Computational challenges in nanoscience: an ab initio perspective

Presented by

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Outline

- Introduction
  - Motivation, Scientific challenges, Methods
- Case studies
  - Application scaling and performance
  - Research areas
- Opportunities
  - Research areas
  - Computational requirements

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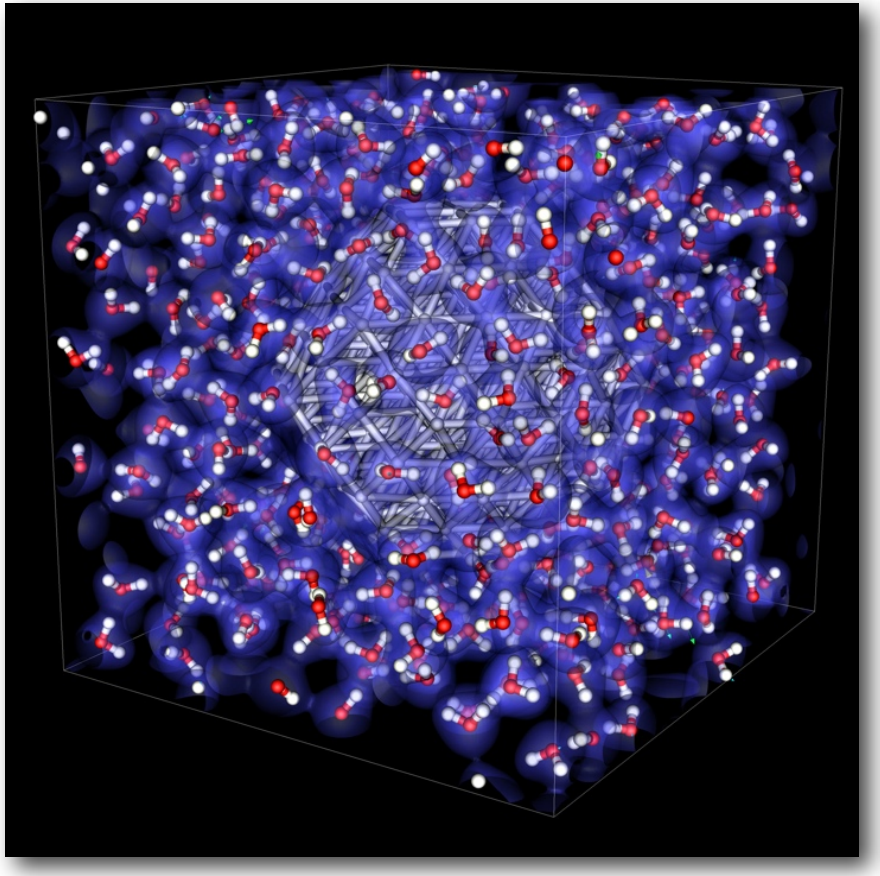
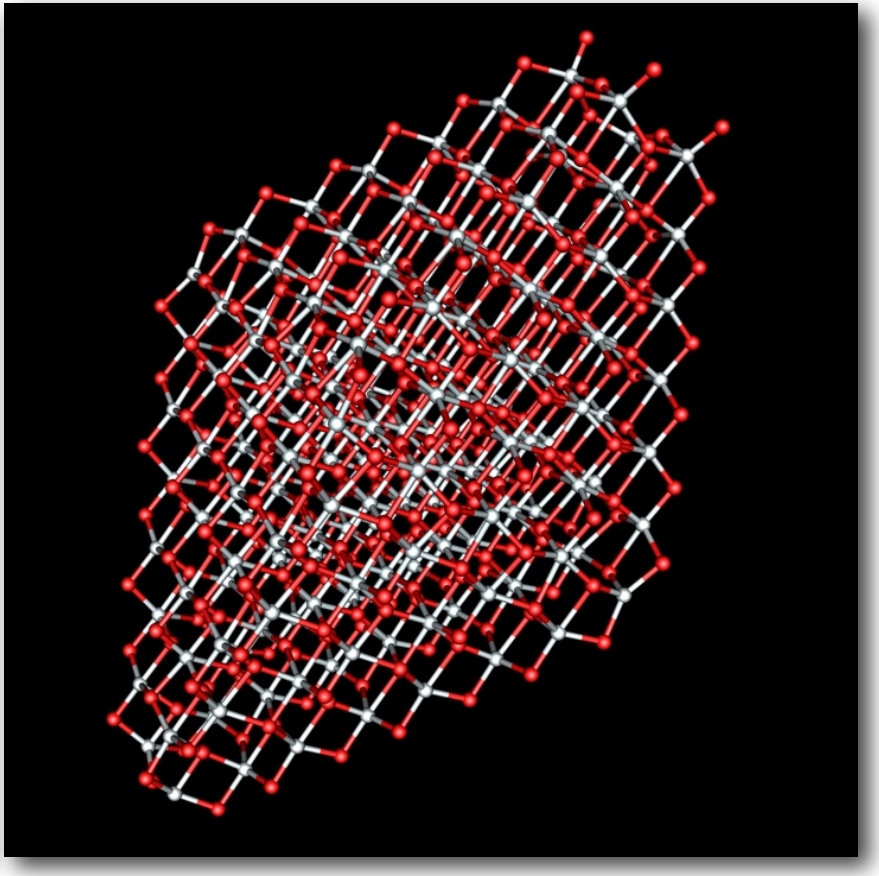
Focus on CNMS User Projects



