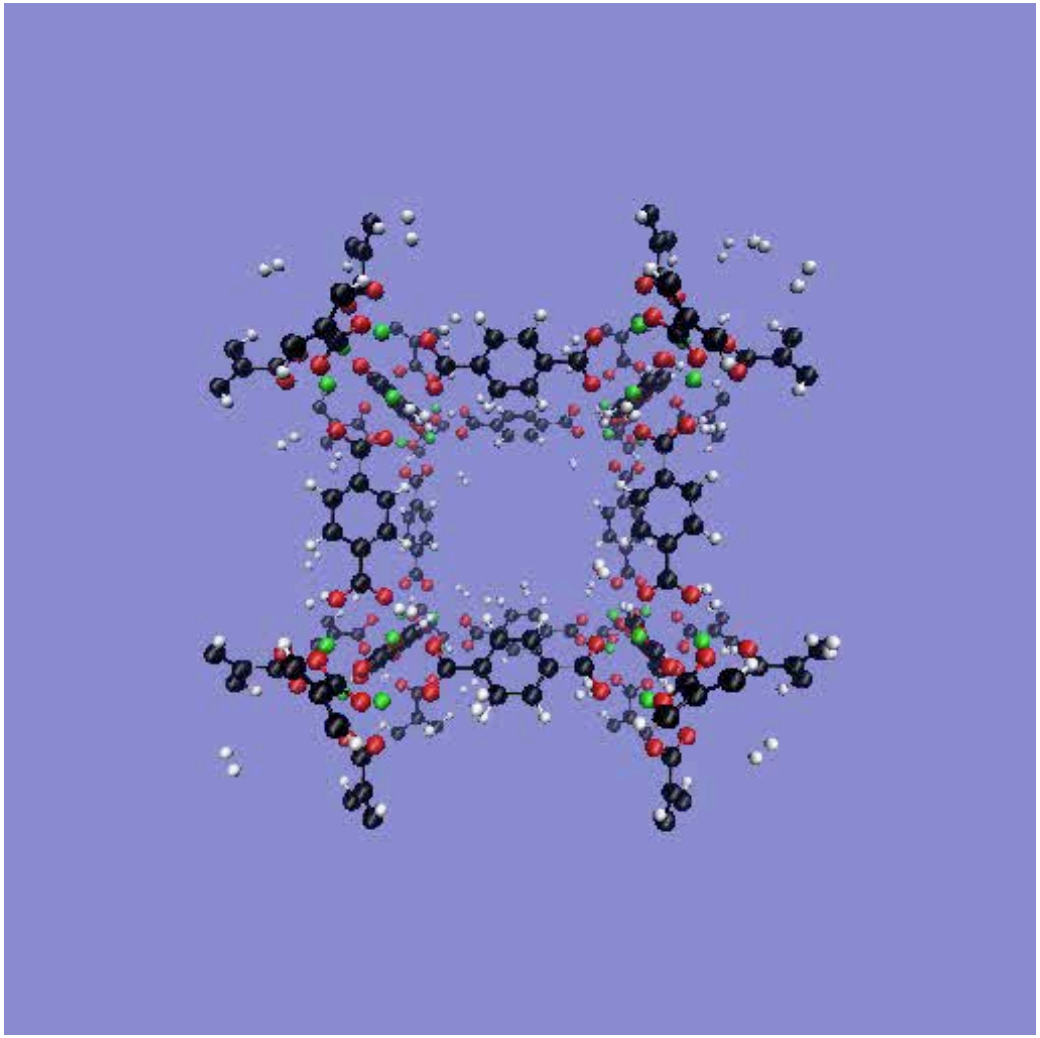
Understanding hydrogen storage in metal organic frameworks from first principles



Understanding hydrogen storage in metal organic frameworks from first principles

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http://charm.cs.illinois.edu/OpenAtom

Overview

- We plan to study metal organic frameworks (MOFS)
  - What is a MOF?
  - Why study hydrogen in MOFs?

- Software we run on Blue Waters: OpenAtom
  - What does it do?
  - How is it parallelized?
  - How does it perform?
  - Why do we need Blue Waters?

- What we learned so far

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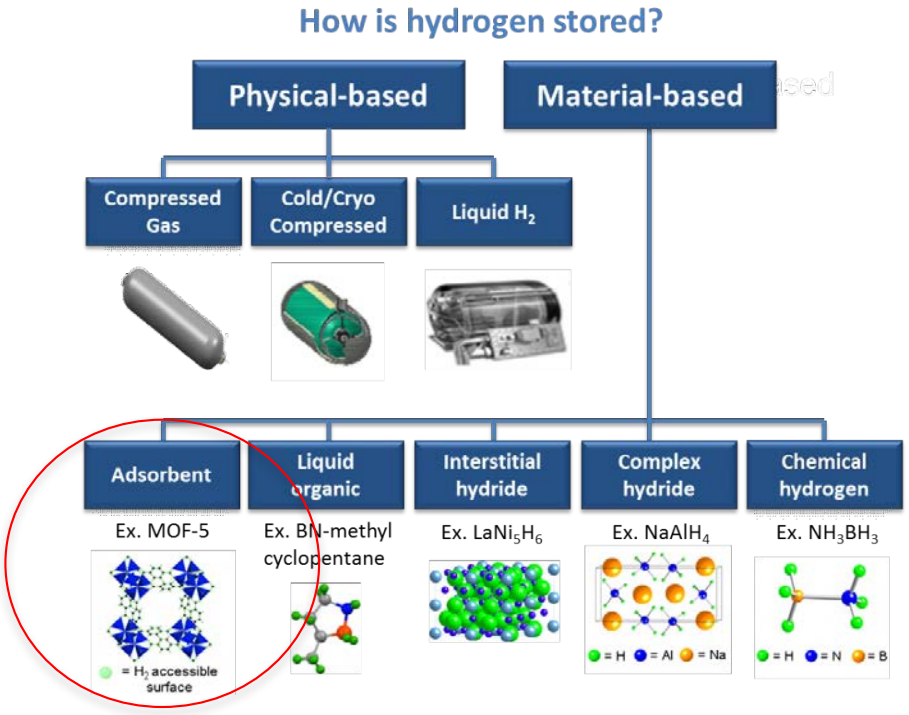
Hydrogen storage for green energy

