Multiscale Analysis of Nanomechanics

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Outline

- Part I: Overview of calculations on Frontera
- Part II: Mechanically deformed graphene-based structures
- Part III: Water on functionalized graphene
- Conclusion

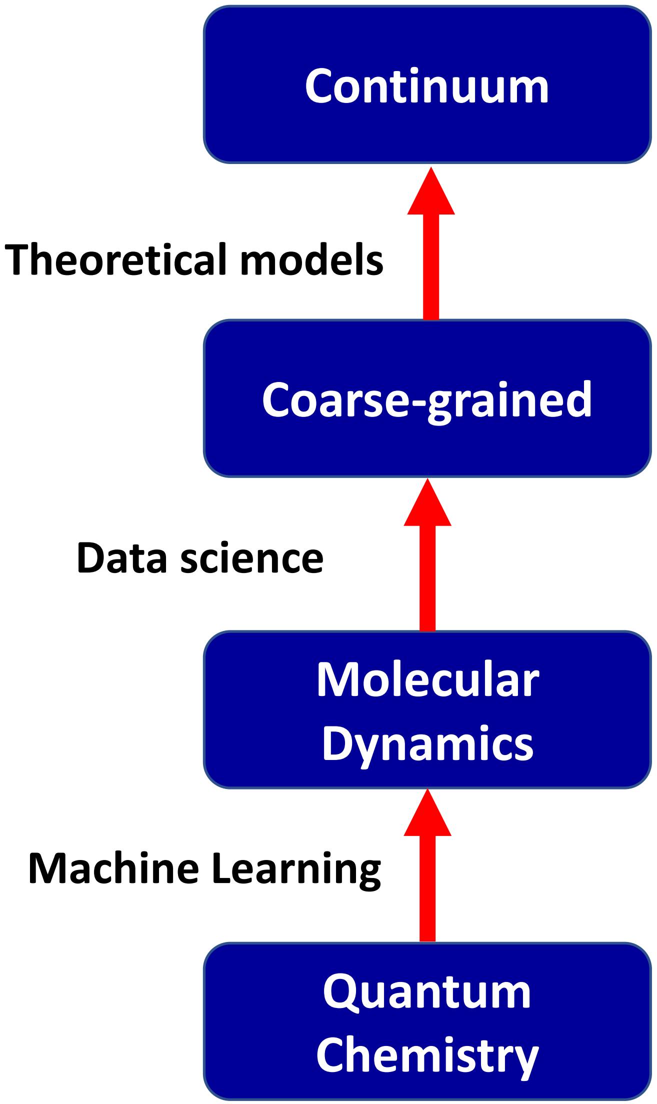
Overview

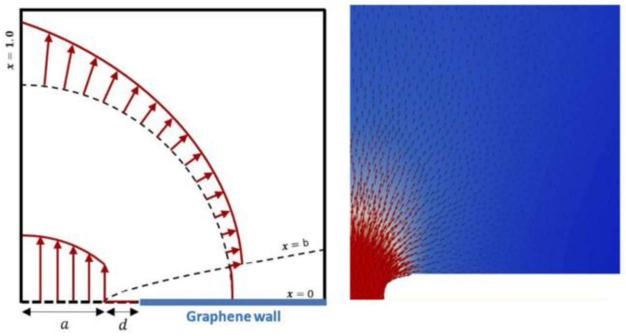
- Multidisciplinary research with focus on applications involving:

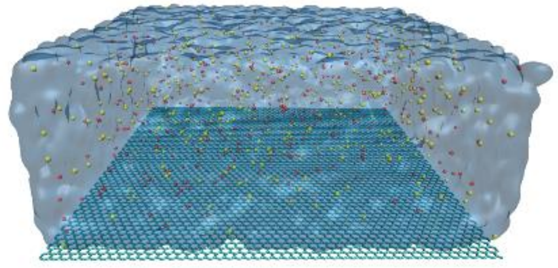
1) Energy harvesting
2) Desalination
3) Biological sensing

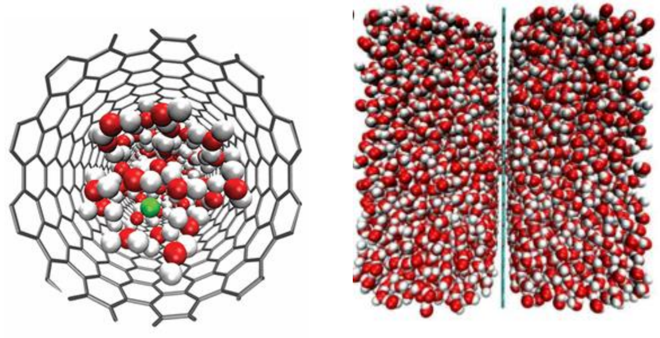
- Frontera allows us to perform complex calculations on:

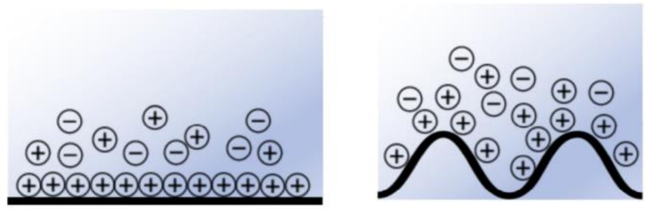
1) Nanomaterials (VASP)
2) Angstrofluidics/Nanofluidics (NAMD/LAMMPS)
3) Coupling scales (VASP)



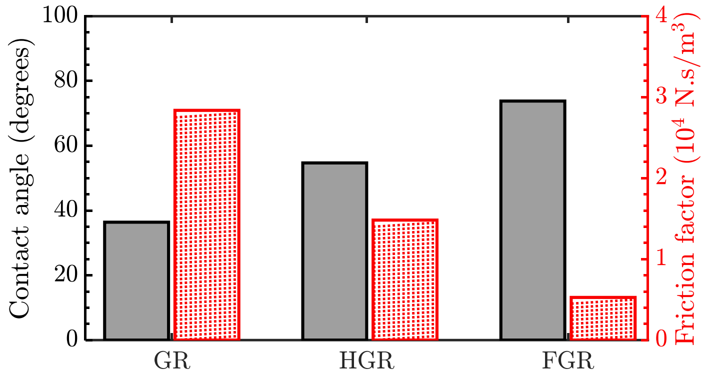








Overview



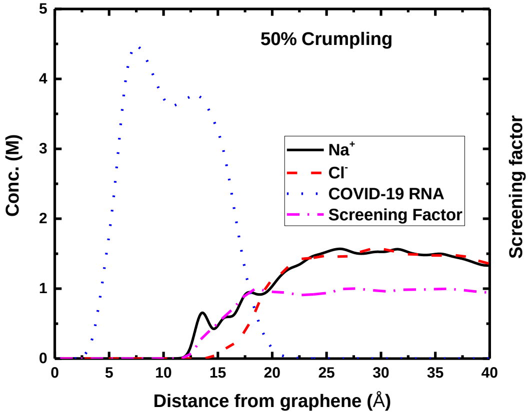
Taqieddin, Heiranian & Aluru. The Journal of Physical Chemistry C, 124 (39), 21467–21475 (2020).