Molecular dynamics
$Pt_{201}+427H_2O=1482$ atoms



Configuration search
$Ti_{161}O_{322}$ (~2nm)=483 atoms

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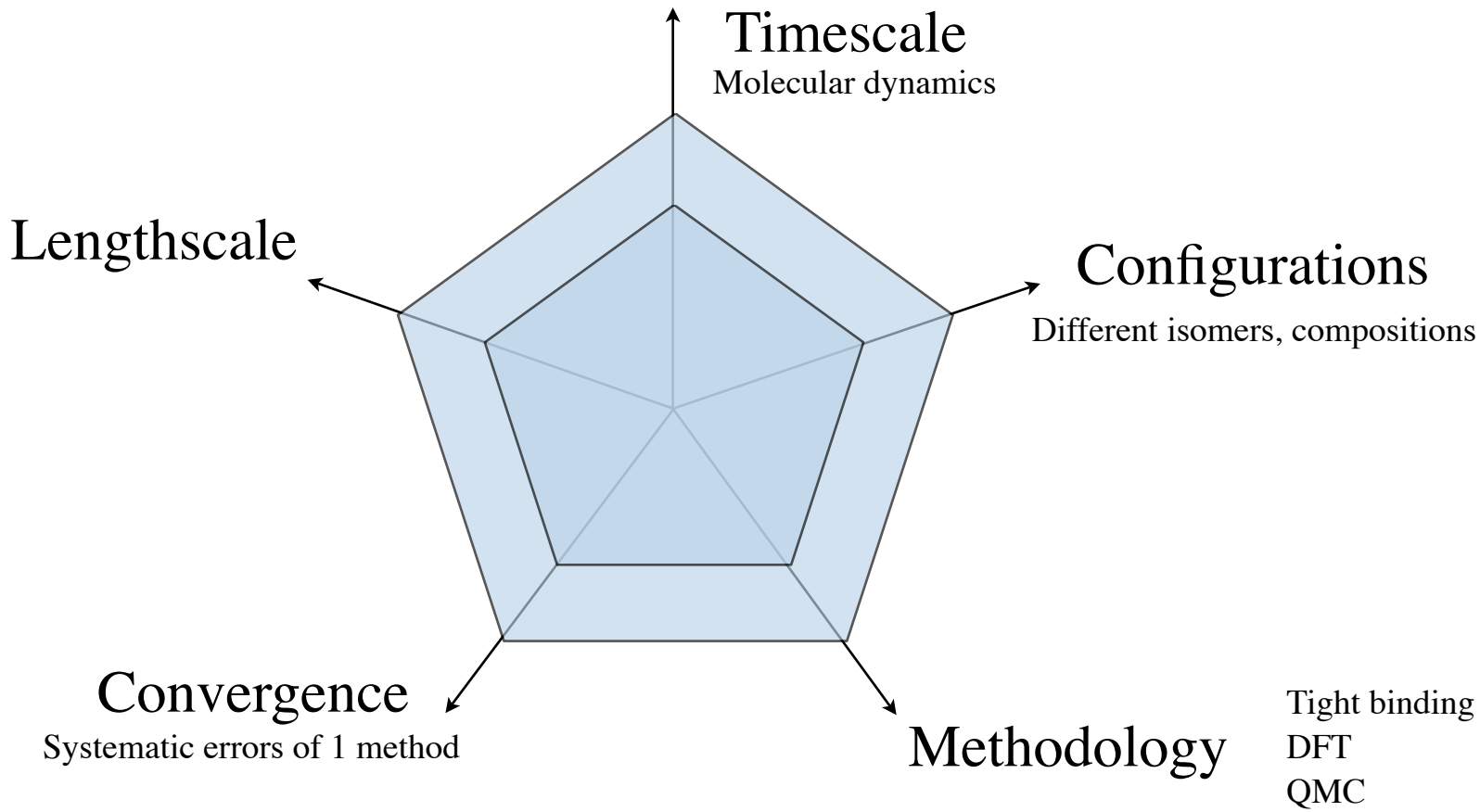
Computational challenge: moving beyond increased length scales