- Hydrogen as fuel
  - energy dense
  - with clean burn
  - hard to store

- Need lightweight material that can store and release a lot of H_2

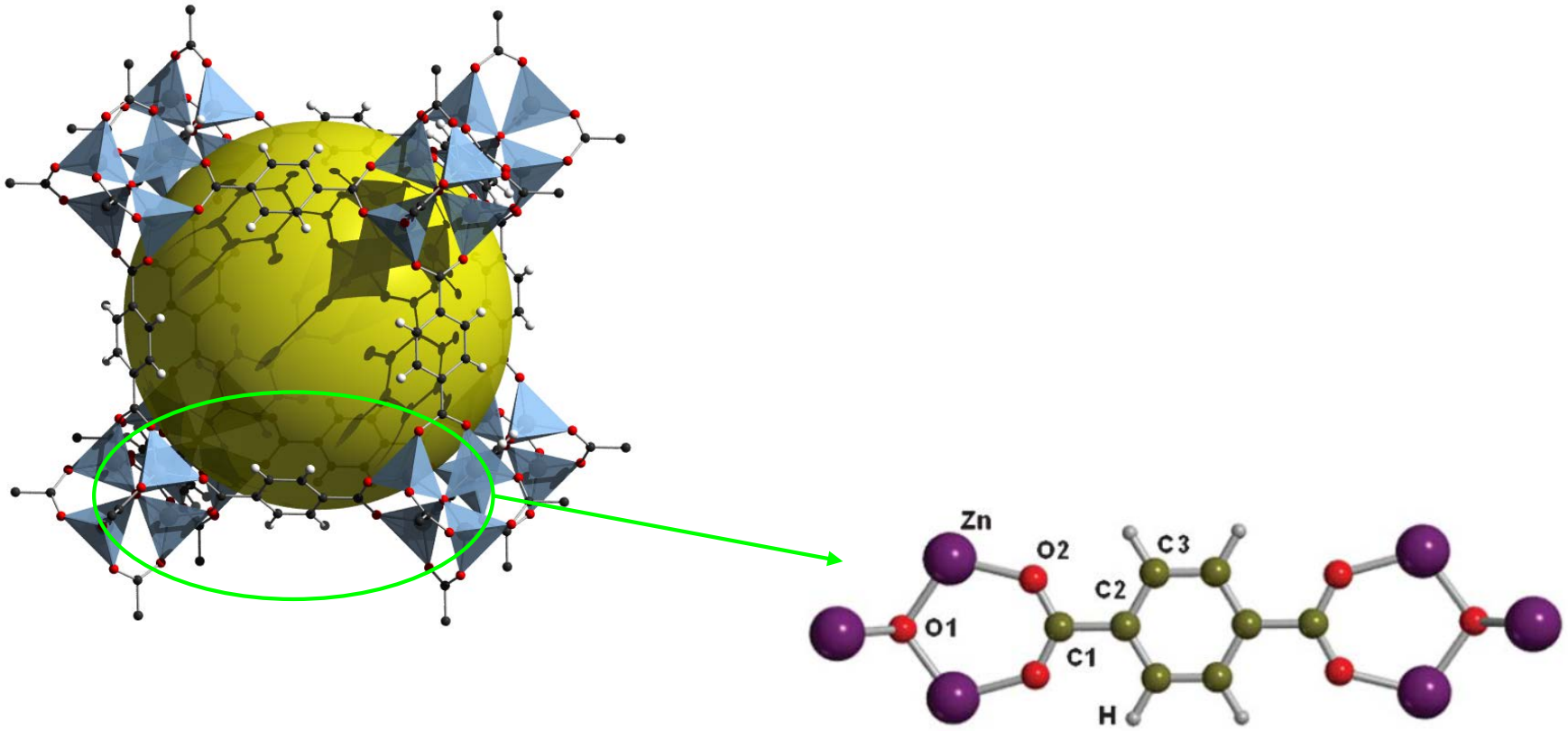
- Metal organic frameworks (MOFs)
  - Porous
  - Large interior surface area
  - Can store reasonable amount of H_2
  - Complex material, details of process not known
  - Optimization of H_2 storage not great to date