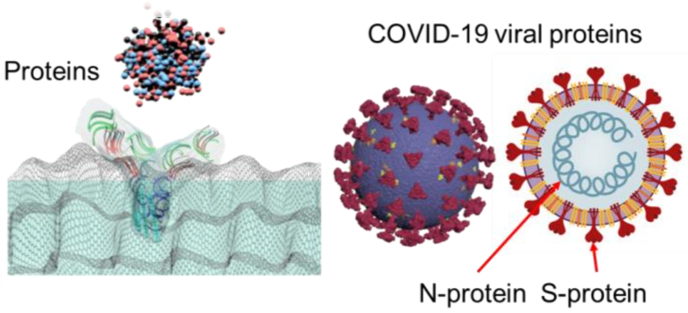
Taqieddin & Aluru, under preparation

Cho, Kang, Taqieddin, ….,Aluru & Nam. Nature Electronics 2021 (In press)

Parsons, Taqieddin,…,Aluru,… & Lyding under preparation

Cho, …, Taqieddin,…,Aluru,… & Atwater under preparation





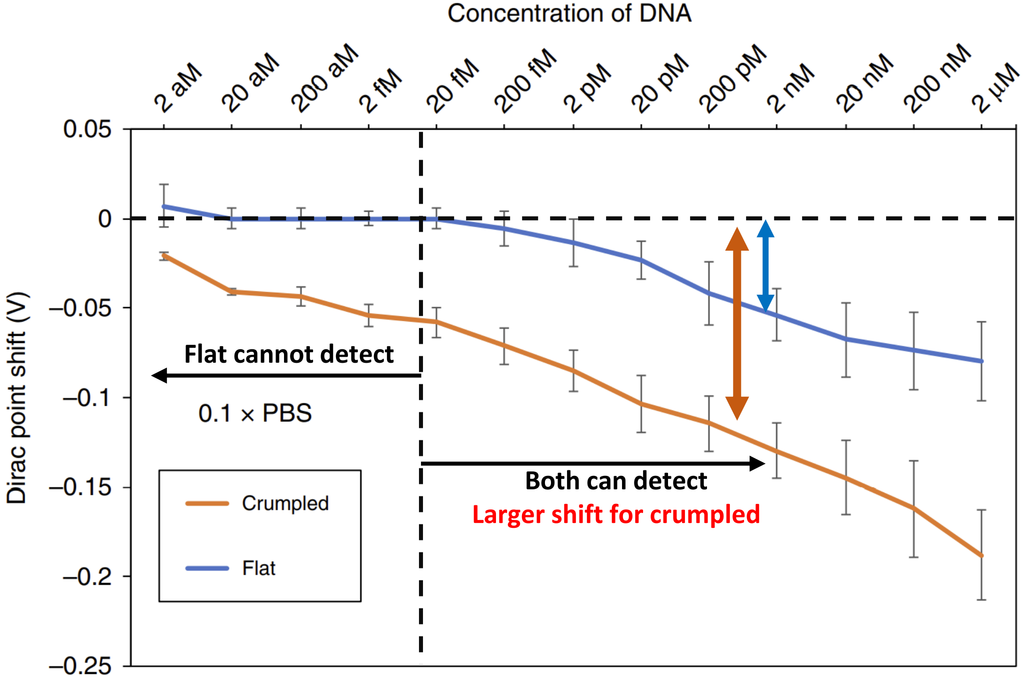
Hwang, …, Taqieddin, Aluru & Bashir under preparation

Mechanically deformed graphene-based structures

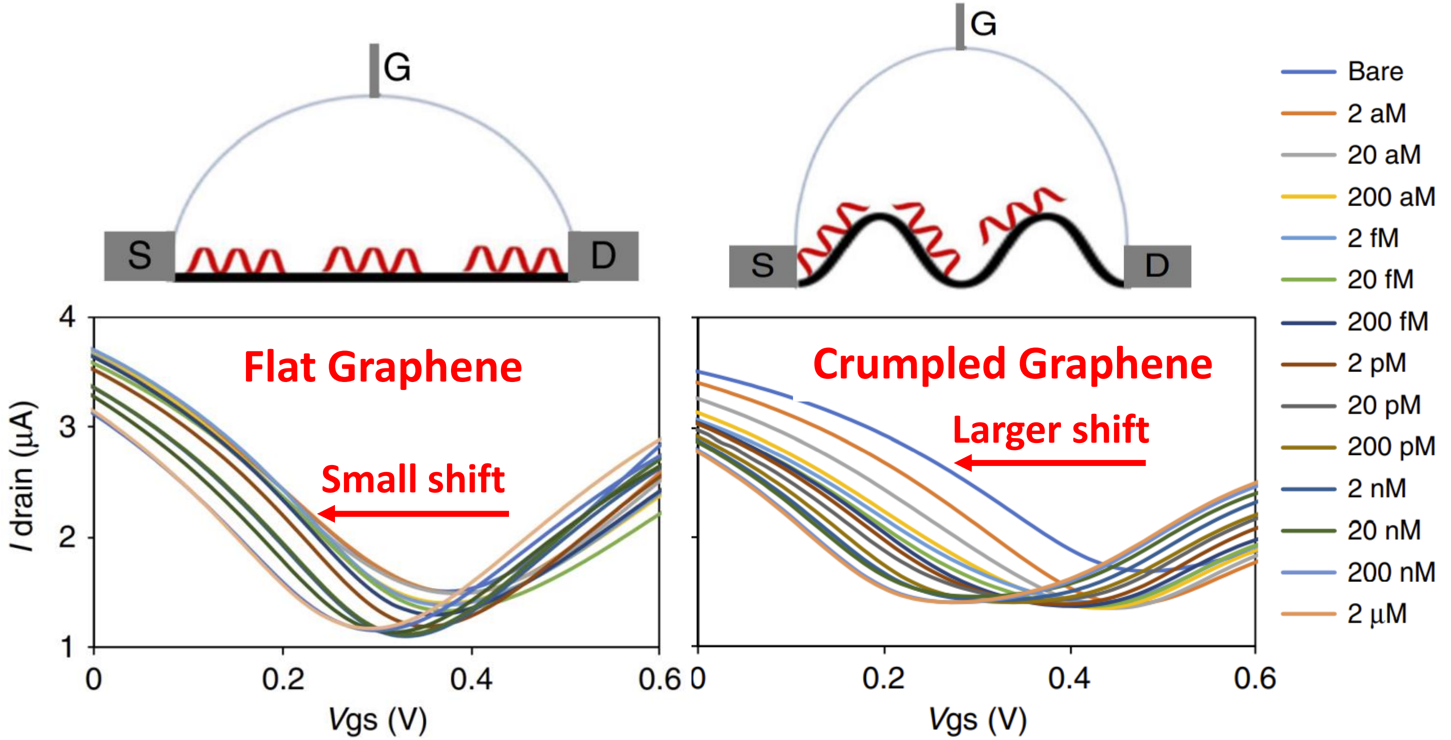
Biological detection

Deformed Graphene Structures

- DNA detection is important for disease diagnosis and environmental monitoring.
- A fast, label-free and inexpensive methods are needed.
- One big challenge: most of disease biomarkers are at ultra-low concentrations.
- This technology is about 10,000 times more sensitive than prior biosensors.



- Dirac point shift can be obtained from the change in carrier charge density of graphene.



- The threshold gate-voltage (Dirac point) shift is larger for the crumpled graphene indicating aM detection (18 molecules).
- We extended this technology to detect COVID-19.

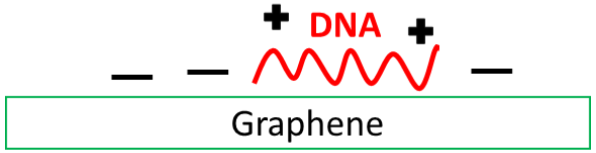
Hwang, Heiranian, Kim, You, Leem, Taqieddin, Faramarzi, Jing, Park, van der Zande, Nam, Aluru & Bashir.
Nature Communication, 11 (1), 1-11 (2020).

