Graphene-Water Non-Bonded Interaction From First Principles

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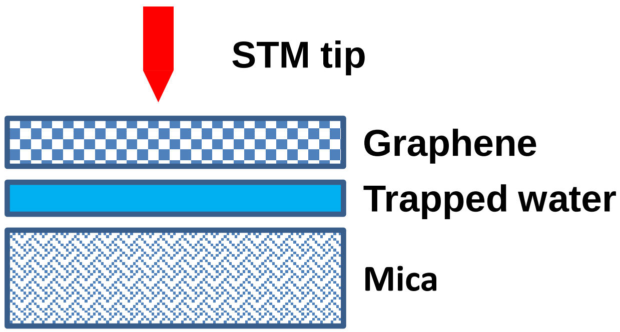
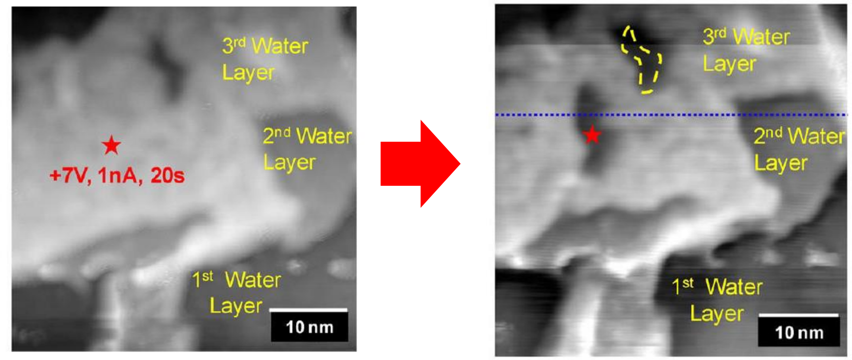
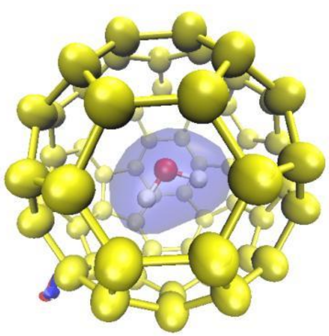
Control Water at Nanoscale

Nanoscale water interfaces

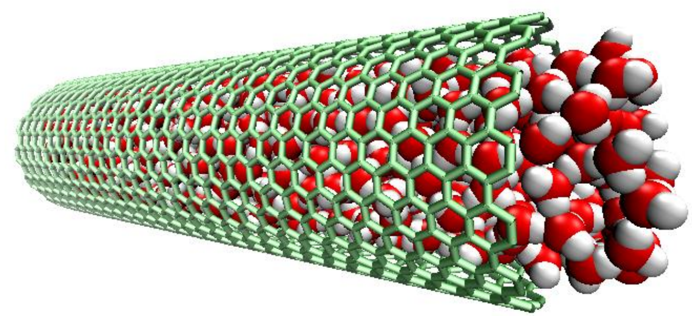




Manipulating trapped water using STM tip: Reduce water layer thickness locally by applying a voltage bias (He, et al. Nano Lett. 2012)



Fast water rotation inside C_{60} (Kurotobi and Murata, Science 2011)

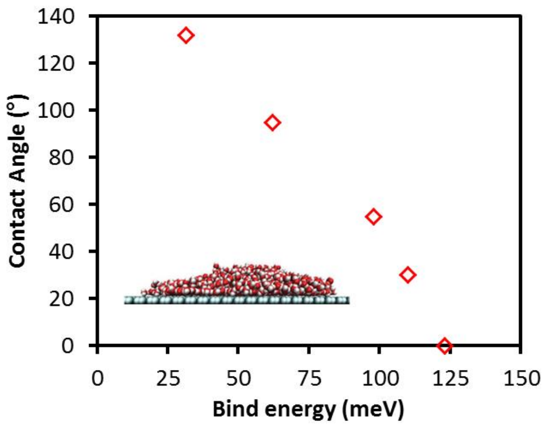


Fast water transport in carbon nanotube (Holt et al. Science 2006)

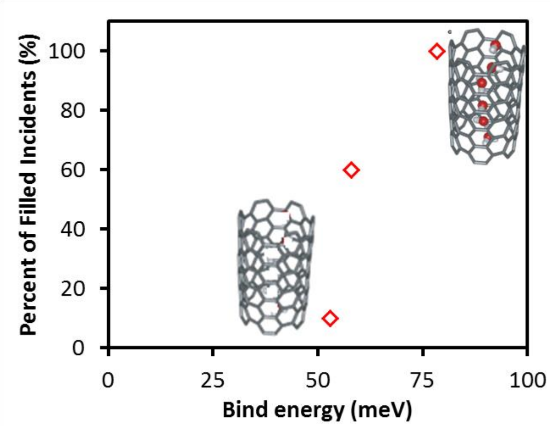
We'd like to have control of these interfaces for design in applications like desalination, sensing, nanomanufacturing, etc.