http://energy.gov/eere/fuelcells/hydrogen-storage

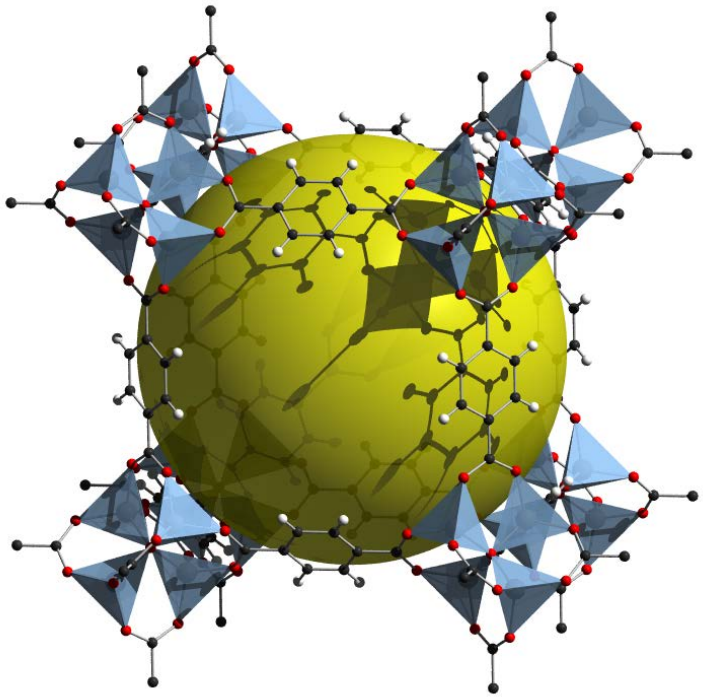
DOE target for a H_2 storage system not yet been reached: e.g., capacity of 40 g H_2 per L.

Typical MOF structure



MOFs we study

- MOF-5 : $Zn_4O(1,4\ benzenedicarboxylate)_3$
- 424 atoms in a simulation cell
- Can change Zn to other metals



Questions to answer:

- How do H_2 bind / diffuse inside MOF?
- Temperature & loading dependence

Molecular dynamics (MD) needed

- Simulate motion of MOF + H_2 to see what happens in real time
- Dynamics & thermodynamics

Technical challenge: H_2 is very light

- Standard MD: point-like atoms move due to interatomic forces
- Hydrogen is *quantum mechanical*: not point-like but *wavy*…

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What is OpenAtom

Sohrab Ismail-Beigi
Applied Physics
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Computer Science
UIUC

Glenn Martyna
Physical Chemistry
& Materials
IBM

NSF SI2-SSI: Scalable, Extensible, and
Open Framework for Ground and
Excited State Properties of Complex Systems

- OpenAtom software package : DFT (now) , GW (next)
- Plane waves and pseudopoentials
- charm++ parallel infrastructure

OpenAtom: what does it do?

- Massively parallel *ab initio* molecular dynamics (AIMD)

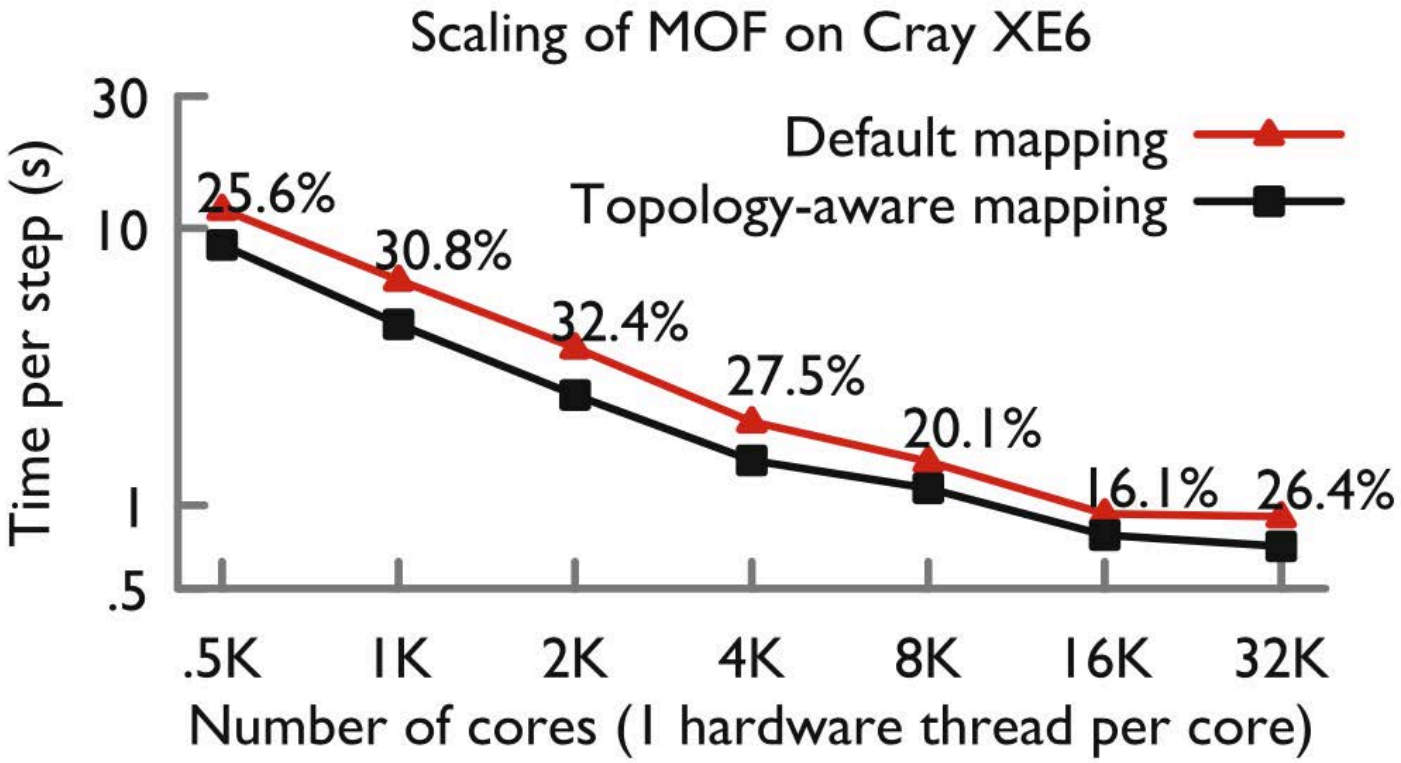
- Describes electrons quantum mechanically, i.e., bonding, *explicitly* using basic physics (no fudge parameters or fits)