Deformed Graphene Structures

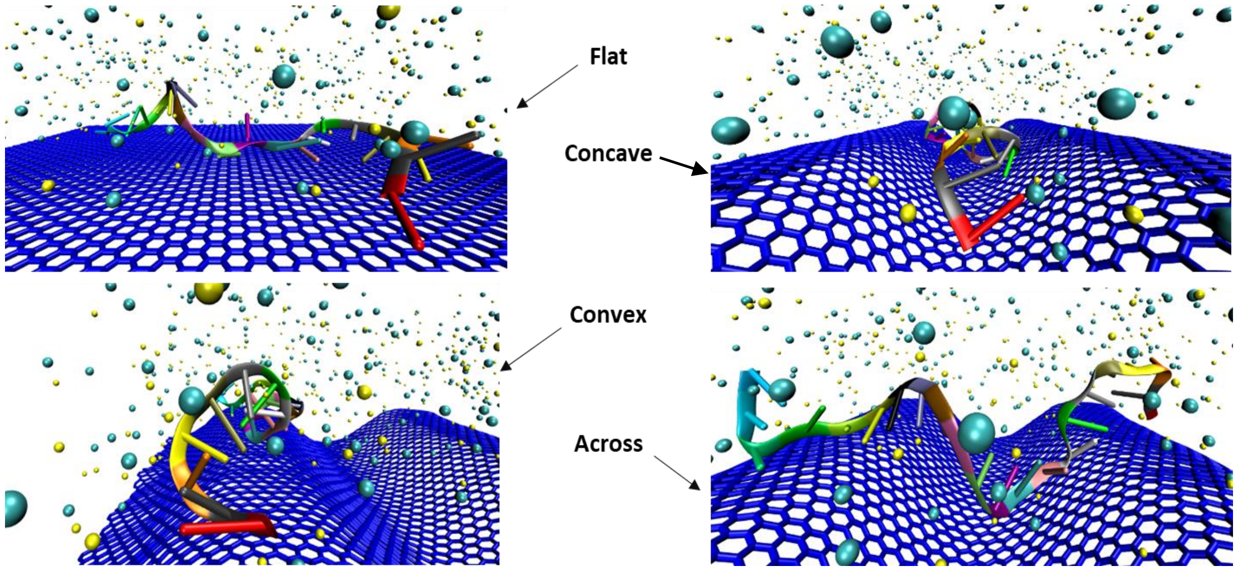
- **We worked on explain the experimental observation using *ab initio* and MD simulations.**

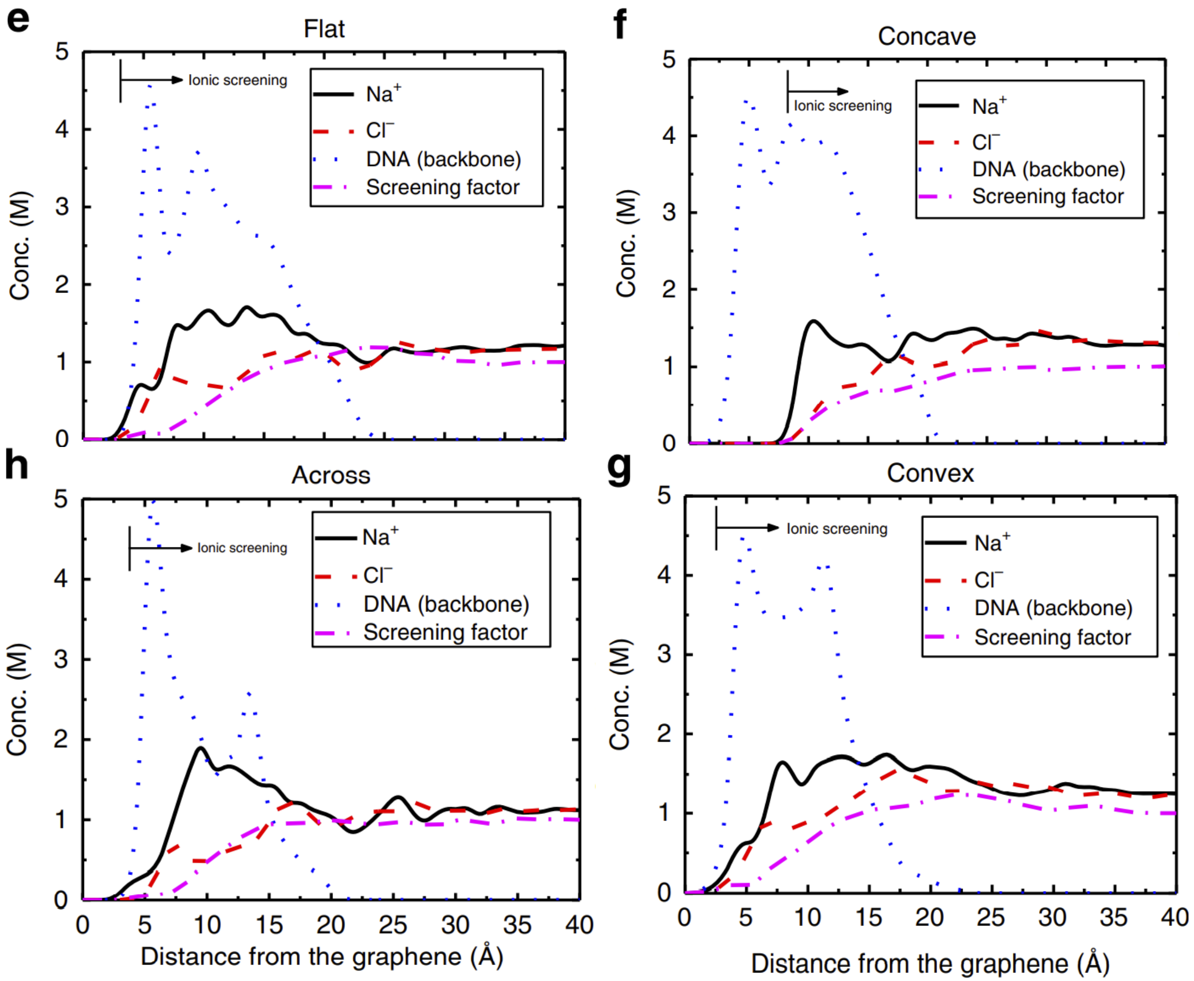
- **In presence of DNA molecules, charge transfer from DNA to graphene modifies the charge carrier density.**

$$\Delta V_{\mathrm{D}} = \frac{e\Delta n}{C_{\mathrm{T}}} = \frac{eN^{unscreened}_{charge}}{C_{\mathrm{T}}}$$



- **DNA charges are less screened by ions in crumpled graphene biosensor.**





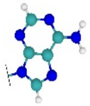
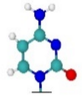
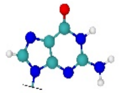
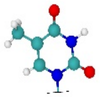
- **Ionic layer forms further away from the concave graphene because of the stronger confinement.**

- **The least ionic screening is observed in the concave region.**

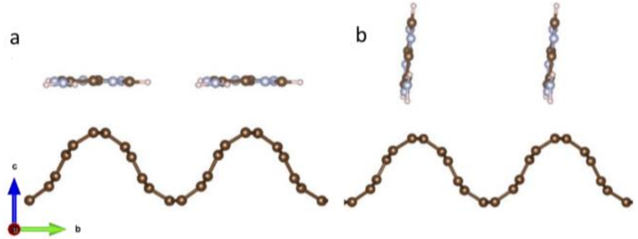
Deformed Graphene Structures

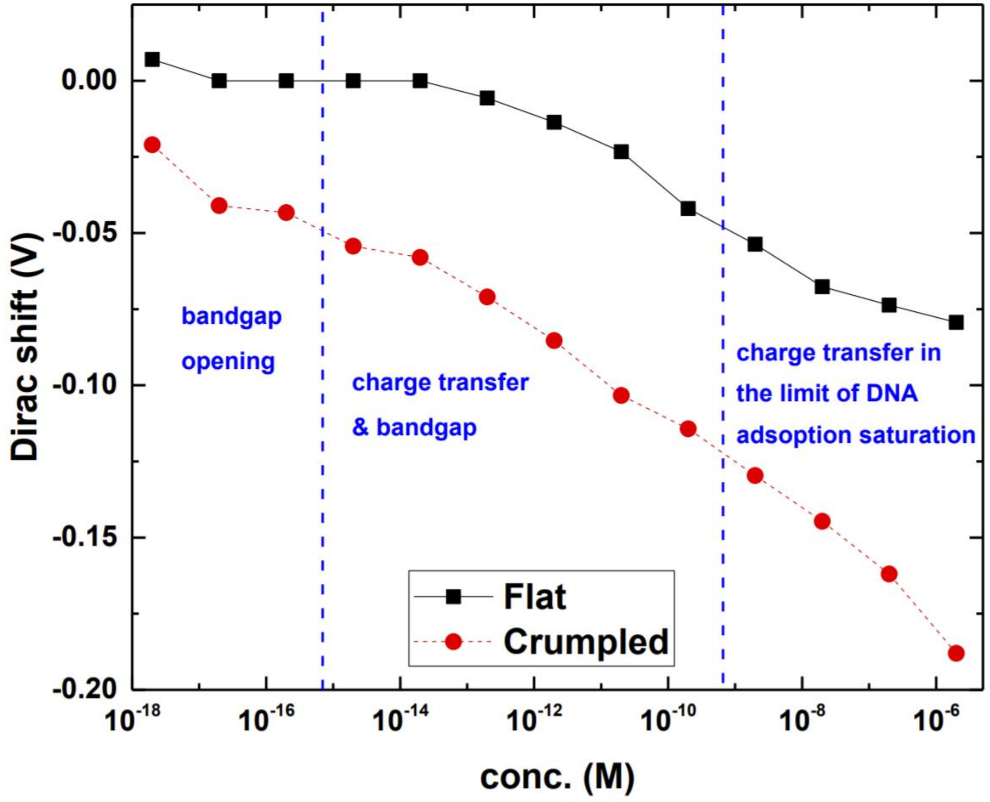
- As change in the bandgap the Dirac shift increases.