- **Human limits: e.g. time to solution. 10ps at 1 minute/ ionic step, 0.5fs steps=> 2 weeks wall clock time**

Electronic structure calculations

- Use a well validated approach: traditional plane-wave $O(N^3)$ density functional methods
- Nanostructure calculations require computation of the structures, energetics and dynamics of highly inhomogeneous systems; Often few-of a kind runs

- PAW and ultrasoft plane wave pseudopotential formalism
- Real space evaluation of PAW projectors
- Single k-point in results presented here
- Tuned VASP 4.6; also applies to PWSCF, CPMD...

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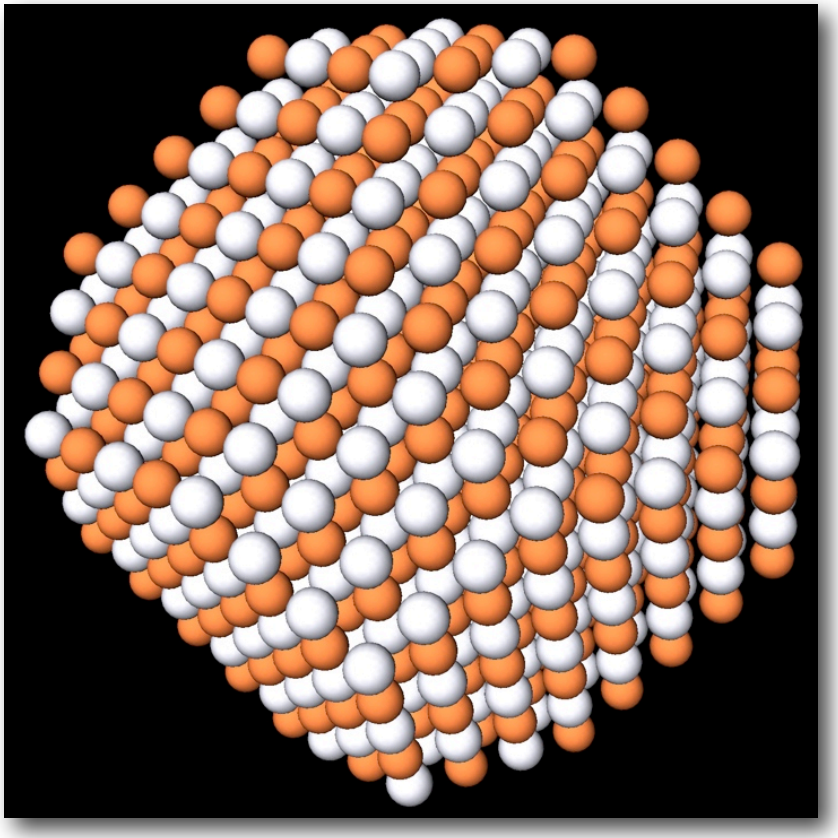
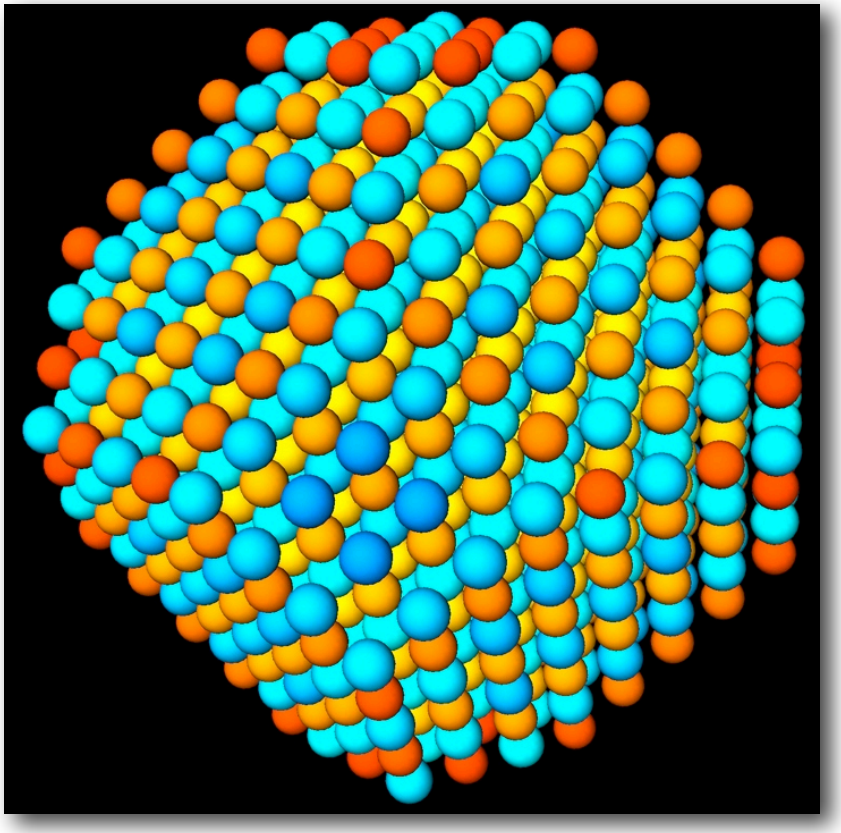
FePt nanomagnetism benchmark

- High density magnetic storage candidate,
- Industry target: ordered arrays of ~3nm particles