- Uses general Fourier basis to represent electron waves
- Uses Charm-FFT *library*: 2D decomposed parallel FFT with spherical cutoff awareness

For the experts:

- Plane waves and pseudopoentials
- Car-Parrinello and Born-Oppenheimer MD

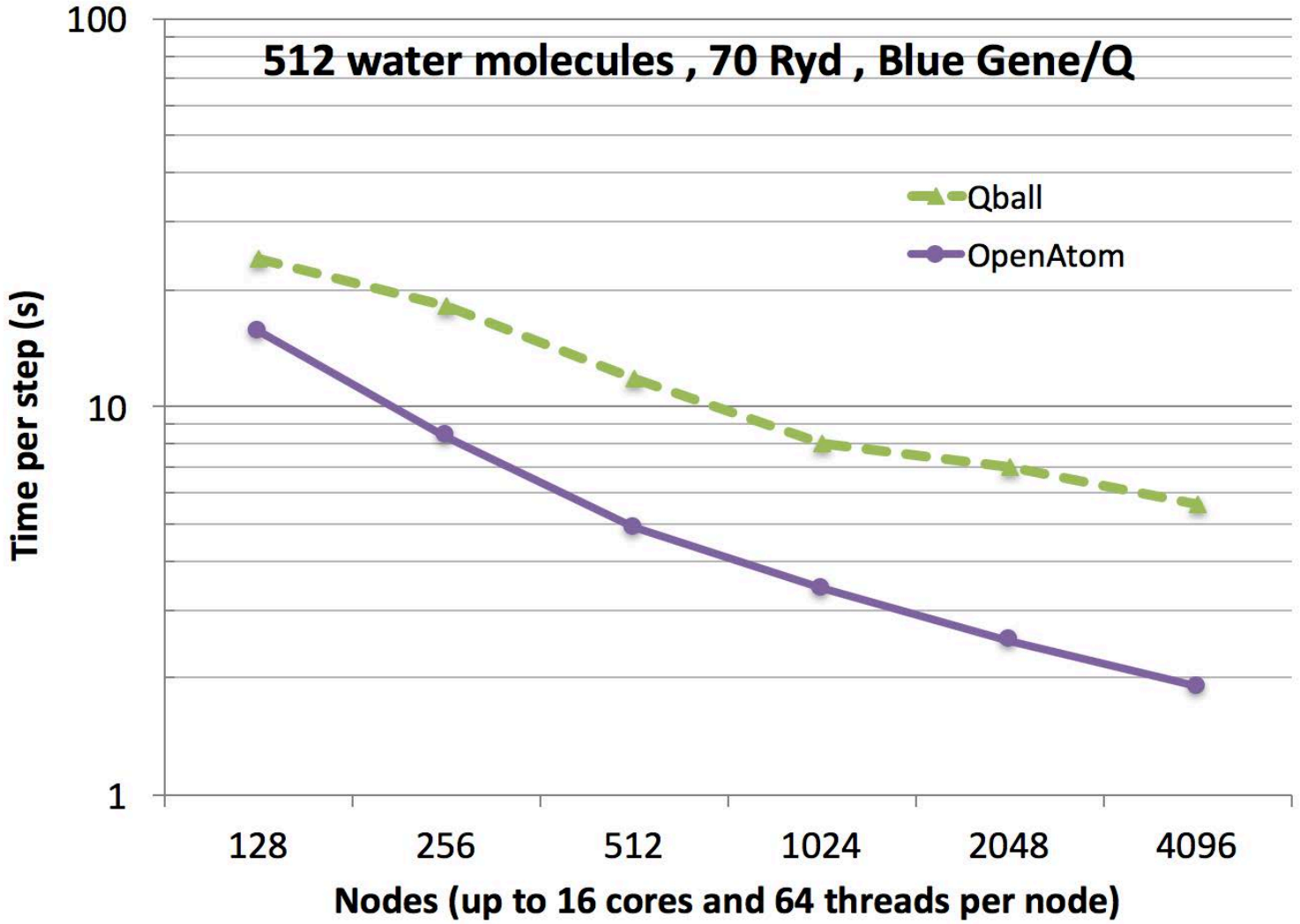
Strong scaling performance



- Few MD steps of AIMD with MOF
- Fixed problem size (strong scaling)
- Time to solution with increased of cores

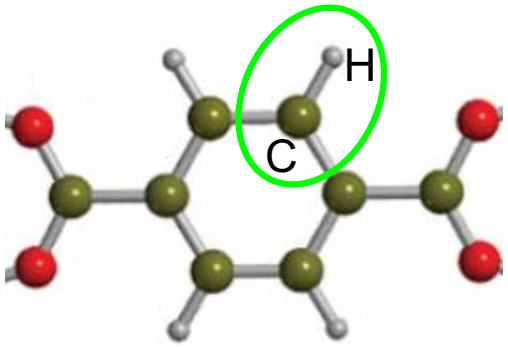
Is OpenAtom competitive?