$$\Delta V_{\mathrm{D}} \propto \Delta n \propto f\left(E_{\mathrm{g}}^{\mathrm{Graphene+DNA}} - E_{\mathrm{g}}^{\mathrm{Graphene}}\right)$$

Case		Flat graphene	Crumpled zigzag graphene	Crumpled armchair graphene
No DNA		0 (GW: 0)	Metal (GW: metal)	0.1325 (GW: 0.4224)
A 	Orientation I	0.0046 (GW: 0.8535)		0.1434 (GW: 1.7641)
	Orientation II	0	Metal	0.1430 (GW: 0.5508)
	Orientation III	0		0.1491
C 	Orientation I	0.0053 (GW: 0.8568)		0.1418 (GW: 1.7493)
	Orientation II	0	Metal	0.1434
	Orientation III	0		0.1467
G 	Orientation I	0.0049 (GW: 0.8518)		0.1522 (GW: 1.7504)
	Orientation II	0	Metal	0.1481
	Orientation III	0		0.1305
T 	Orientation I	0.0054 (GW: 0.8562)		0.1485 (GW: 1.7477)
	Orientation II	0	Metal	0.1474
	Orientation III	0		0.1448

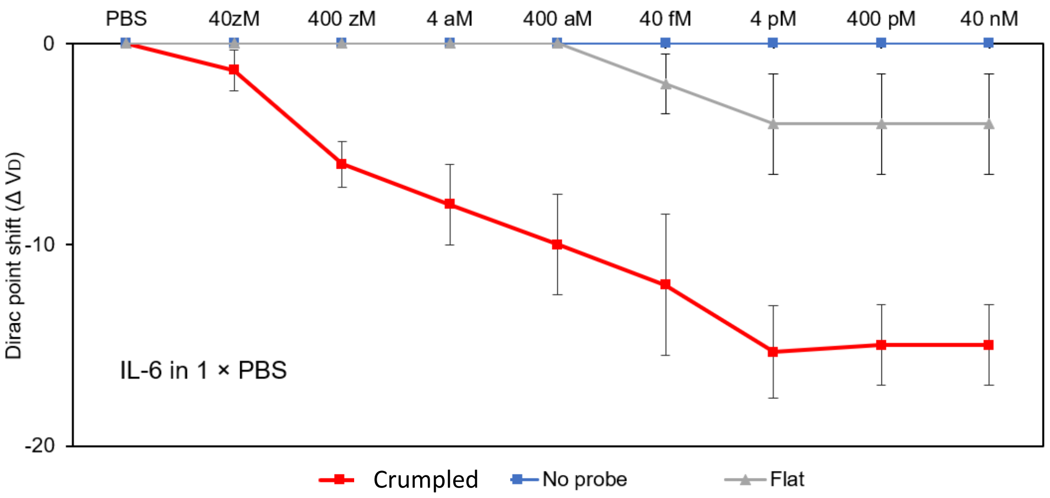
- DFT and GW simulations showed a significant bandgap change in case of the crumpled graphene.

