Multi-scale Computational Exploration of Two-Dimensional Materials in Nanofluidics and DNA Sequencing

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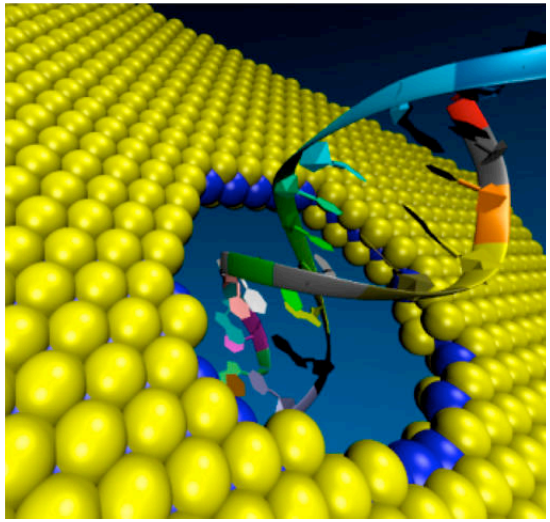
2D Materials in Nanofluidics and DNA Sequencing

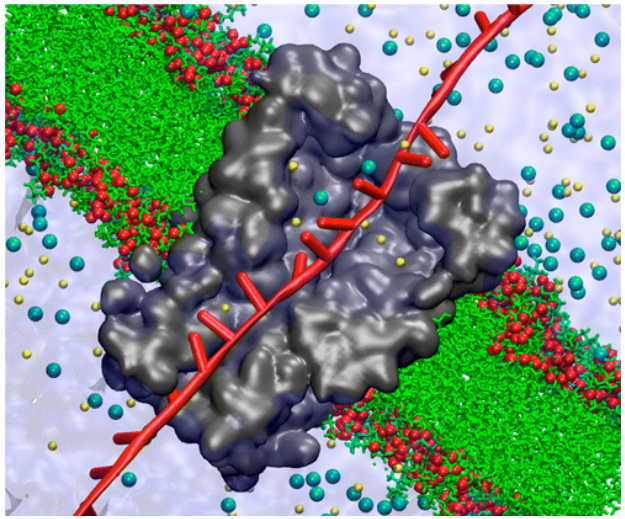
DNA base detection using nano-pore

Molybdenum disulfide (MoS_2)



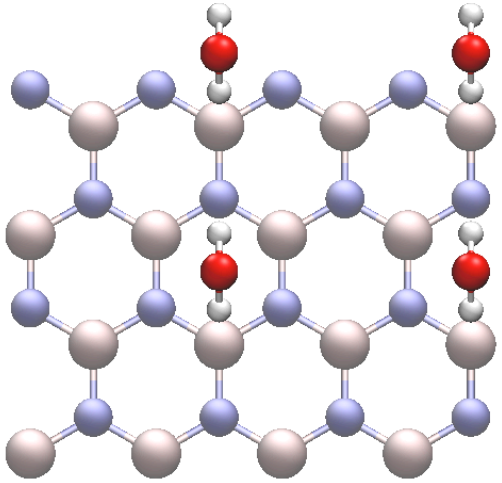
Signal to noise ratio 5 times better than graphene
(ACS Nano 2014)

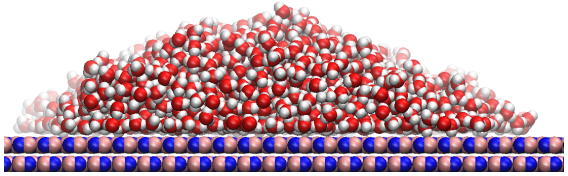
mechanosensitive channel



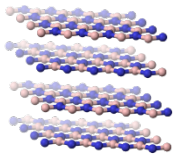
Two distinguishable signals including tension and ionic current
(JPCL 2015)

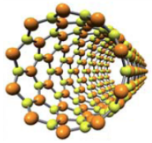
hBN-water interaction using quantum Monte Carlo