$Fe_{399}Pt_{408}$=807 atoms

Structure and ferromagnetic magnetic moments

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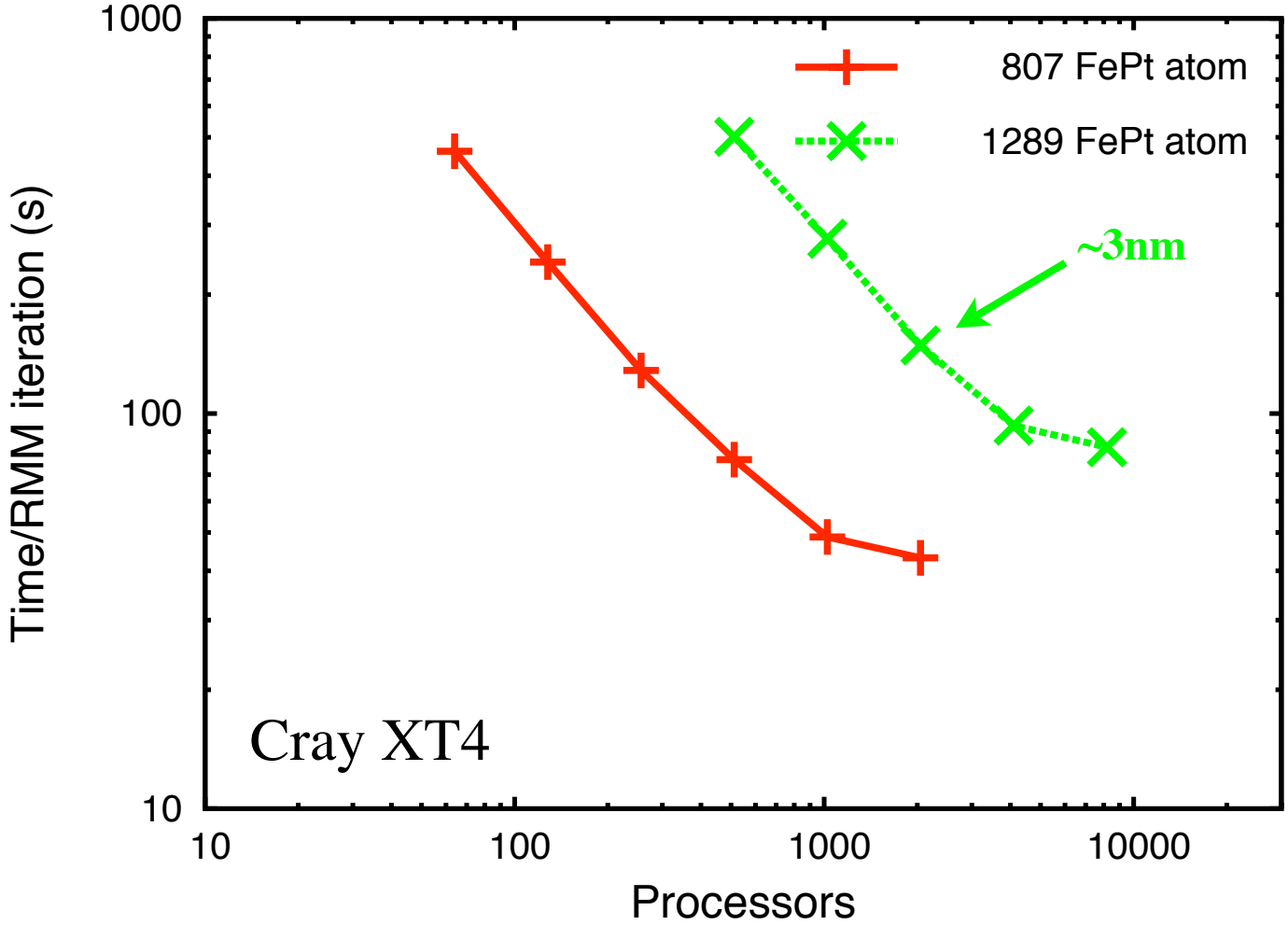
FePt nanoparticle benchmark results



- **Timings for ferromagnetic state (i.e. not spin parallel)**
- **Converged, relaxed geometries have been obtained**

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Scaling summary

- **Nanoscience “nm” length scales are achievable on a few thousand processors**
- **Rule of thumb (dual core Opterons, Cray XT4)**
  - **1 processor/atom good scalability**
  - **Few processors/atom scalability limit**
  - **Many exceptions, but a good starting point[*]**
- **Detailed analysis of the performance and reasons for turnover are available**

[*] Scalability is determined by the electron count (subspace size), plane wave count (cutoff, simulation cell size), k-points, suitability of different diagonalization algorithms (~electronic gap), hardware considerations (interconnect latency+bandwidth...)