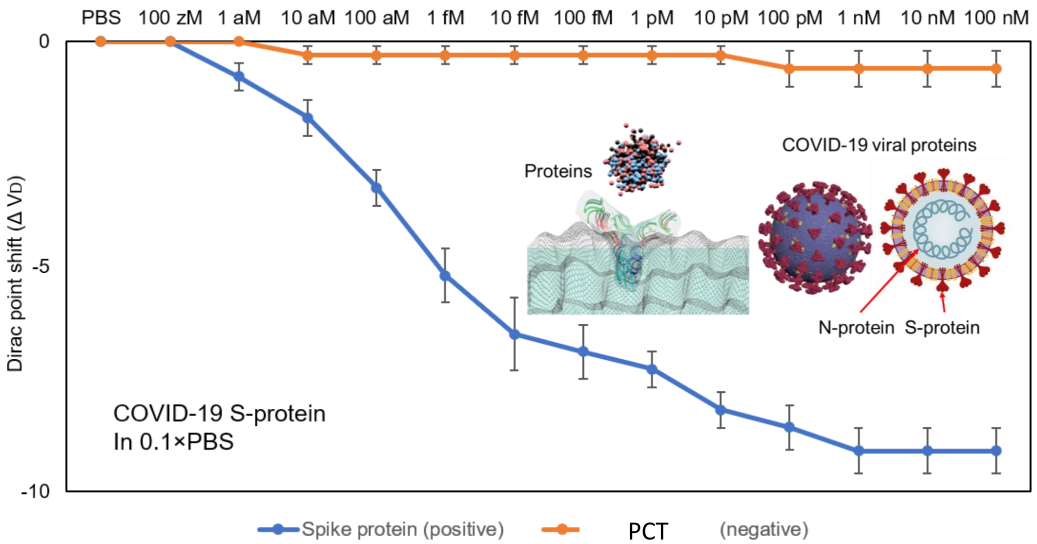




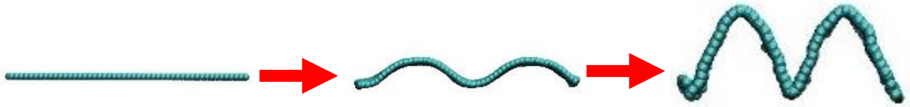
Deformed Graphene Structures

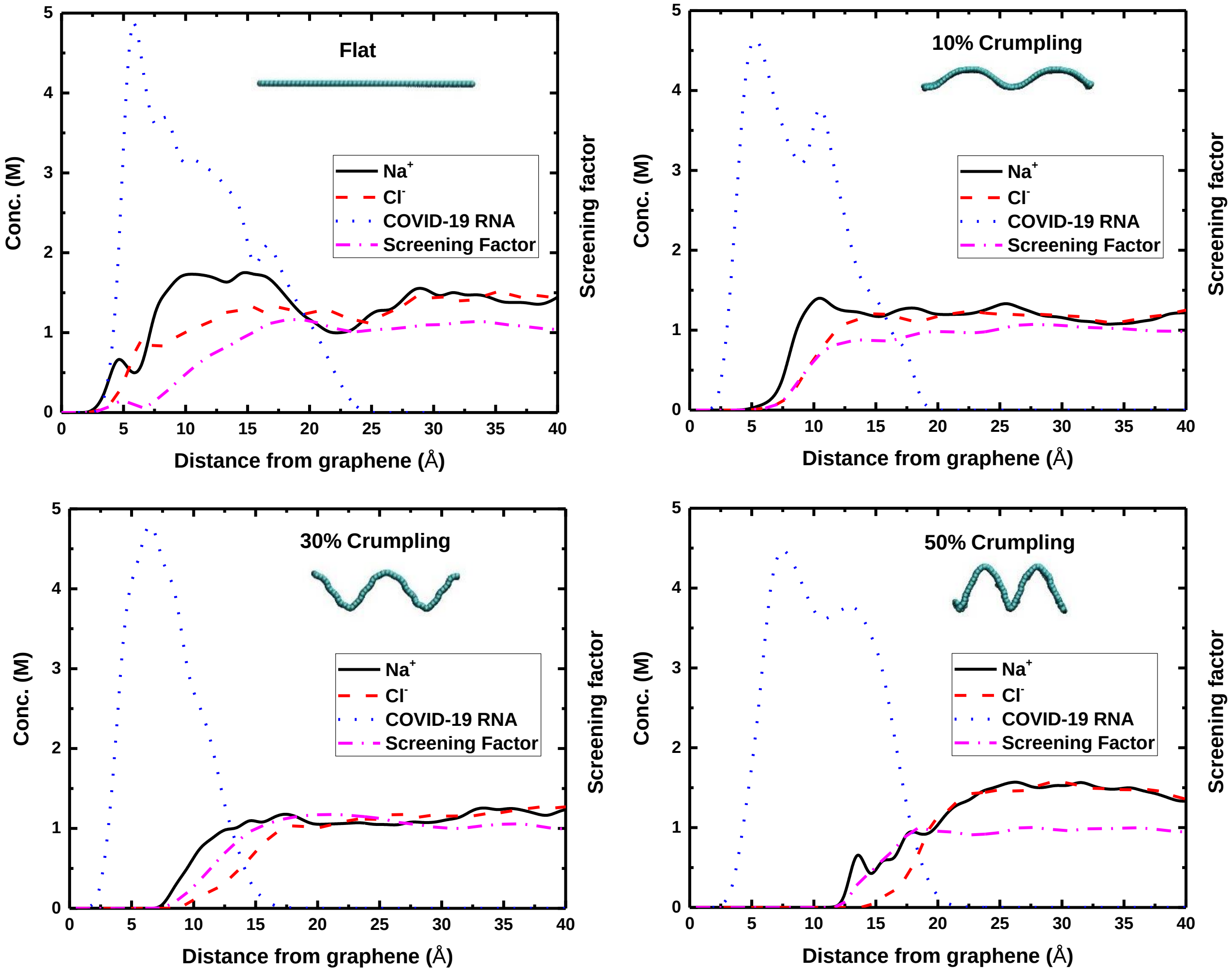
- Crumpled graphene was able to detect very low concentration of interleukin-6 (IL-6) and COVID-19 related proteins.





- Strong coupling between mechanical strain and electronics.

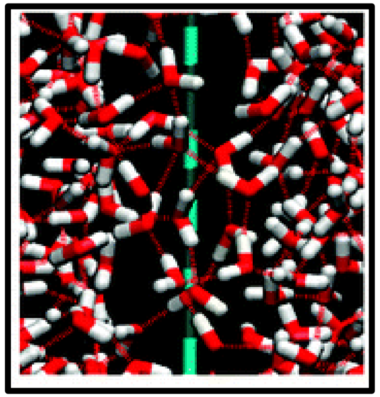




Water on functionalized graphene

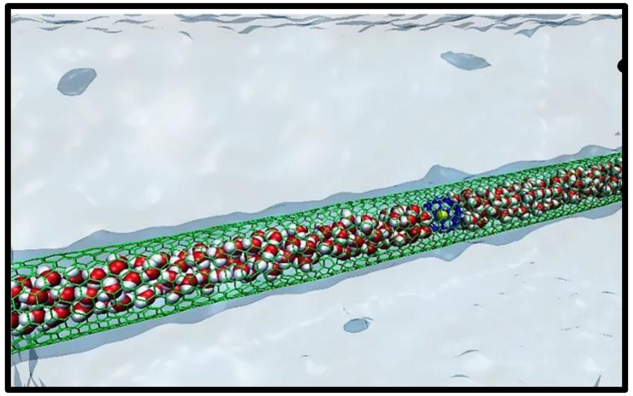
Water- Graphene

- The interactions between water and graphene are crucial in various applications.



Water desalination

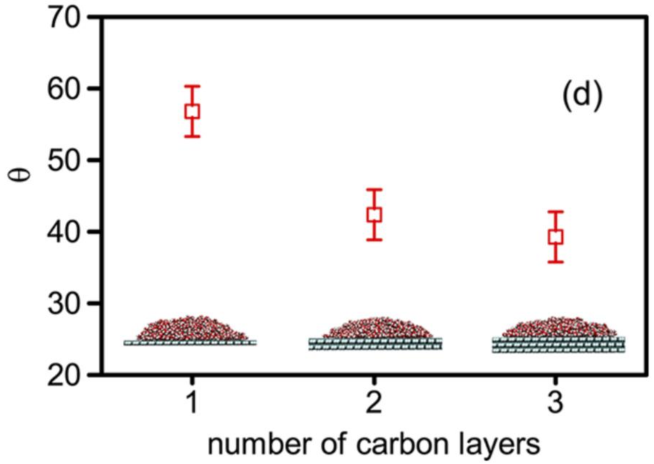
Suk & Aluru. The Journal of Physical Chemistry Letters 1 (10) : 1590-1594 (2010).



Electromechanical pumps

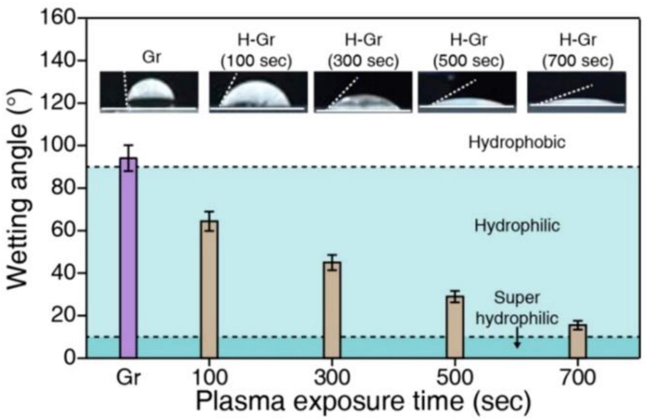
Farimani, Heiranian, & Aluru, Scientific reports, 6(1), 1-6 (2016).

- Developing accurate force field parameters is important to model the interactions between water and graphene.

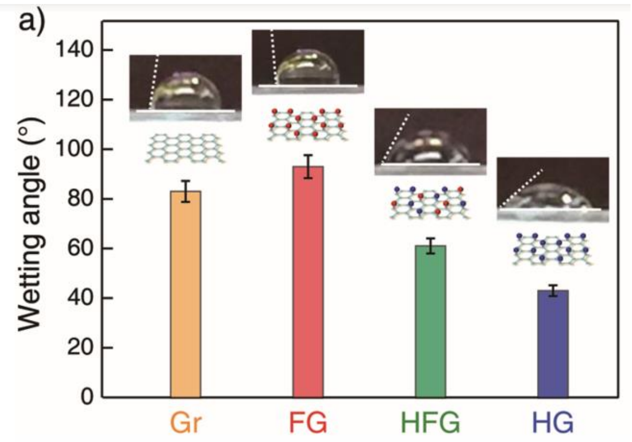


Wu & Aluru. The Journal of Physical Chemistry B, 117(29), 8802-8813 (2013).

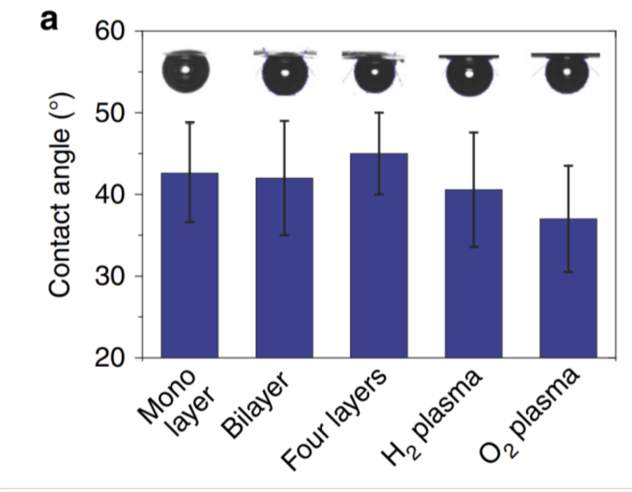
- Experimentally the wetting contact angle of water on graphene shows inconsistency.