We Don’t Know How Water Interacts with Carbon

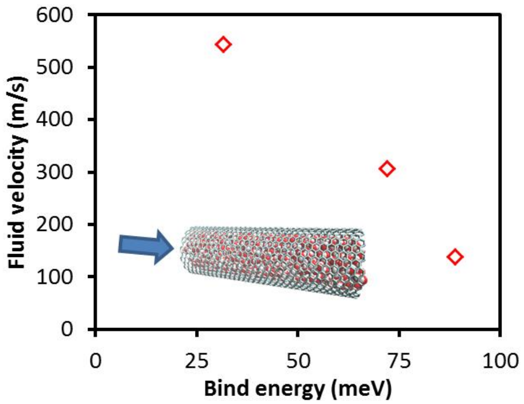
Change in carbon-water interactions leads to different properties



Water contact angle on graphene

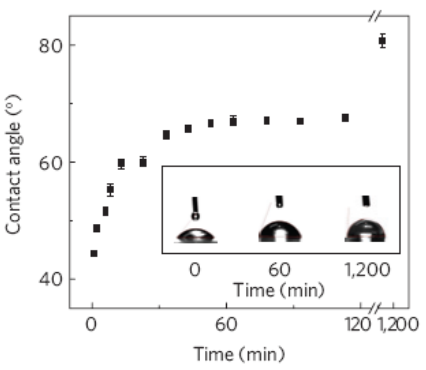


CNT filling behavior



Fast water transport through CNT

Determining carbon-water interaction from experiments is challenging



Change in water contact angle on graphite with time when exposed to air (Li et al. Nature Material 2013)

Contact angle measurement on clean and defect-free graphene/graphite surface is difficult.

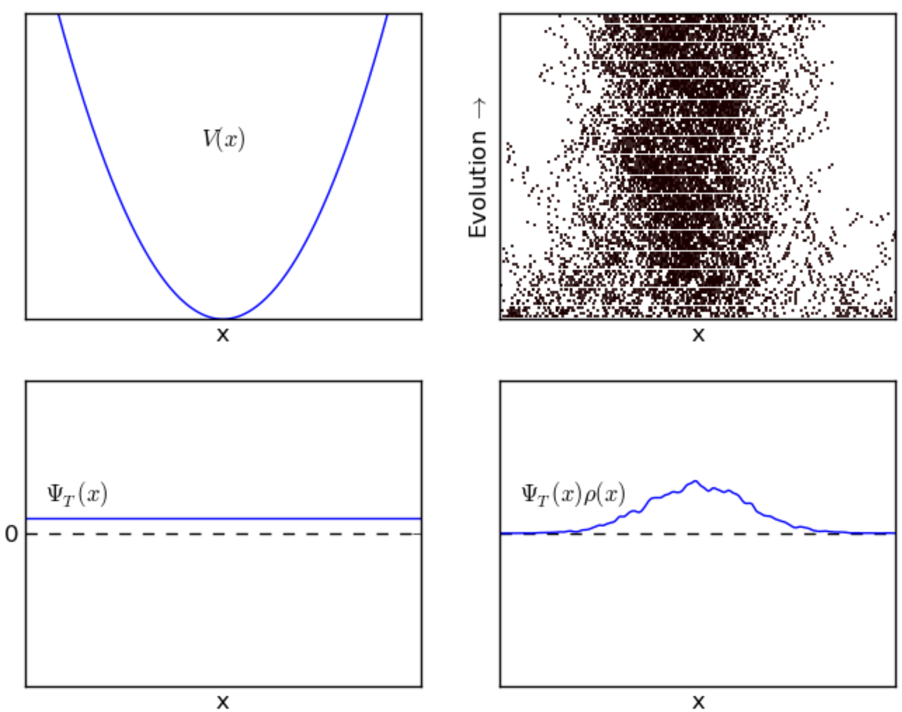
Solve Many-body Schrödinger Equation

Target $H\psi(X) = E\psi(X)$ → **Solve a different equation in Diffusion Monte Carlo (DMC) method** $-\frac{\partial\varphi(X,t)}{\partial t} = (H-E)\varphi(X,t)$