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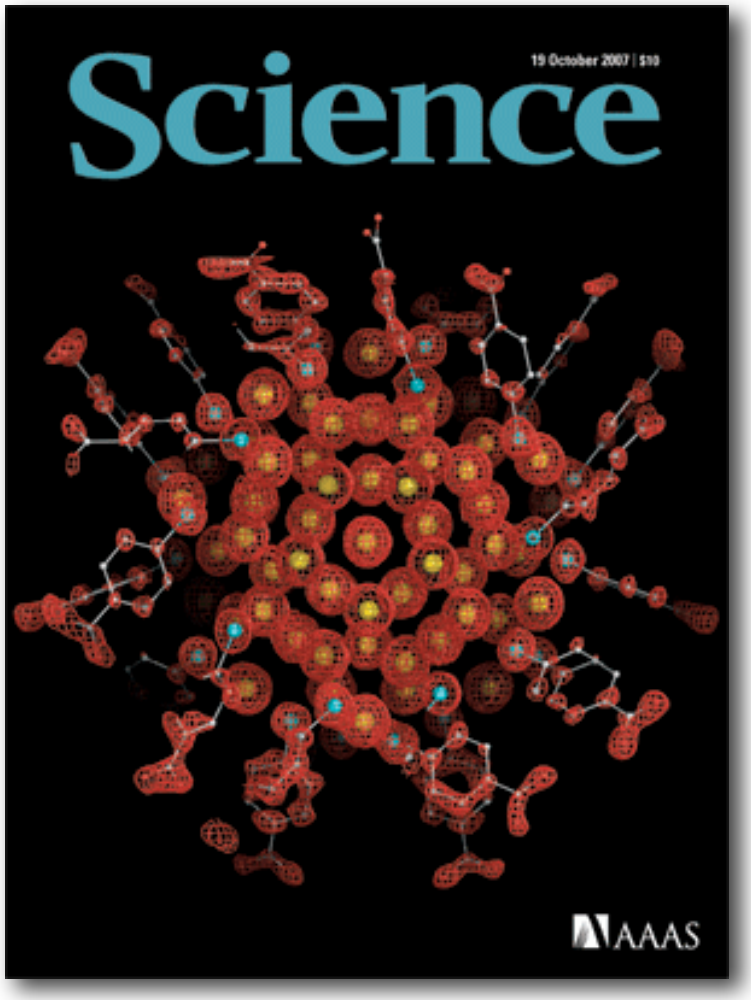
Overlap of experimentally and computational accessible length scales



Jadinski et al. Science **318** 430 (2007)

- X-ray determination of gold nanoparticle structure at 1.1A resolution
- 102 gold atoms + thiol layer
- Likely "only" an O(1000) processor DFT calculation