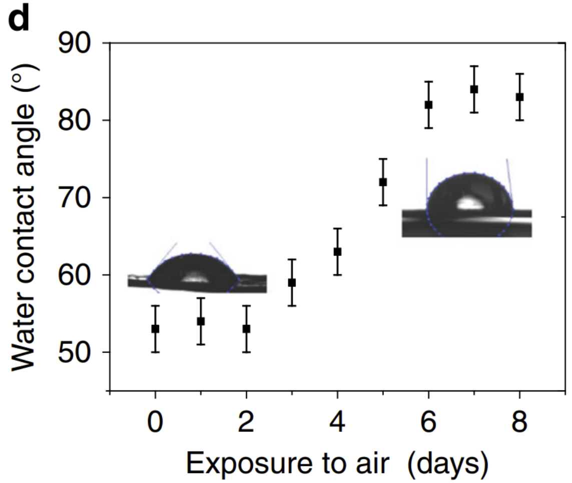


Son, Lee, Han, ... & Hong. Nano letters, 20(8), 5625-5631 (2020).



Son, Buzov, Chen, Sung,... & van der Zande. Advanced Materials, 31(39), 1903424 (2019).

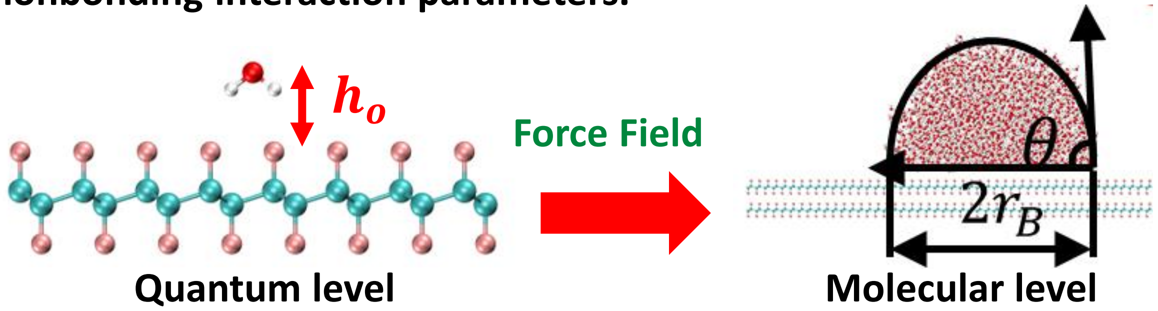




Prydatko, Belyaeva, Jiang,... & Schneider. Nature communications, 9(1), 1-7 (2018).

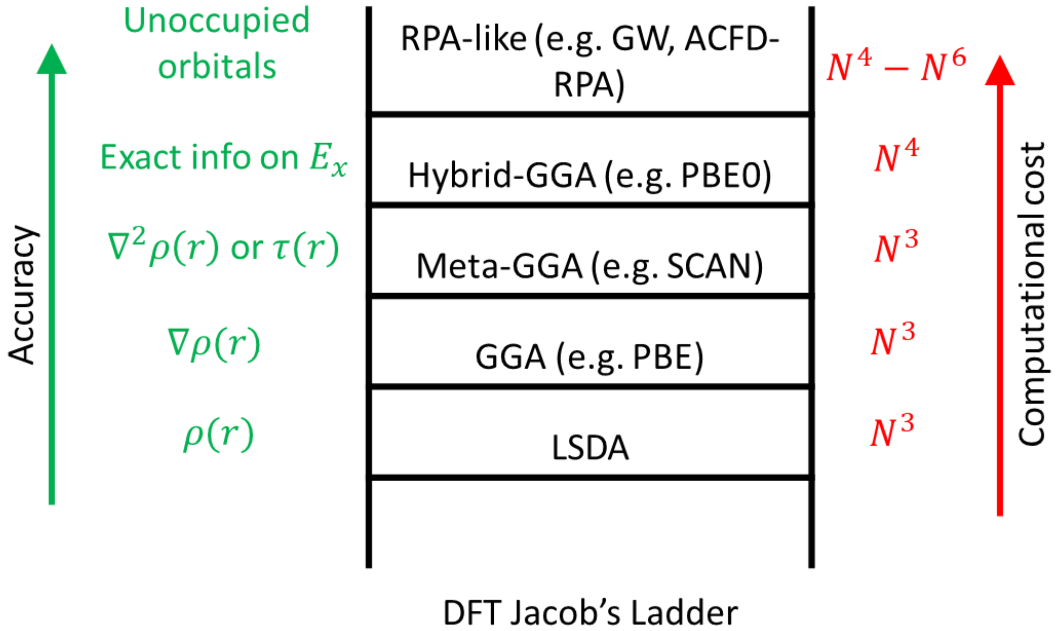
Water- Graphene

- Computational work is limited to the presence of accurate nonbonding interaction parameters.



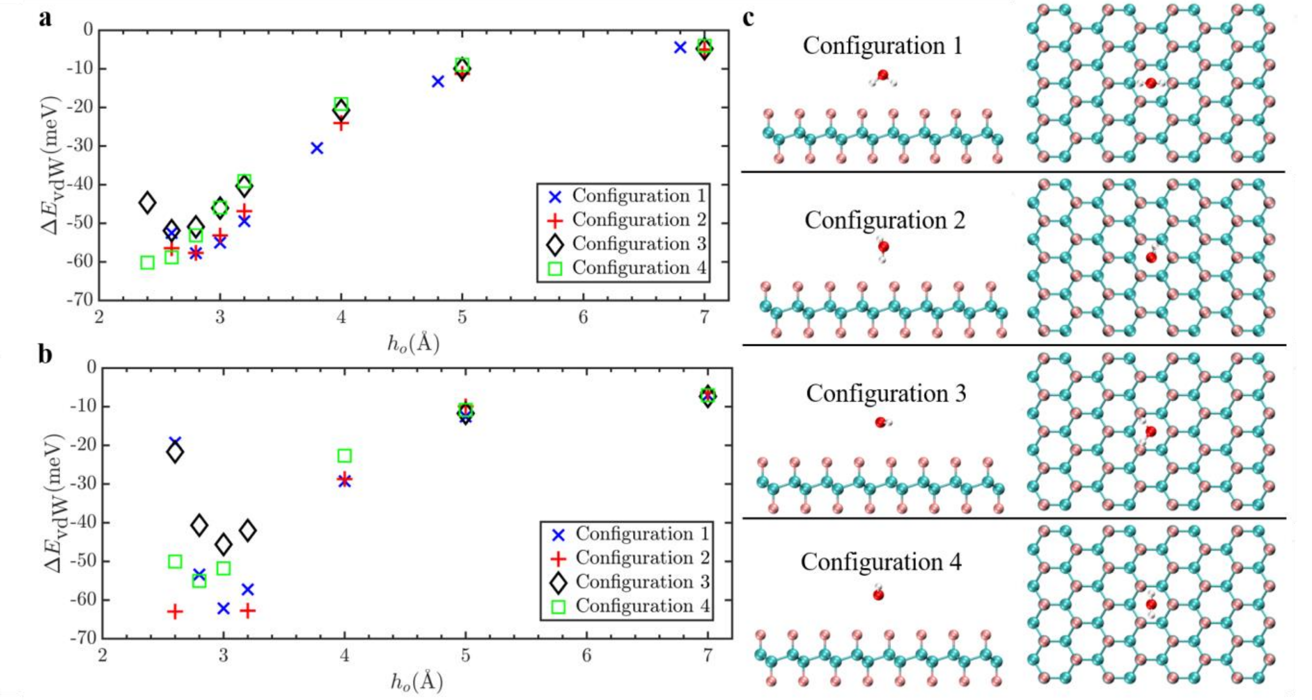
- The interaction energy between water and graphene:

$$\Delta E_{\mathrm{B}} = E_{\mathrm{water+GR}} - E_{\mathrm{water}} - E_{\mathrm{GR}} = \Delta E_{\mathrm{elec}} + \Delta E_{\mathrm{vdW}}$$



