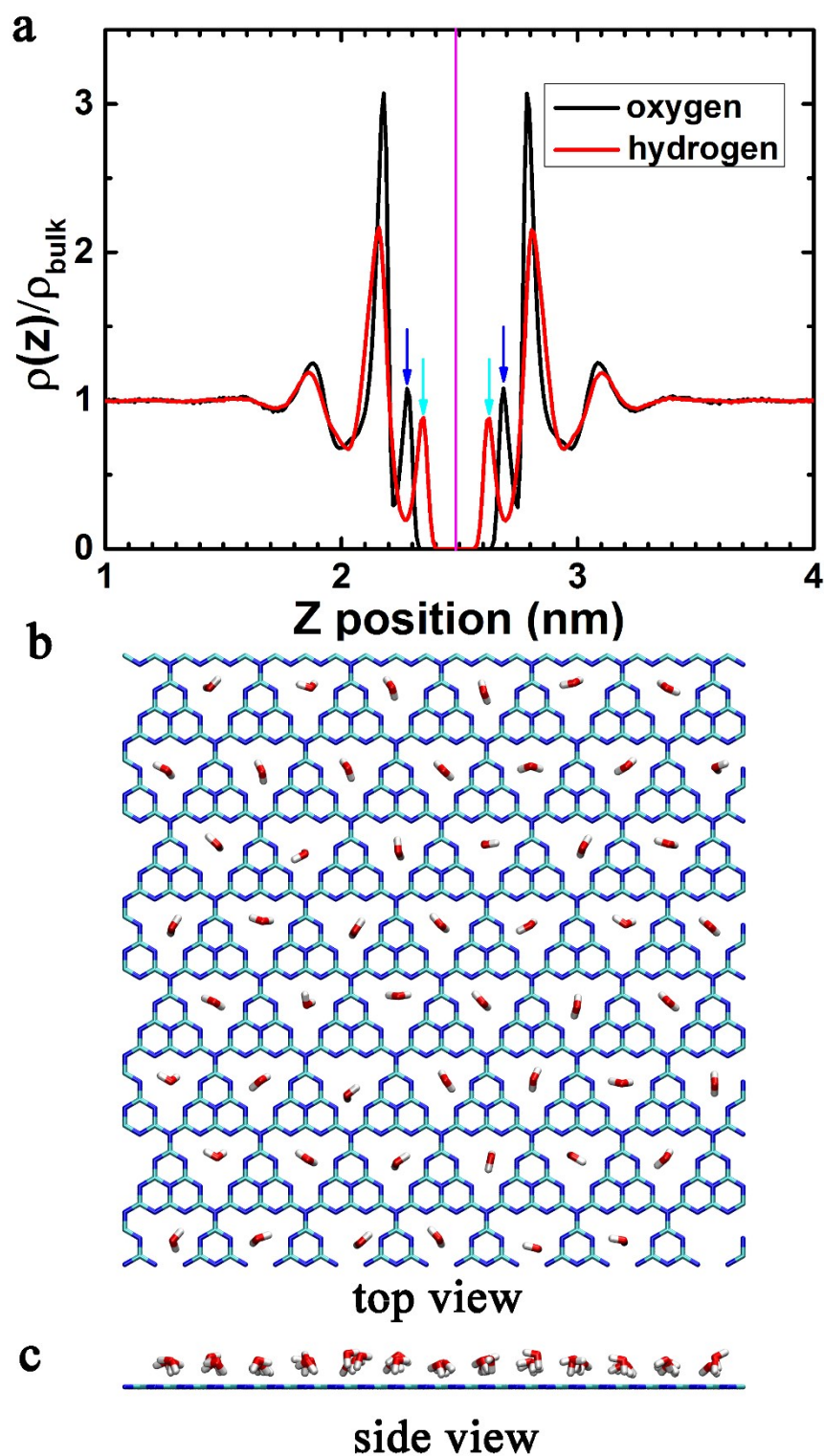


Figure S3. Water distribution on C3N4 nanosheet. (a) Axial distribution of water molecules (including the hydrogens and oxygens of waters) on C3N4 nanosheet. (b-c) Top and side views of the specific water molecule layer.