- Few steps of AIMD
- Compared to Qball : a LANL fork of Qbox AIMD software



Why quantum hydrogen?

- Consider C-H bond in part of the MOF
- Vibration frequency f ~ 9 x 10^{13} Hz



- Classical (point-like atom) picture
  - C-H length vibrates at this frequency
  - Continuous vibrational energy

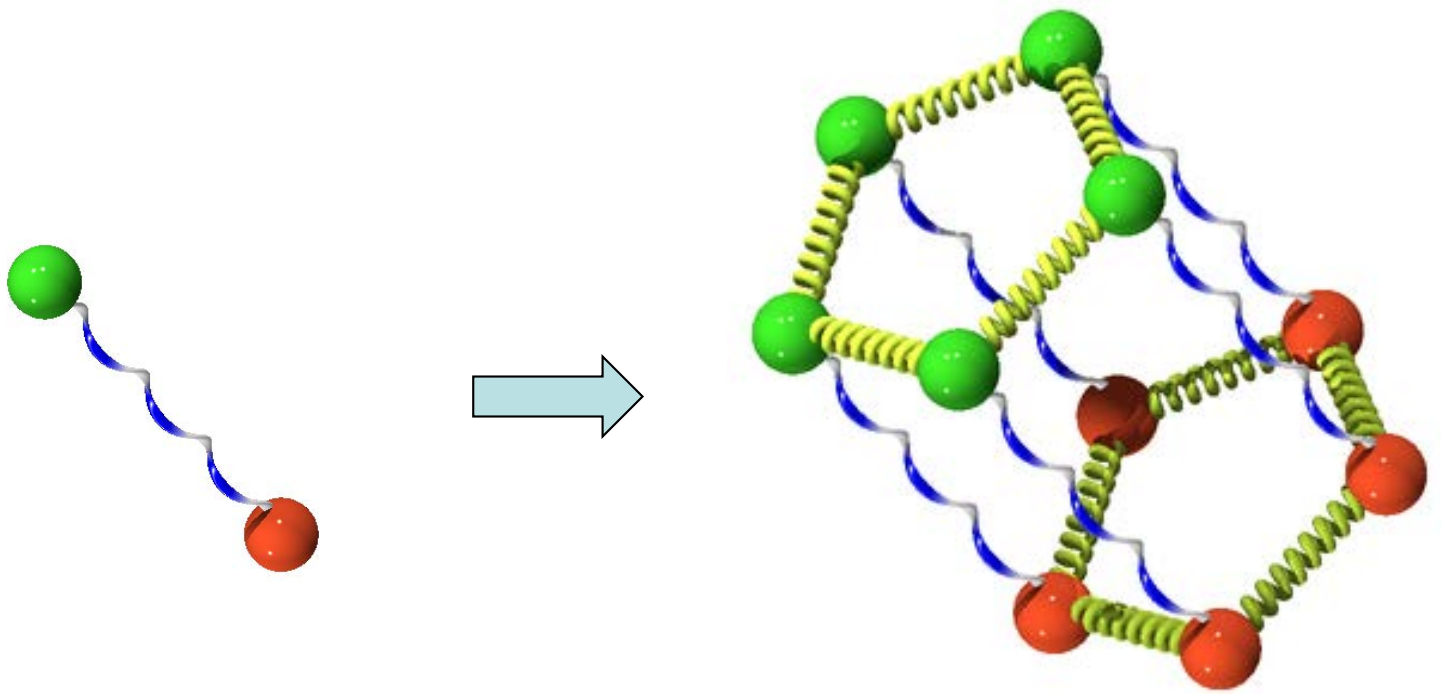
- Quantum systems: energy discrete
  - Einstein says $E = hf$
  - Convert to temperature : 4300 K !

- For any reasonable temperature, the C-H bond
  - does *not* vibrate at all (stuck in ground state)
  - no dynamics per se