$$H = -\frac{\hbar^2}{2m_e}\nabla^2 + V \equiv \hat{T} + \hat{V}$$

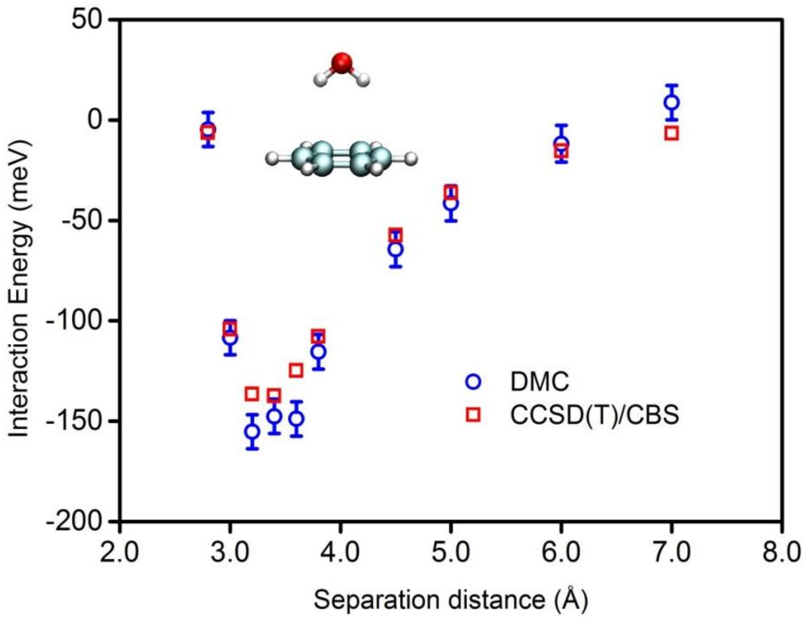
when $t \to \infty$, $\varphi(X,t) \to \psi(X)$

Isomorphism between Schrödinger equation and stochastic process
Wavefunction ↔ Distribution of walkers
Kinetic ↔ Diffusion
Potential ↔ birth/death