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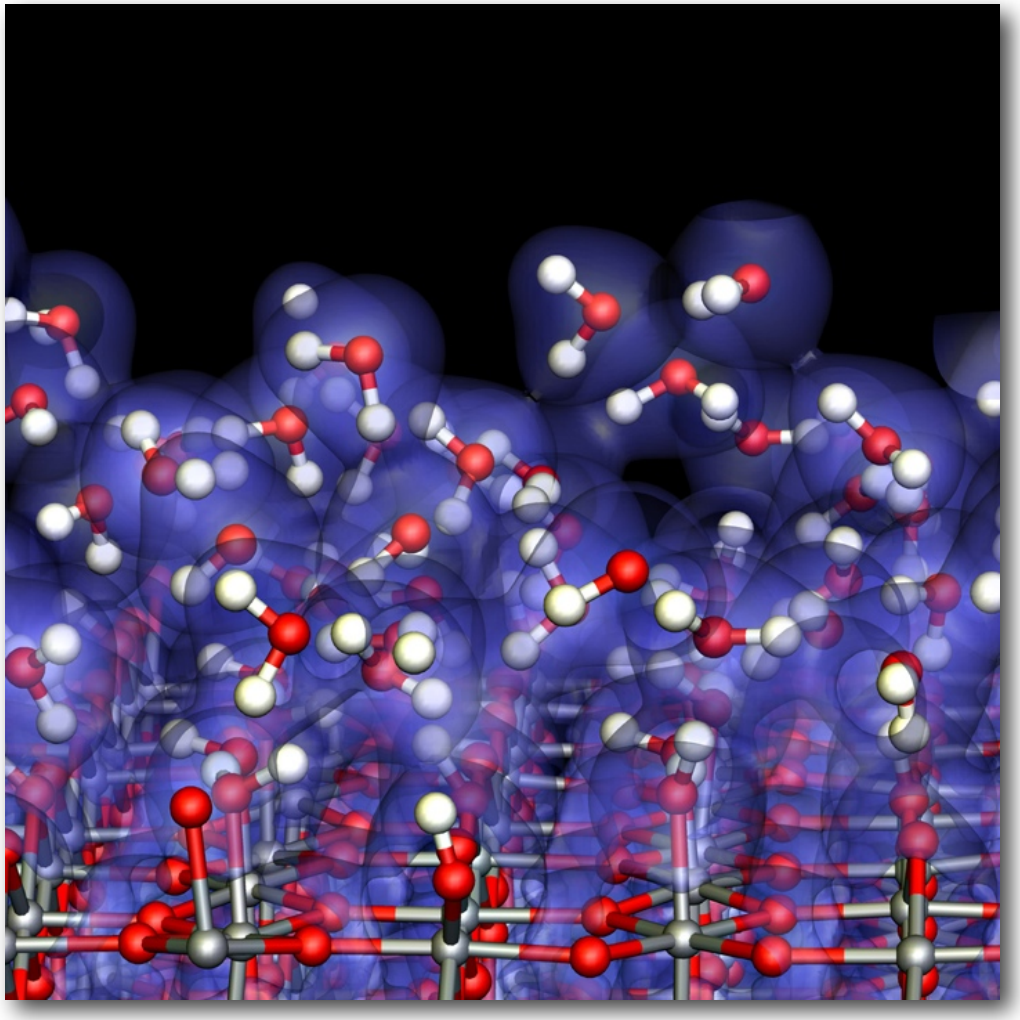
Long time dynamics: Water on TiO_2



- Combined molecular dynamics and neutron scattering study of proton dynamics at oxide surface
- Classical potential inaccurate for dynamics
- ~700 atom cell, 20000 MD steps, 2048 procesors, average ~1min/step including all real-world overhead
- 2 weeks continuous run/3 months timeshare