Path integral Monte Carlo

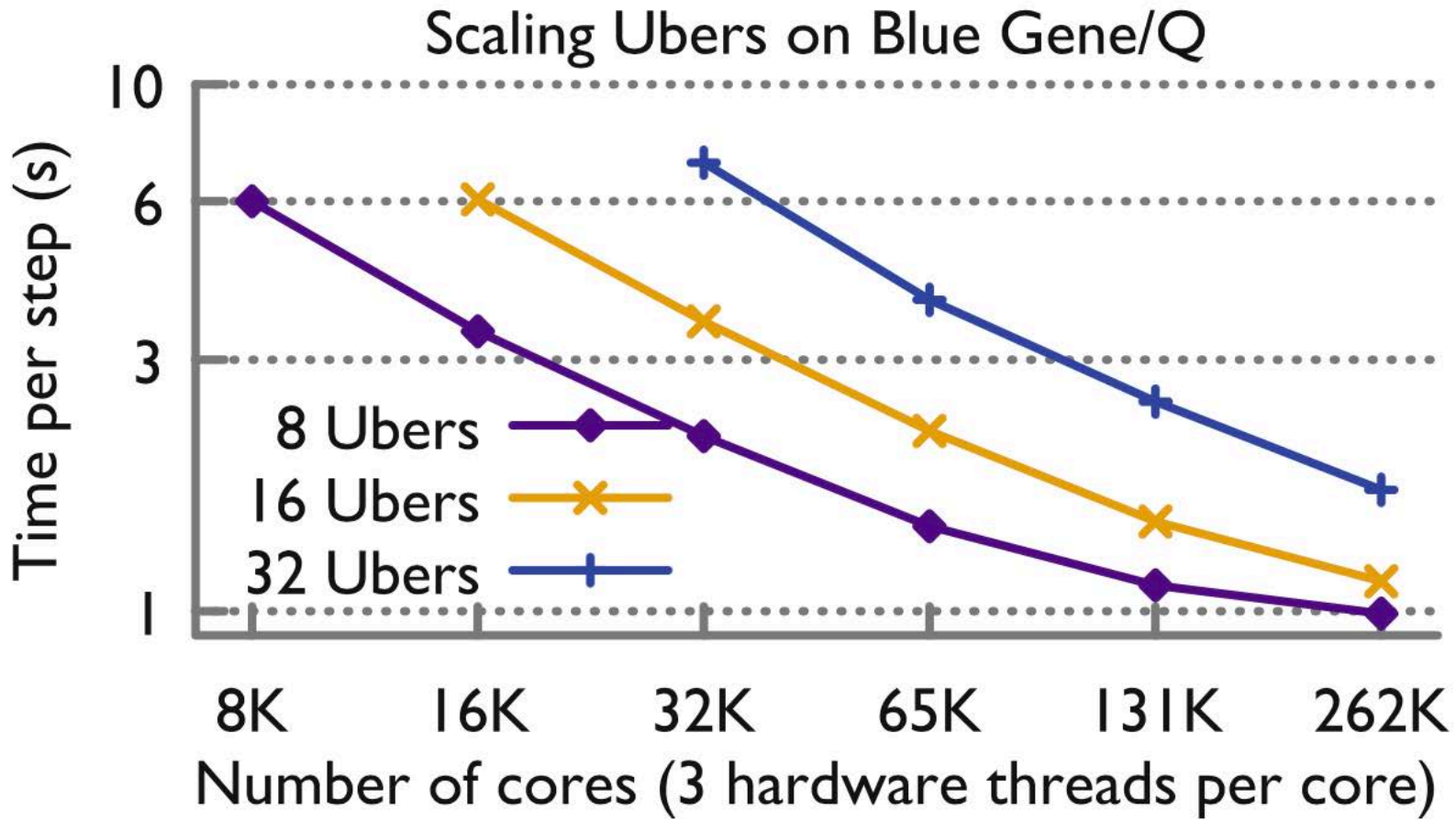
- A way to make atomic coordiantes (nuclei) quantum

- Multiple images of MOF (“beads”)
- Beads connected by appropriate springs
- Entirety evolves as a giant MD