Example: harmonic oscillator

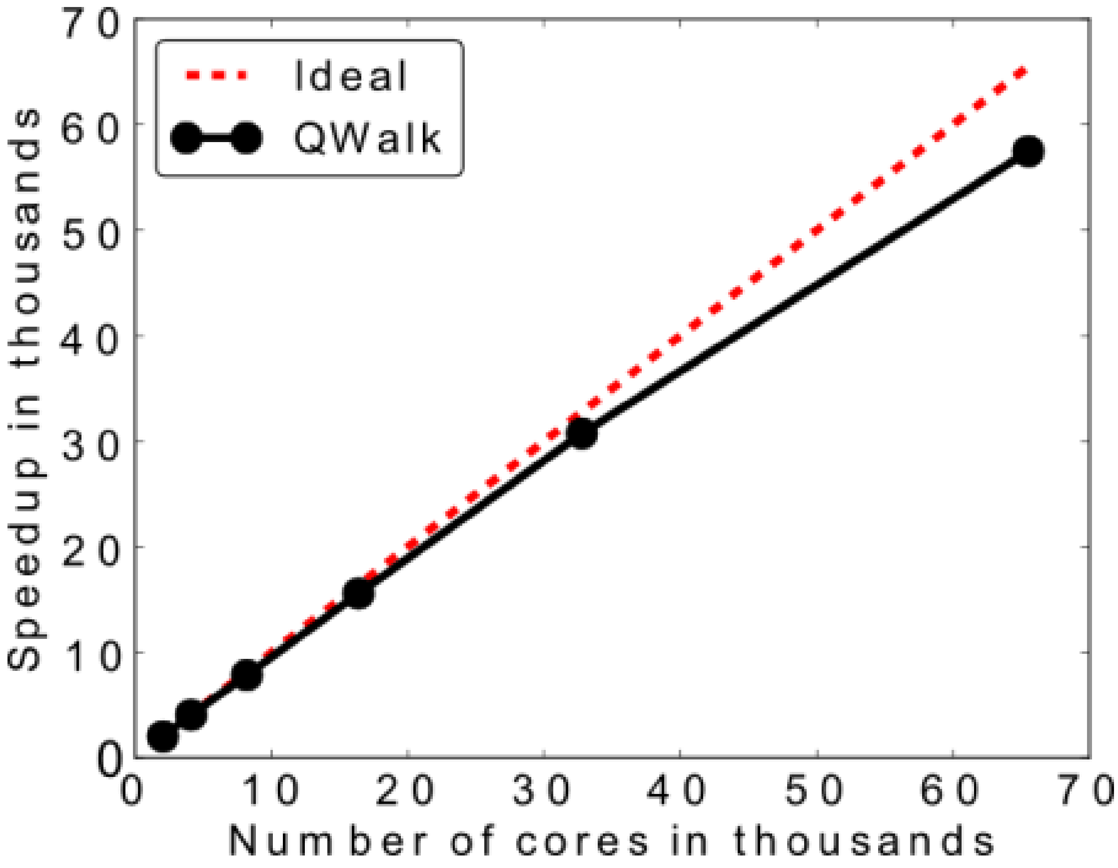
Verify and calibrate DMC method



The DMC method is verified and calibrated by comparing to couple cluster method with complete basis set (CBS) for a small system. Qwalk package is used.

Utilizing the Power of Blue Waters

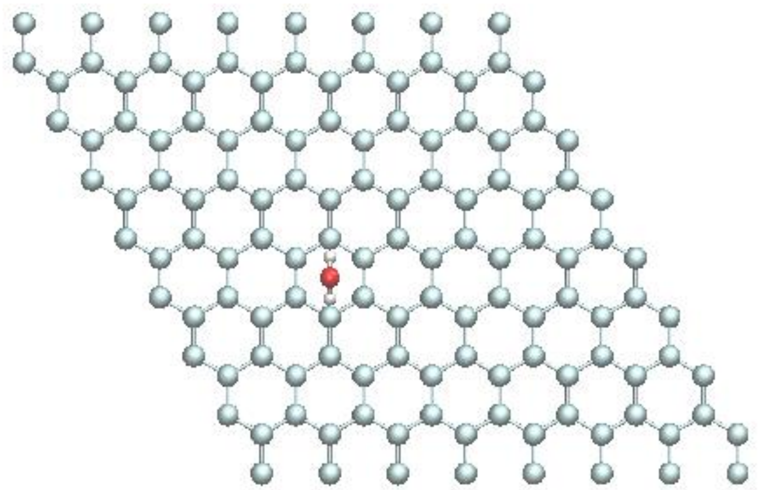
One graphene-water energy point cost 200,000 core hours.

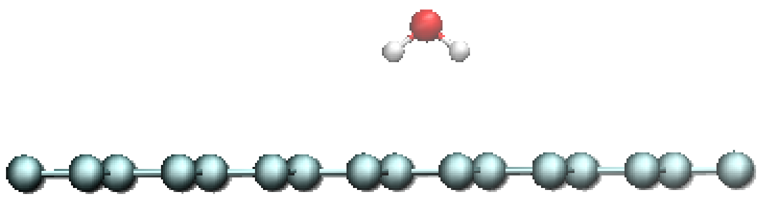


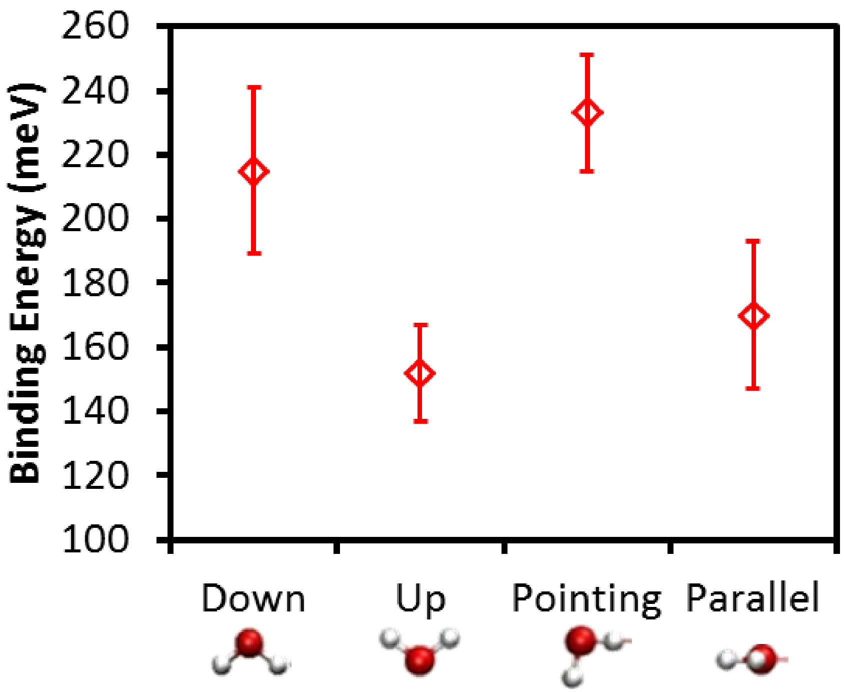
Scaling of Qwalk with number of processors

Our simulation jobs that used to take weeks on other systems can now be done within days or even hours on Blue Waters.