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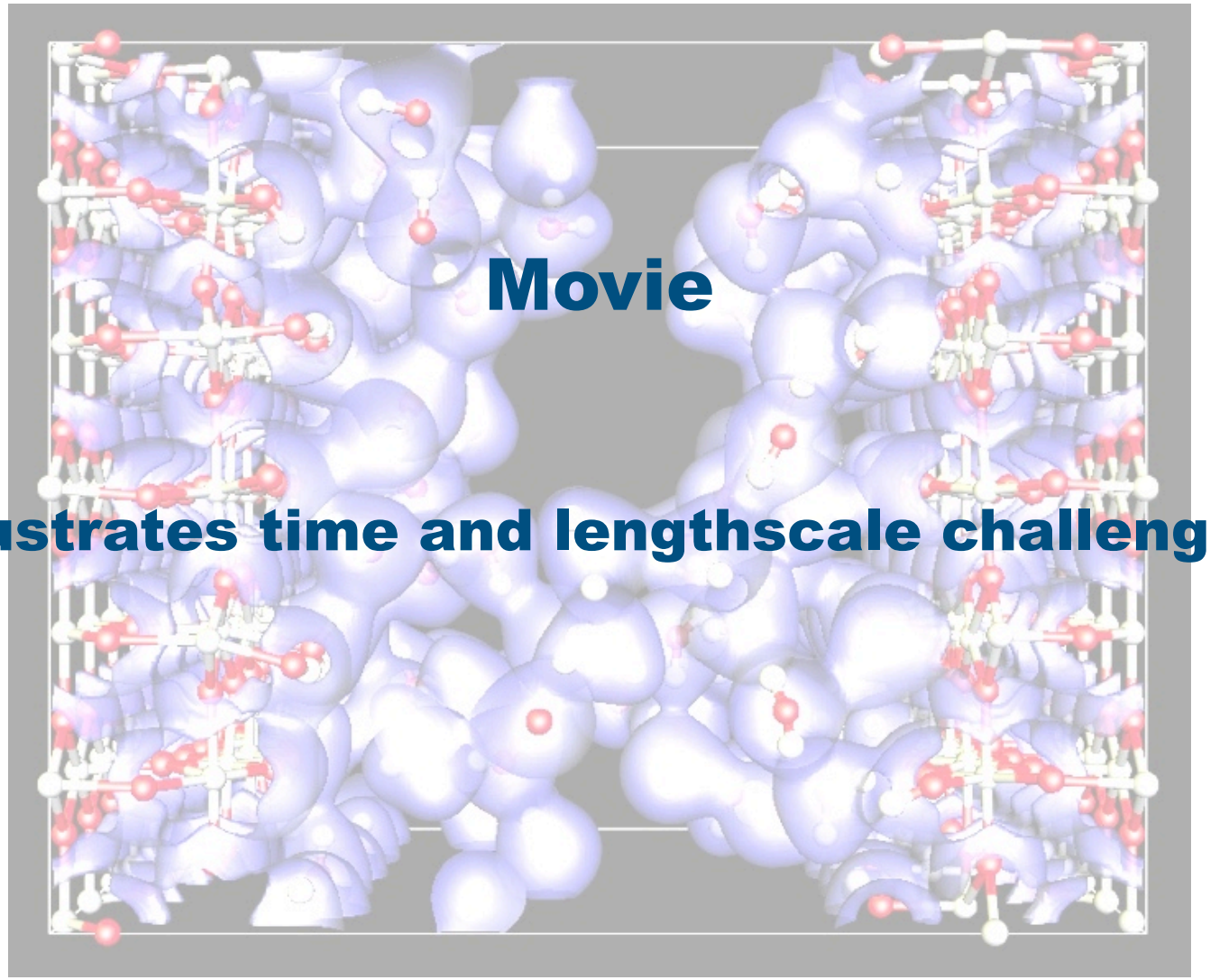

Movie

Illustrates time