Path integral strong scaling

- Multiple beads of MOF (ubers)
- Entirety evolves as giant MD



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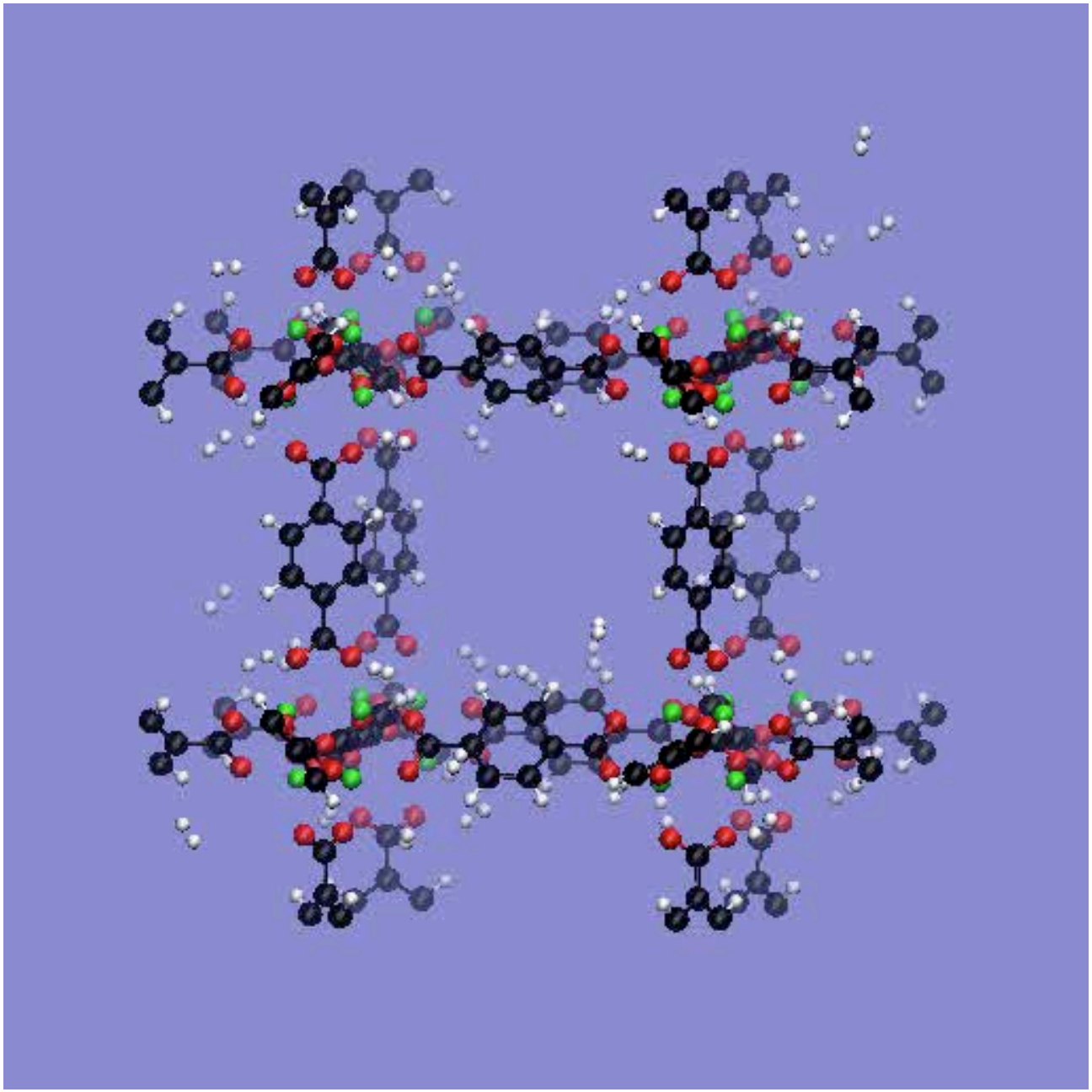
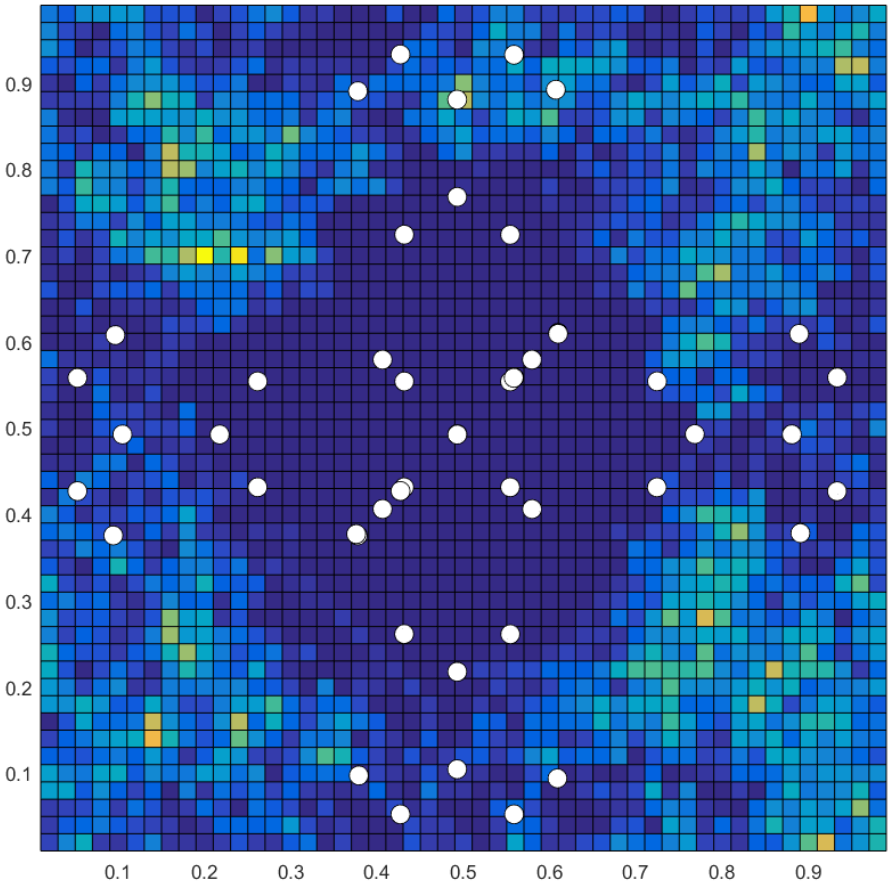
Preliminary results: MD itself