Graphene-Water Interaction is Strong





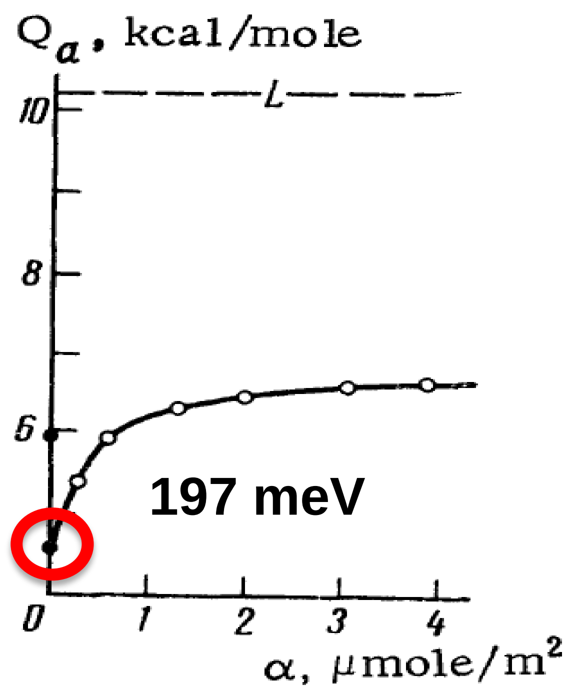


The binding energy is 180±20 meV.

The binding energy has a weak water-orientation dependence.

Insights by Comparing with Experiments

Heat of adsorption of a single water molecule on graphite

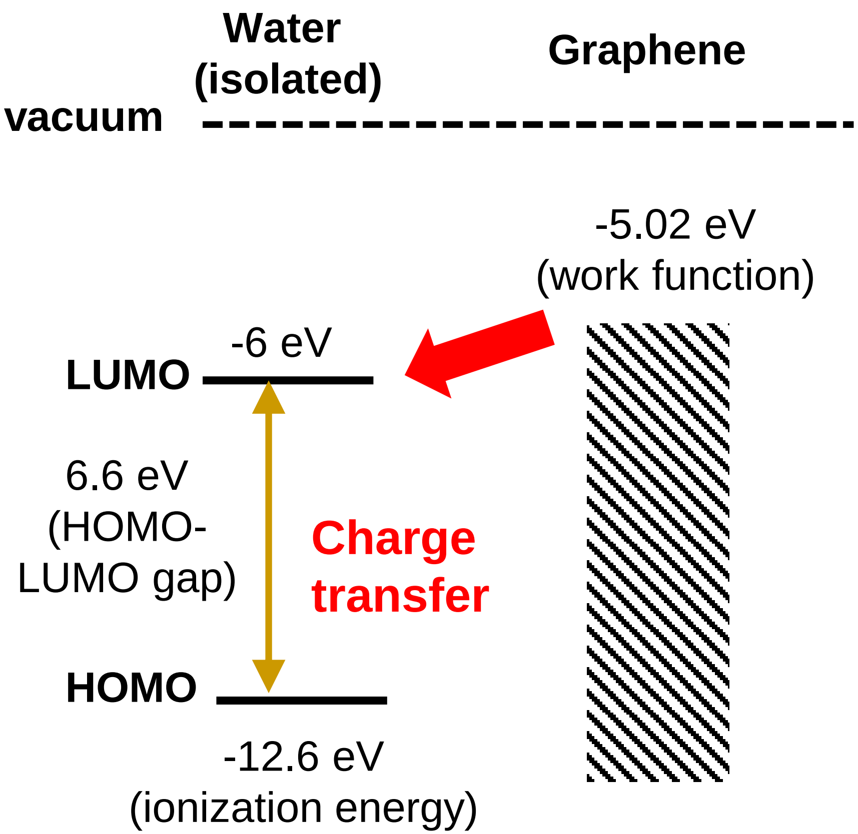


Heat of adsorption energy of water on graphite from gas chromatograms, extrapolated to zero water density (Kiselev et al. 1969)

Our calculation agrees with experiments involving a single water molecule.