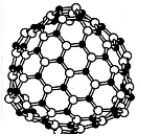


hBN based Materials

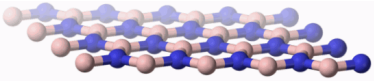




1995

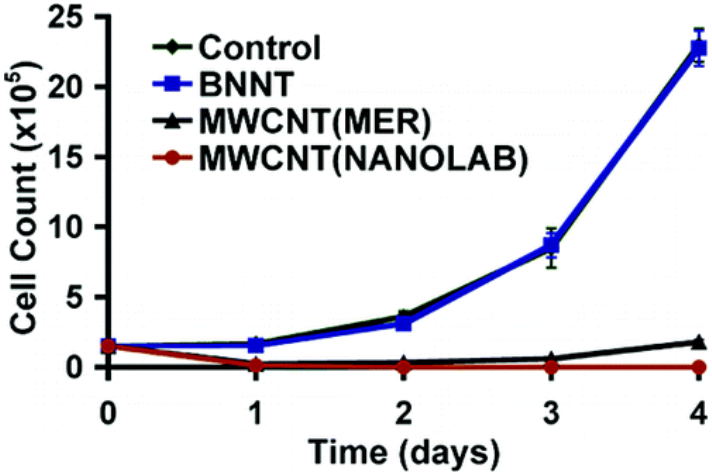


1998



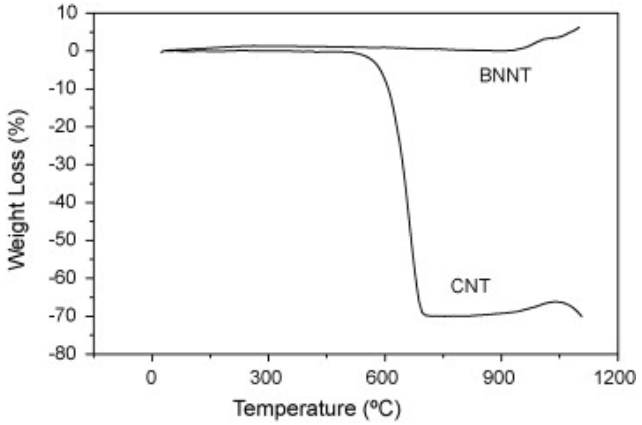
2008

BNNT is noncytotoxic



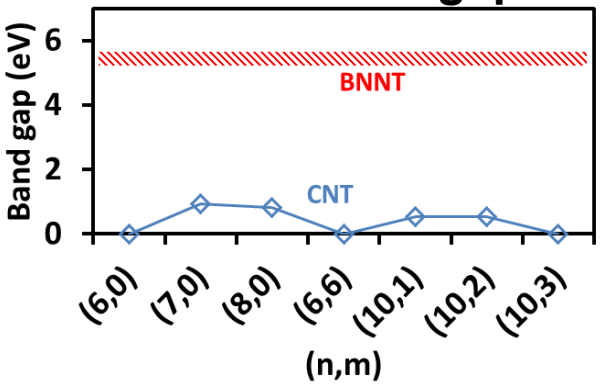
Applications in biological probes and biomaterials (Zettl group, Berkeley and Lawrence Berkeley National Lab, JACS 2009)

BNNT is thermally stable



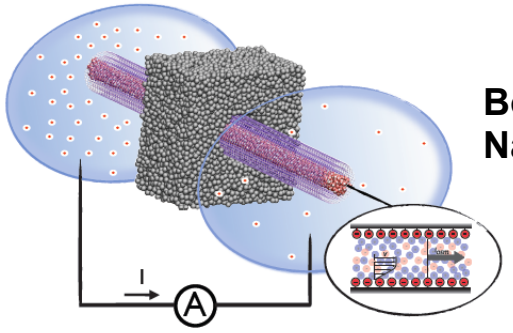
Applications in high-temperature environment (Golberg group, Japan)

hBN has >5 eV band-gap



Blasé group, 1994

BNNT for harvesting "blue energy"

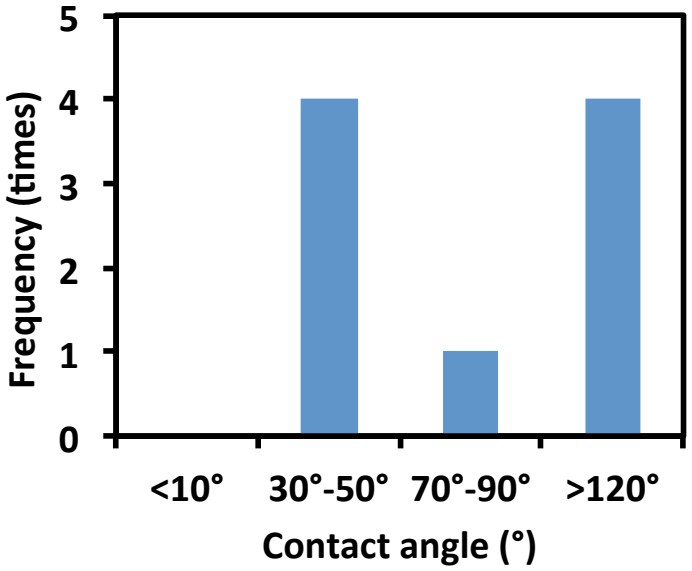


Bocquet group, Nature 2013

Apply the hBN-based materials in nanofluidics and nano medicine.