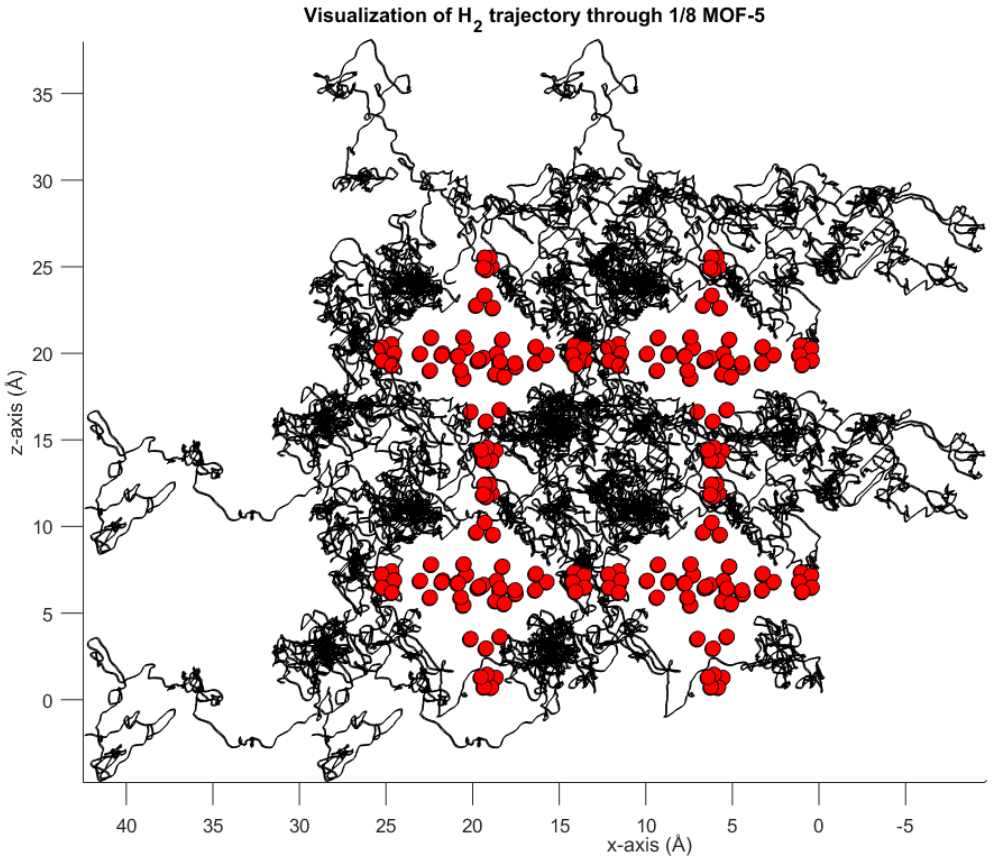


Preliminary results: diffusion

Heatmap: mean H_2 positions in simulation cell



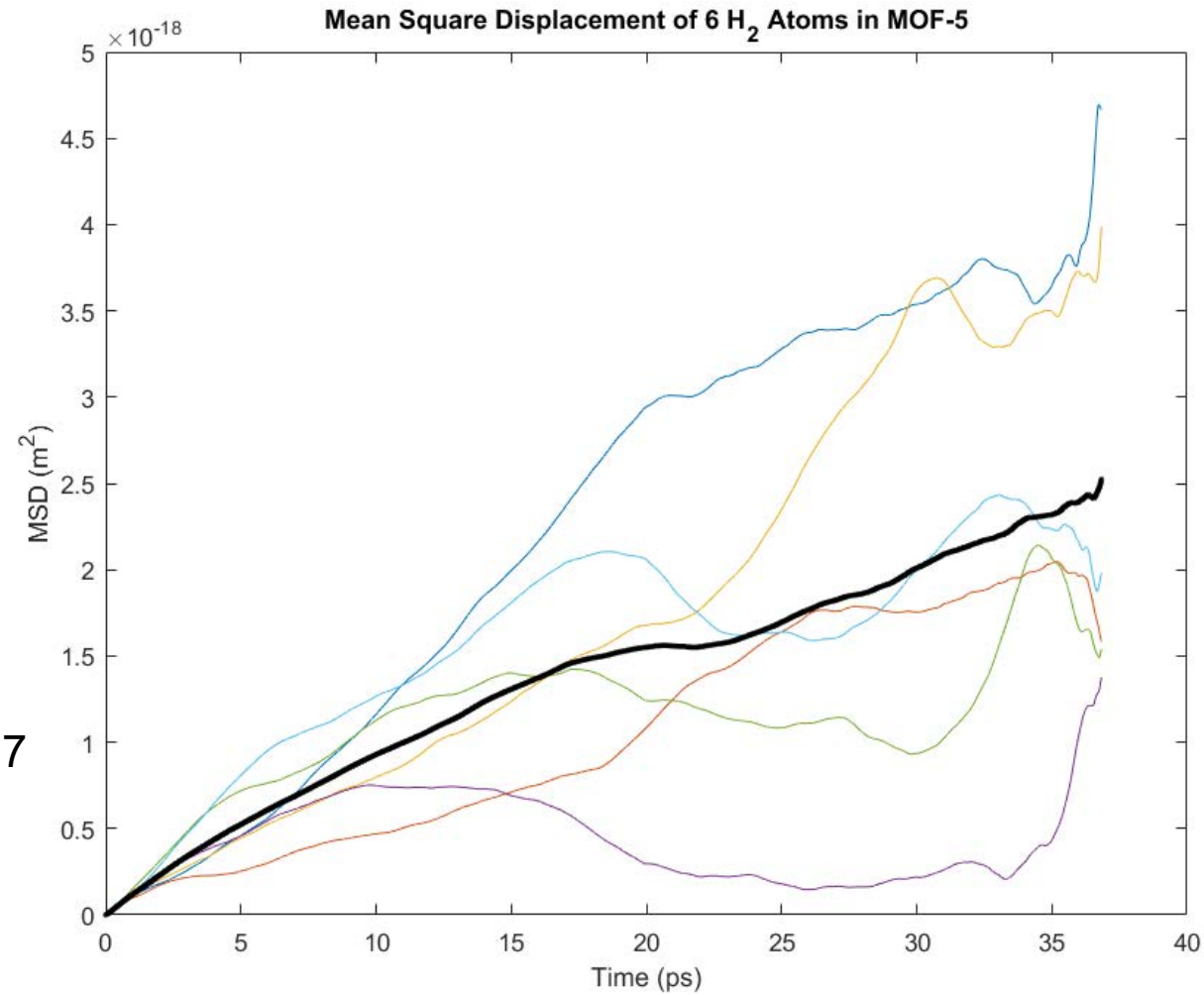
Diffusive paths followed by H_2 molecules over time



Preliminary results: diffusion

- Slope: m = 7 x 10^{-8} m^2/s
  - $\boxed{D \approx 10^{-8} \frac{m^2}{s}}$
- Seems to agree with available literature

$$D \approx 7 \times 10^{-9} \frac{m^2}{s}$$



J. Phys. Chem. C **2008,** *112,* 2911-2917

Summary

- Study metal organic frameworks (MOFS) for H_2 storage

- Use OpenAtom on Blue Waters: OpenAtom
  - Highly parallel
  - Scales well
  - Can simulate quantum nuclear effects

- Preliminary non-quantum simulations
  - Seem reasonable
  - Need longer simulations/more statistics

- Gear up for fully quantum simulations this year