- Then fit the obtained vdW energies to Lennard-Jones potentials:

$$\Delta E_{\mathrm{vdW}} = \sum_{i \in \mathrm{GR}} 4\varepsilon_{iO}\left[\left(\frac{\sigma_{io}}{r_{io}}\right)^{12} - \left(\frac{\sigma_{io}}{r_{io}}\right)^{6}\right]$$



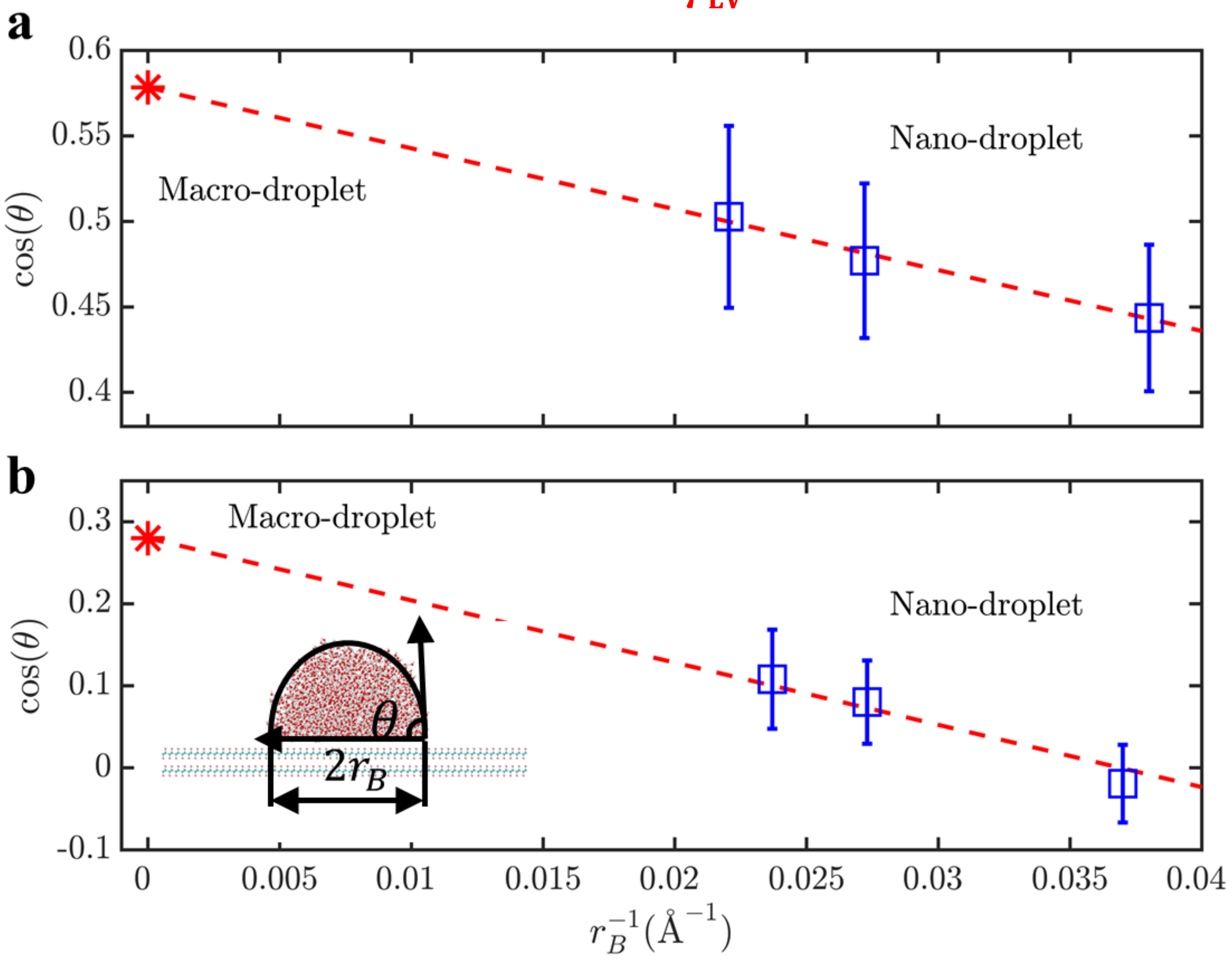
Graphene surface	σ_{C-O} (Å)	ε_{C-O} (kcal /mol)	q_c (e)	σ_{i-O} (Å)	ε_{i-O} (kcal /mol)	q_i (e)
Hydrogenated ($i = H$)	3.436	0.0850	0.00	3.773	0.0607	0.00
Fluorinated ($i = F$)	3.436	0.0850	0.56	4.231	0.0535	-0.56

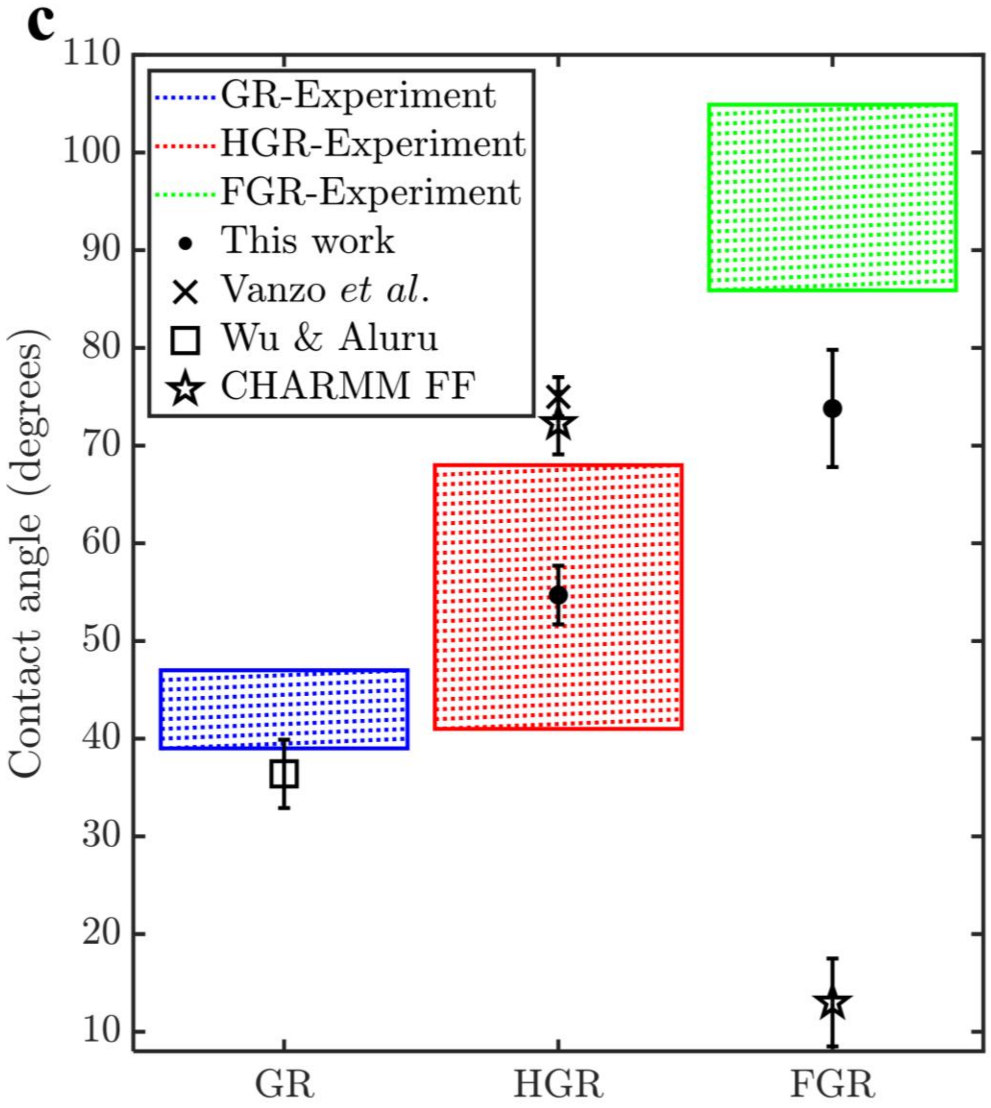
Water- Graphene

- Using Young's equation, we extrapolate the macro-droplet contact angle (equivalent to experimental angle):