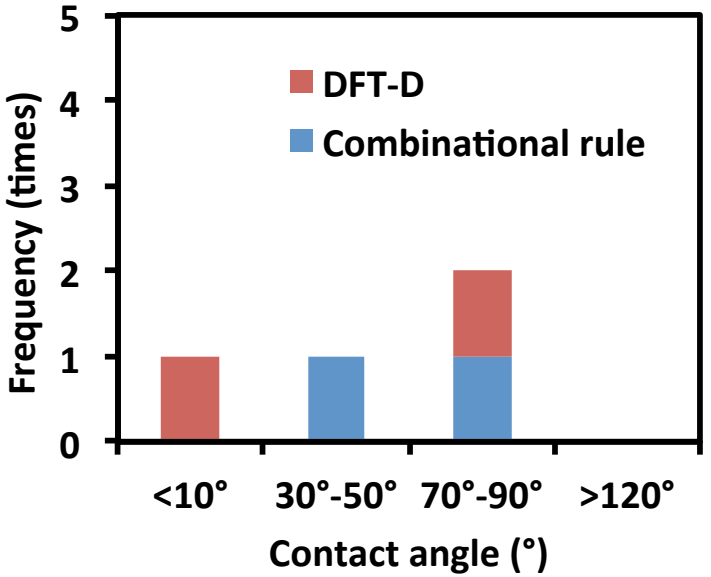
We Don’t Know How Water Interacts with hBN

Experiments

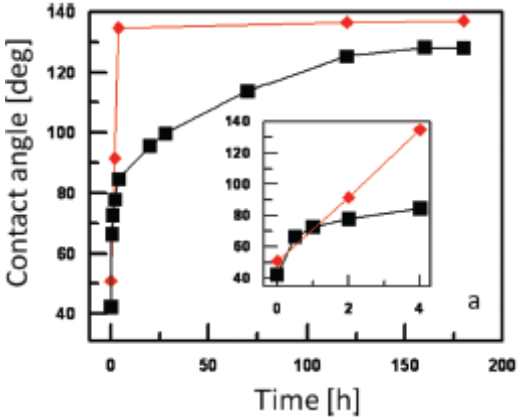


Challenging due to surface contamination, defects, different fabrication methods

Simulations



Lorentz-Berthelot combinational rule or DFT-D calculation



Contact angle changes with time when exposed to air (Boinovich et al. 2013)