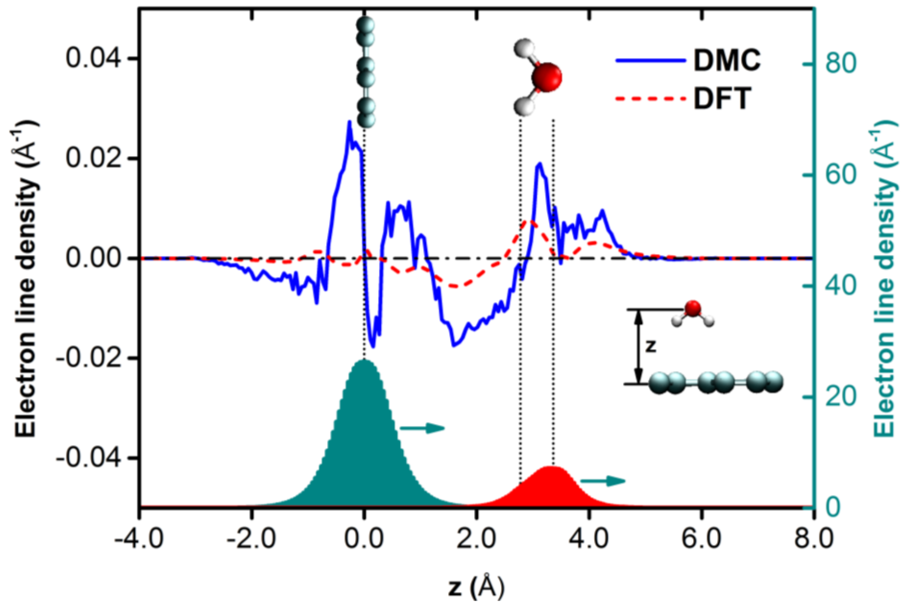
Charge Transfer Between Graphene and Water

Charge transfer by looking at energy levels



Energy levels of a single water molecule and graphene from experiments.

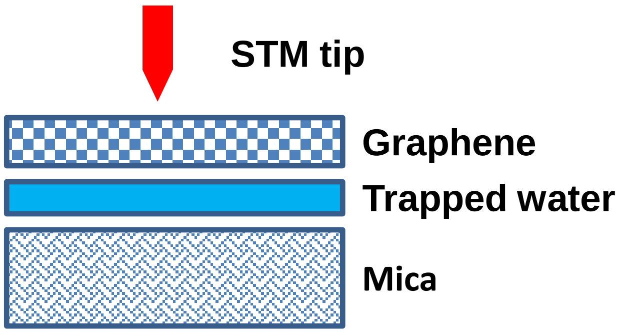
Charge transfer is observed for single water from our electron density analysis!!!



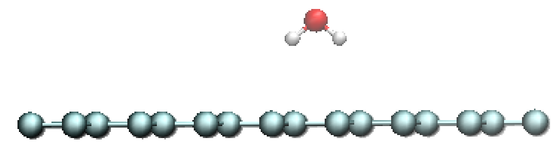
Electron density re-distribution when a single water molecule approach graphene.

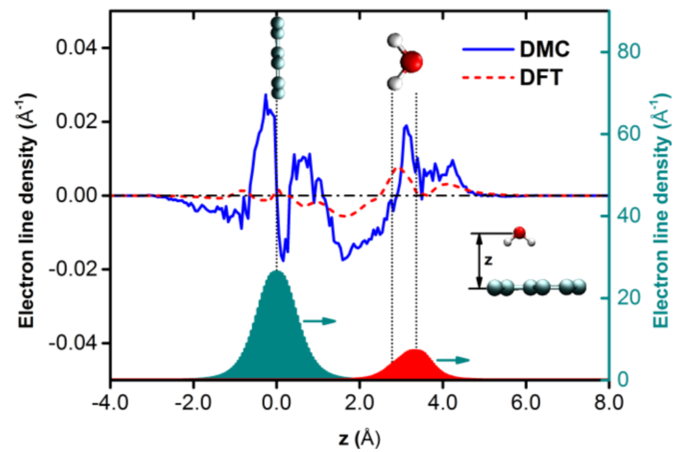
Charge transfer between a single-water molecule and graphene may contribute to the interaction.

Conclusion and Acknowledgment



Control water at nanoscale





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Thank you for your attentions.
Questions and suggestions are most welcome.