$$\cos(\theta_\infty) = \cos(\theta) - \frac{\tau}{\gamma_{LV}} r_B^{-1}$$

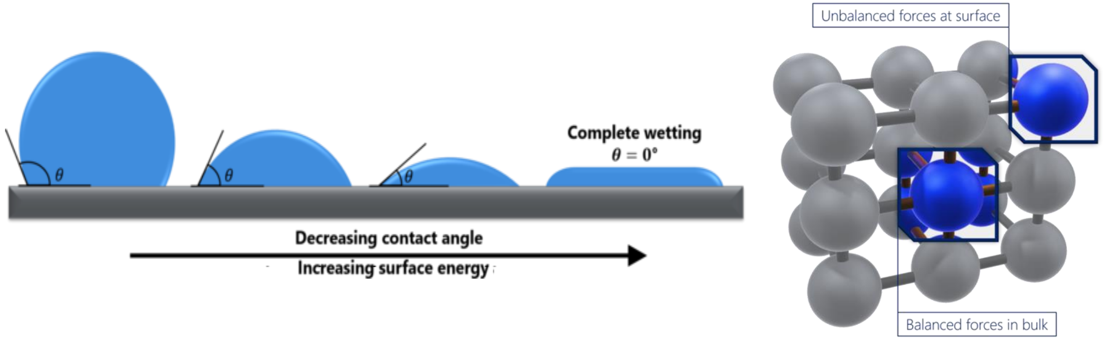
- The average macro-droplet contact angle for GR, HGR, and FGR is 36.2, 54.7, and 73.8 degrees, respectively.

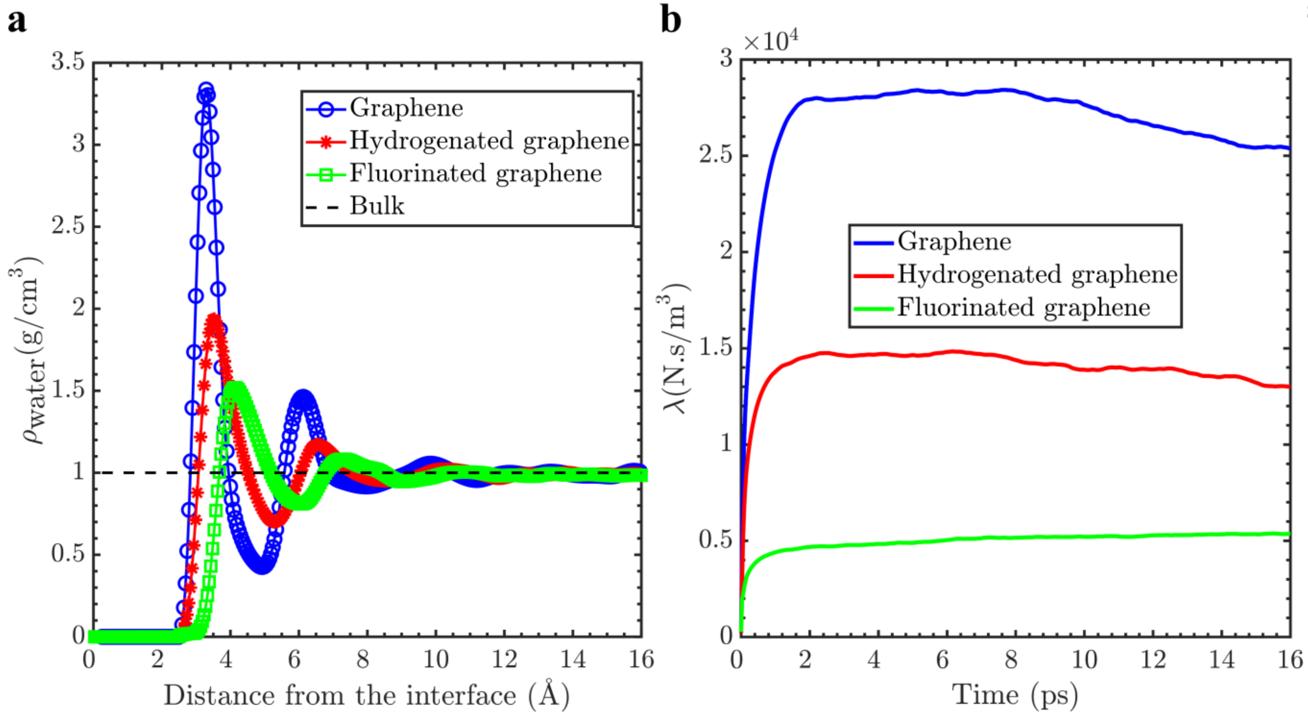


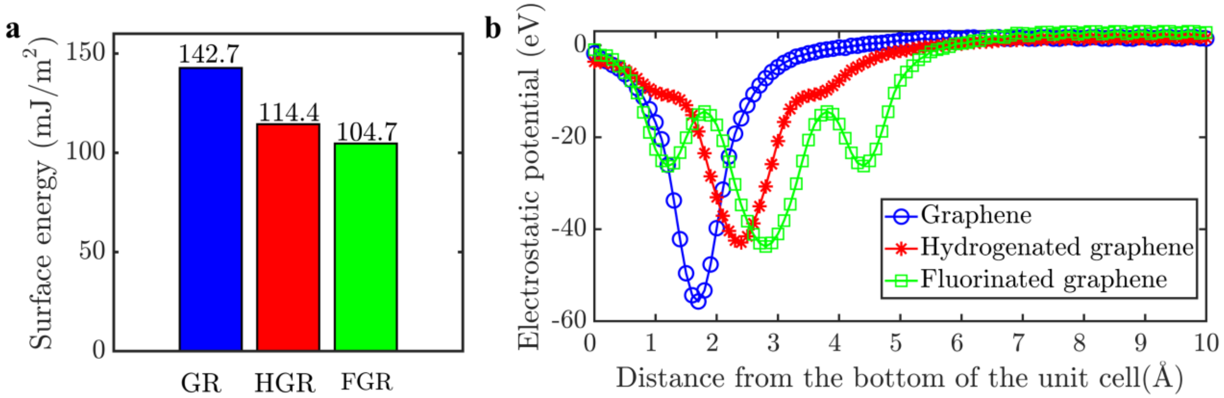


Water- Graphene

- The computed interfacial density and friction factor follow the contact angle trend ($\theta_\infty^{FGR} > \theta_\infty^{HGR} > \theta_\infty^{GR}$).
- A slip length of **138.3, 49.2, and 25.7** nm is observed on the interface of **FGR, HGR, and GR**, respectively.







- The obtained surface energies support the computed wetting behavior using MD.
- Finally, the electrostatic potential profile shows graphene has largest negative energy (attractions) compared to the other surfaces.

Conclusion

- Our simulations focus on investigating multiscale and multi-physics problems in nanotechnology applications.
- We developed a crumpled graphene biosensor which is about 10,000 times more sensitive than prior biosensors and extended the technology to detect COVID-19.
- We investigated the coupling between mechanics and electronics in the crumpled graphene.
- We developed accurate force field parameters to describe the interactions between water and functionalized graphene.
- Our developed parameters show that chemical modification makes graphene more hydrophobic ($\theta_{\infty}^{\mathrm{FGR}} > \theta_{\infty}^{\mathrm{HGR}} > \theta_{\infty}^{\mathrm{GR}}$).

Thank you!