Solve Many-body Schrödinger Equation

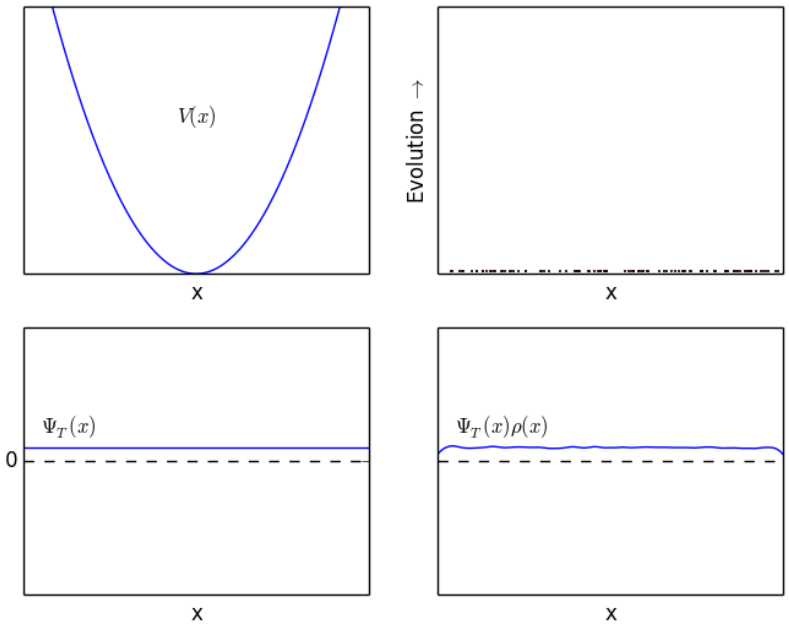
Target $\hat{H}\psi(X) = E\psi(X)$

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

$$X = (X_1, X_2, \ldots, X_n)$$

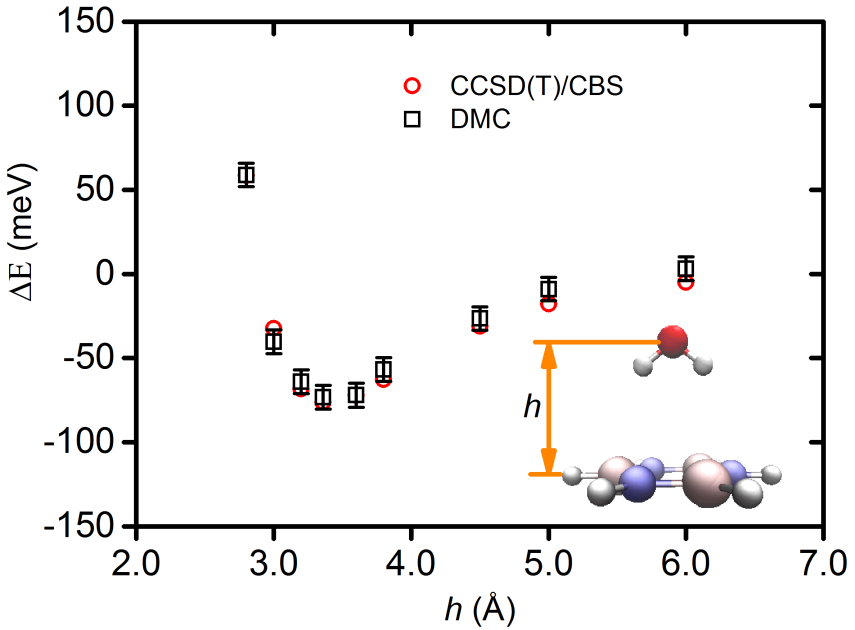
$$\hat{V} = -\frac{1}{4\pi\varepsilon_0}\sum_i^n\sum_I^N \frac{Z_I e^2}{r_{iI}} + \frac{1}{4\pi\varepsilon_0}\sum_{i<j}^n \frac{e^2}{r_{ij}}$$

Diffuse Monte Carlo (DMC)
Isomorphism between Schrödinger equation and stochastic process
Wavefunction ↔ Distribution of walkers
Kinetic ↔ Diffusion
Potential ↔ birth/death