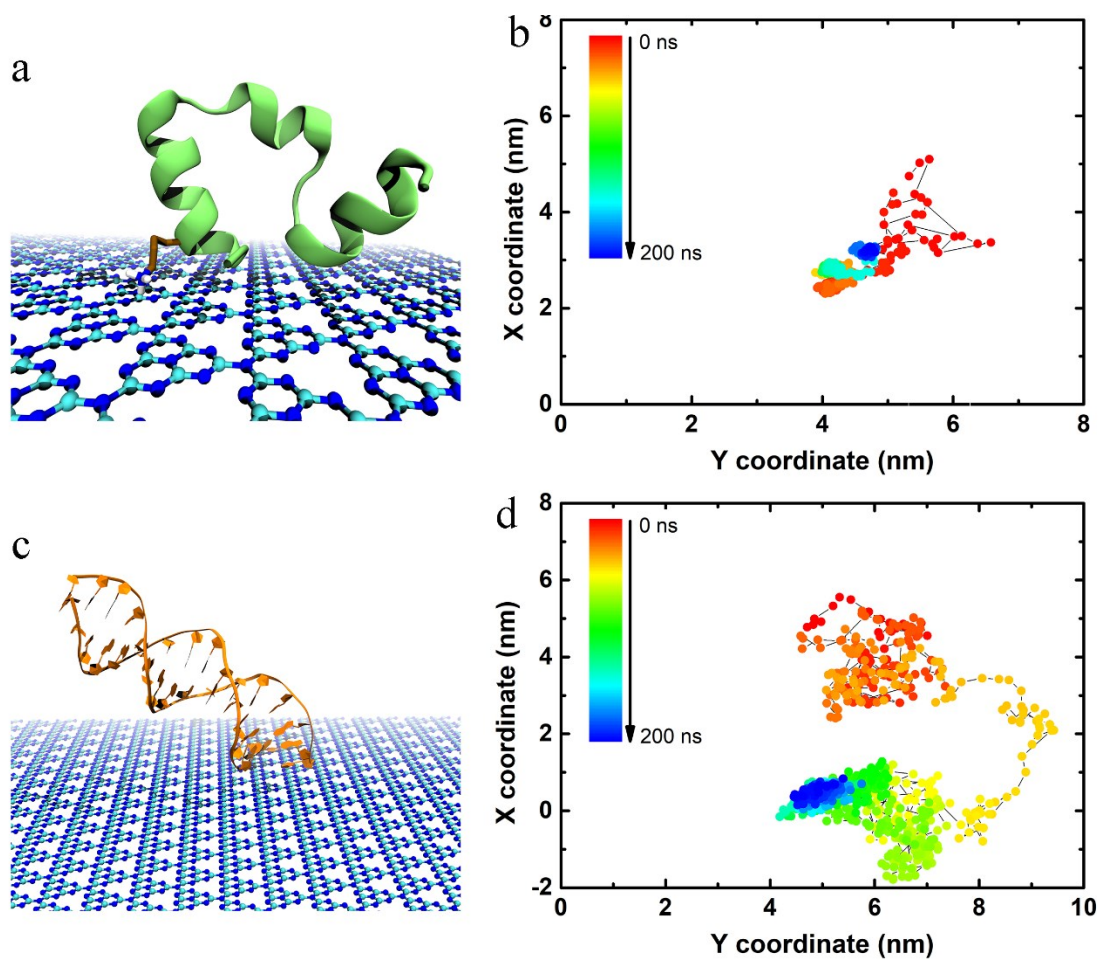


Figure S4. HP35 and dsDNA binding to C3N4 nanosheet. (a, c) The final binding conformations of HP35 protein and dsDNA to C3N4 nanosheet. (b, d) The CoM tracks of HP35 and dsDNA binding to C3N4 nanosheet.

Table S1. Structural influences of biomolecules binding onto 2D nanomaterials.

<b>2D Material</b>	<b>biomolecule</b>	<b>consequences</b>	<b>Ref.</b>
graphene	HP35 protein	potential unfolding	1
Carboxyl functionalized graphene defect	HP35 protein/WW domain	robust unfolding	2-3
graphene oxide	protein G-related albumin-binding module (GA53)	no influence	4
MoS <sub>2</sub>	HP35 protein/ $\alpha$ -helical polyalanine	limited influence	5
C <sub>2</sub> N	HP35 protein/double-stranded DNA	no influence	6-7
C <sub>3</sub> N <sub>4</sub>	$\lambda$ -repressor protein	no influence	This work
C <sub>3</sub> N	HP35 protein	robust unfolding	8
phosphorene	HP35 protein	No influence	9
silicene	HP35 protein/A $\beta$ 1-42 fibrils	No influence	10
Pt nanocrystal	HP35 protein	Unfolding on (111) facet but no influence on (100) facet	11
boron nitride	HP35 protein	potential unfolding	12

## Reference

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