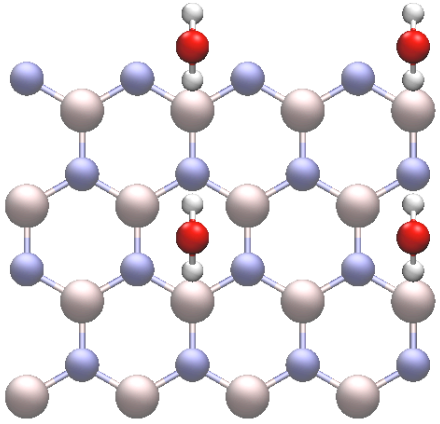
Example: harmonic oscillator

Verify DMC method

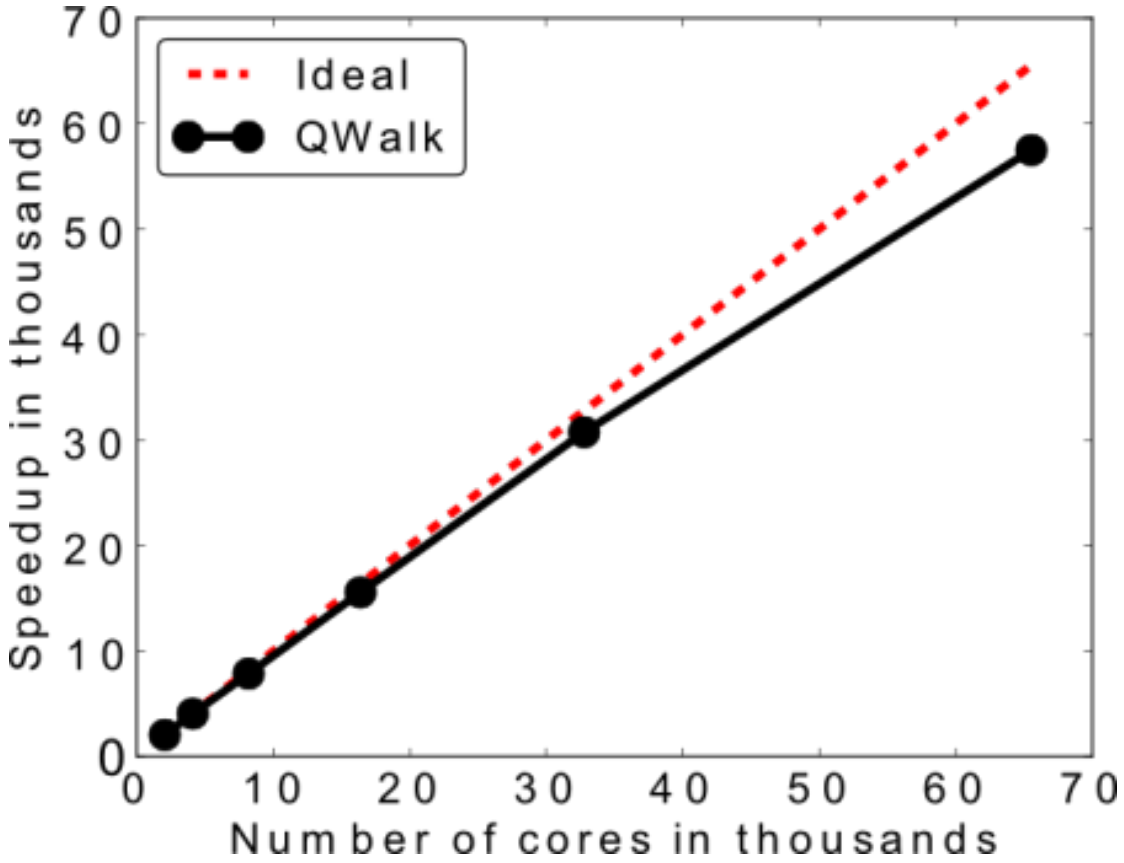


The DMC method is verified and calibrated by comparing to couple cluster method with complete basis set (CBS) for a small system. Qwalk package is used.

Utilizing the Power of Blue Waters



One hBN-water energy point cost 980,000 core hours.

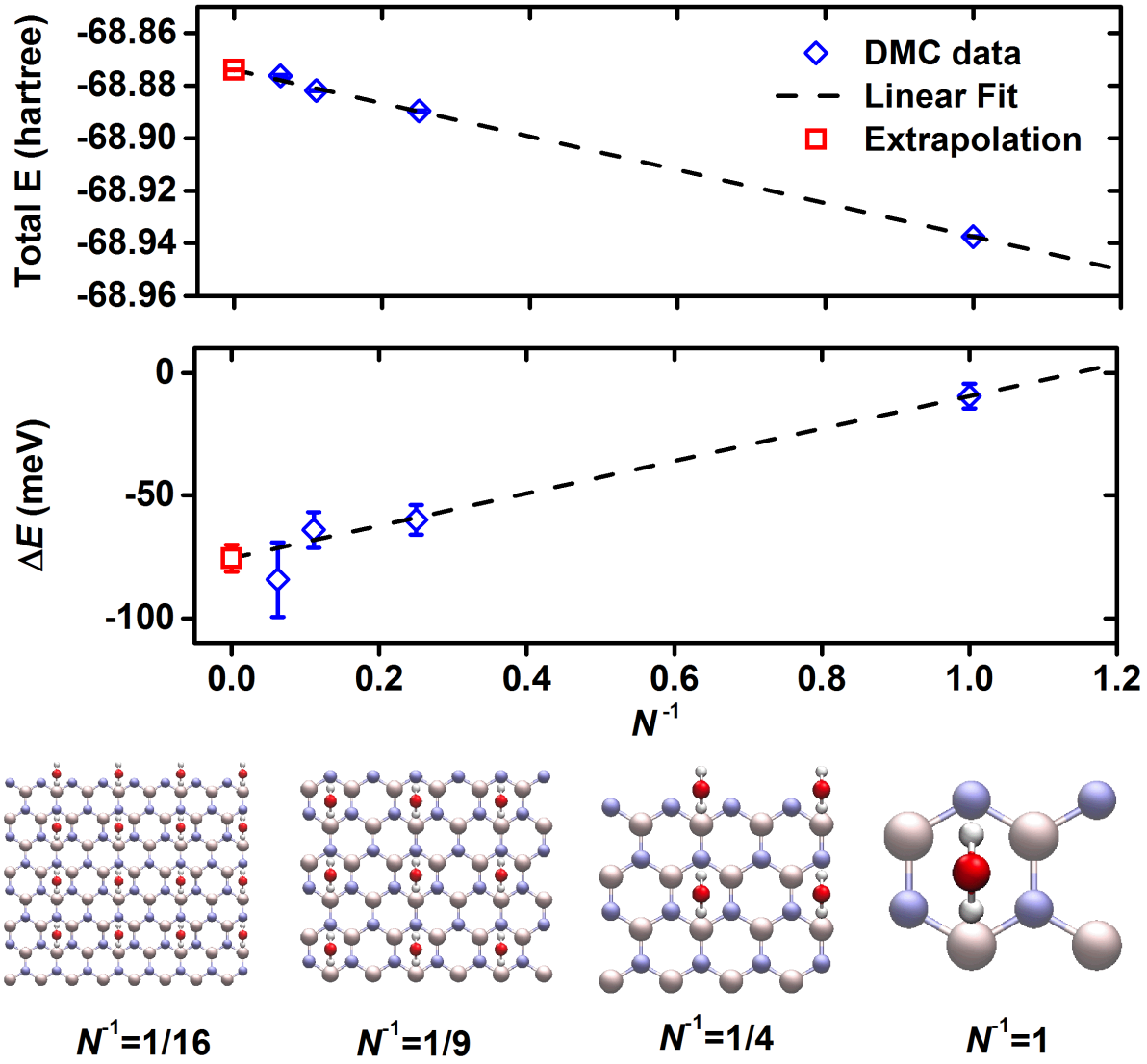


Scaling of Qwalk with number of processors

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

Error Control in DMC

Finite Size Error



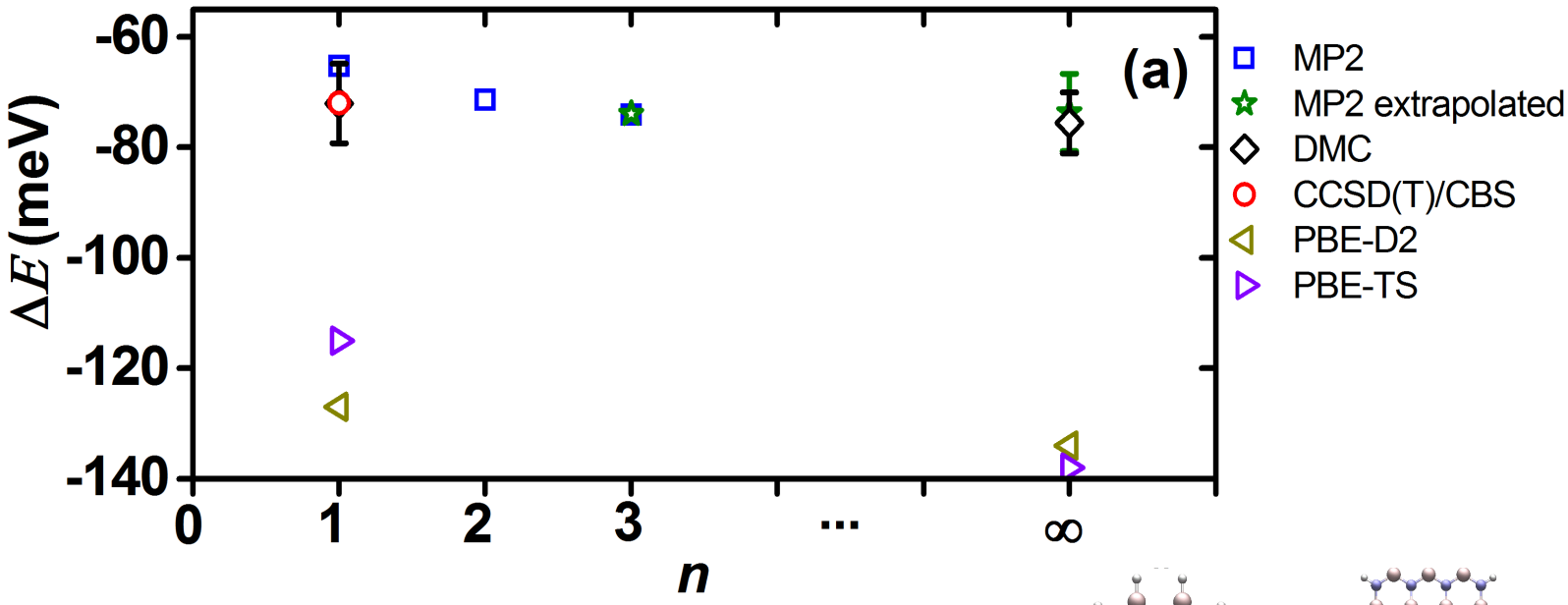
Binding energies between a single water molecule and hBN monolayer: 76 ± 6 meV

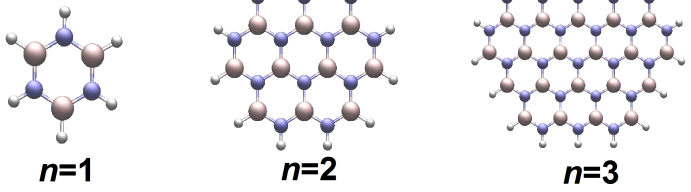
Finding an Efficient Method Using DMC as Reference

Developing force field parameters needs Δ*E* for multiple configurations.

DMC is computationally expensive: ~980,000 core hours for one data point.

Use DMC to verify MP2 for one configuration.
Then use MP2 to compute interaction energy of other configurations.

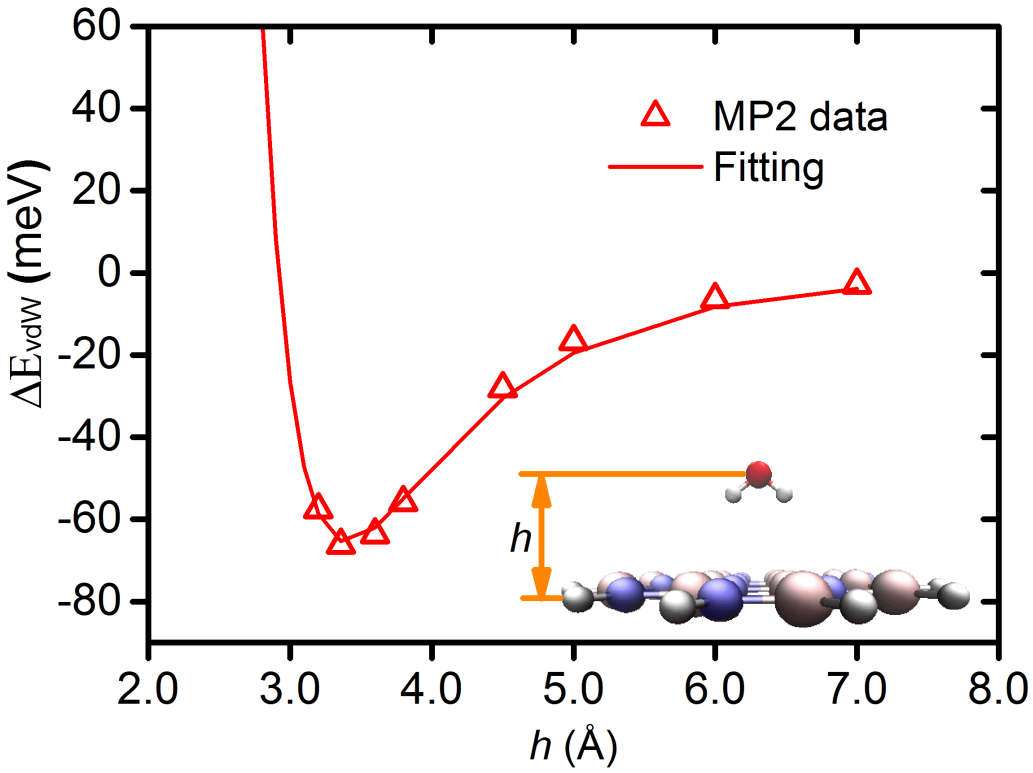




For hBN, Δ*E* = -74 ± 7 meV
Compared to -76 ± 6 meV by DMC

Develop Parameters by Fitting to MP2 Energies

$$\Delta E = \sum_{i\in\{\mathrm{N}\}} 4\varepsilon_{\mathrm{O\text{-}N}}\left[\frac{\sigma_{\mathrm{O\text{-}N}}^{12}}{r_{\mathrm{O}\text{-}i}^{12}} - \frac{\sigma_{\mathrm{O\text{-}N}}^{6}}{r_{\mathrm{O}\text{-}i}^{6}}\right] + \sum_{j\in\{\mathrm{B}\}} 4\varepsilon_{\mathrm{O\text{-}B}}\left[\frac{\sigma_{\mathrm{O\text{-}B}}^{12}}{r_{\mathrm{O}\text{-}j}^{12}} - \frac{\sigma_{\mathrm{O\text{-}B}}^{6}}{r_{\mathrm{O}\text{-}j}^{6}}\right] + E_{\mathrm{vdWO\text{-}H}} + E_{\mathrm{ESP}}$$

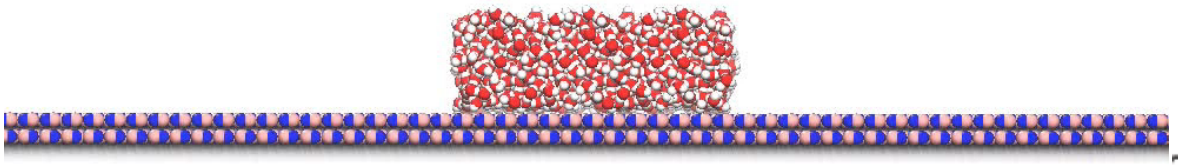


$\sigma_{\mathrm{B\text{-}OW}}$ (Å)	$\varepsilon_{\mathrm{B\text{-}OW}}$ (kcal/mol)	$\sigma_{\mathrm{N\text{-}OW}}$ (Å)	$\varepsilon_{\mathrm{N\text{-}OW}}$ (kcal/mol)	$q_{\mathrm{B}}/q_{\mathrm{N}}$
3.444	0.1016	3.398	0.1255	±0.30

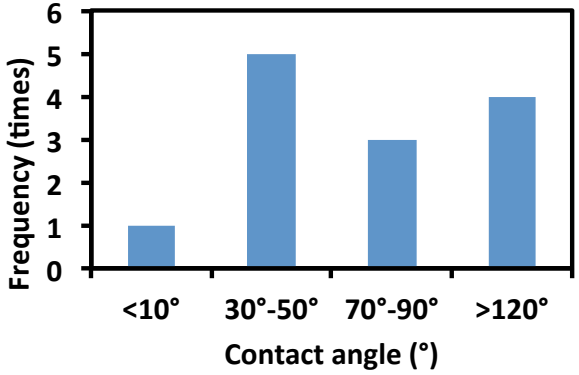
Simulate Contact Angle using Parameters

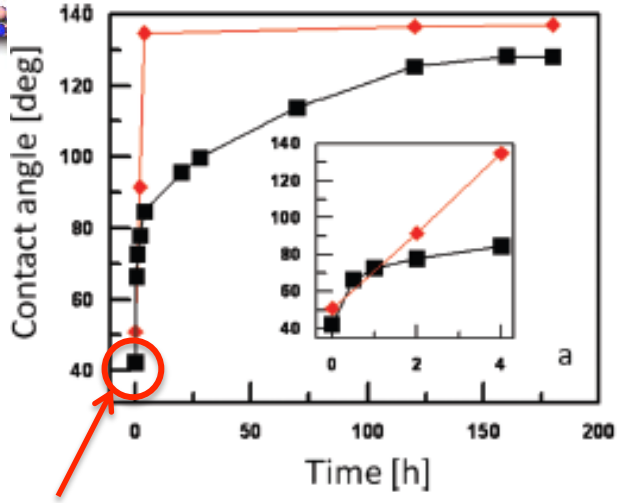
Use molecular dynamics simulations with the developed force field parameters

$\sigma_{B\text{-}OW}$ (Å)	$\varepsilon_{B\text{-}OW}$ (kcal/mol)	$\sigma_{N\text{-}OW}$ (Å)	$\varepsilon_{N\text{-}OW}$ (kcal/mol)	q_B/q_N
3.444	0.1016	3.398	0.1255	±0.30



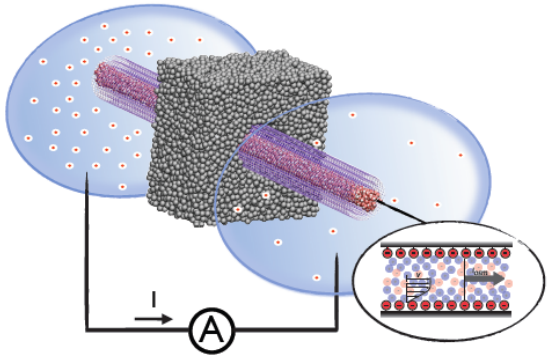
Contact angle: 45 ± 4°



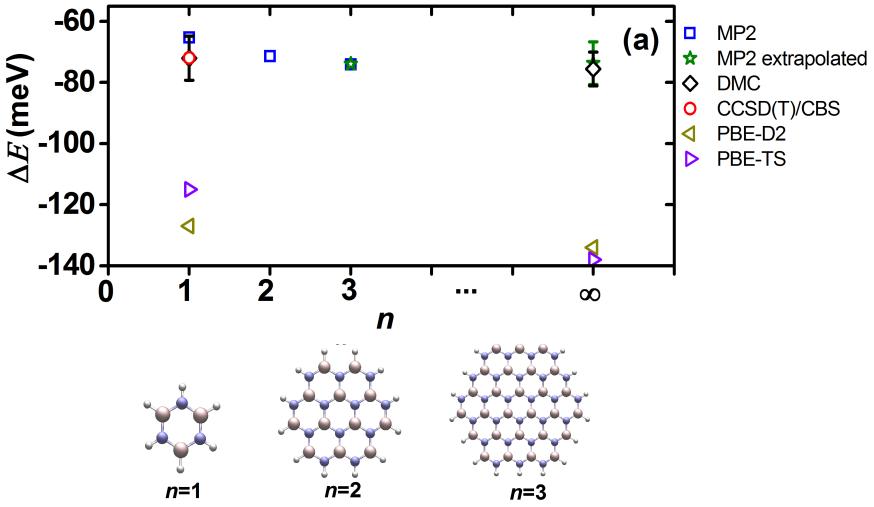


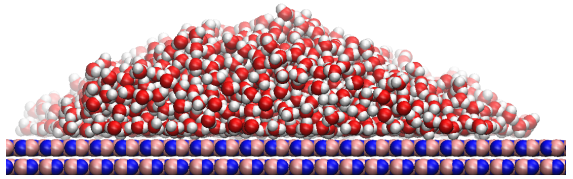
Experiment: Surface with minimum contamination

Conclusion and Acknowledgment



Apply hBN in nanofluidics





Special Thanks to NSF, Air Forces, and Blue Waters (ILL).

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