and lengthscale challenges

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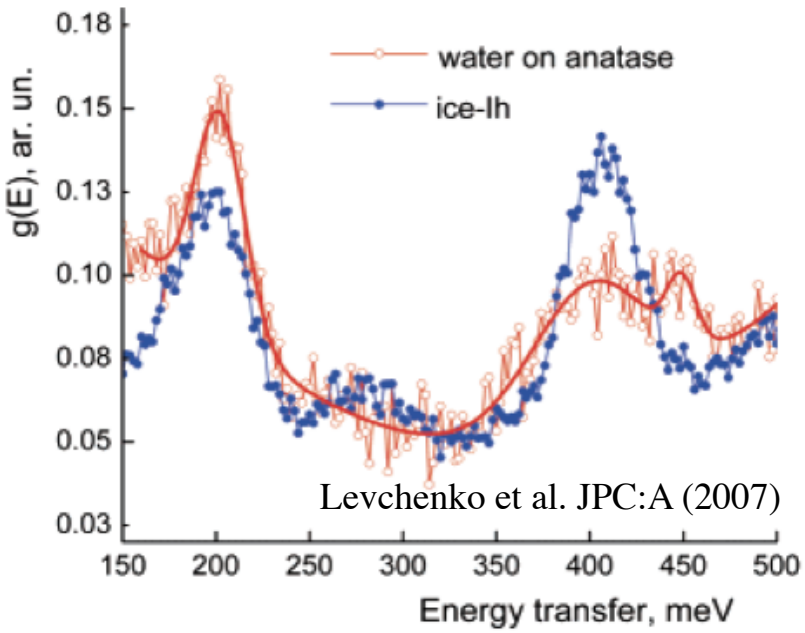
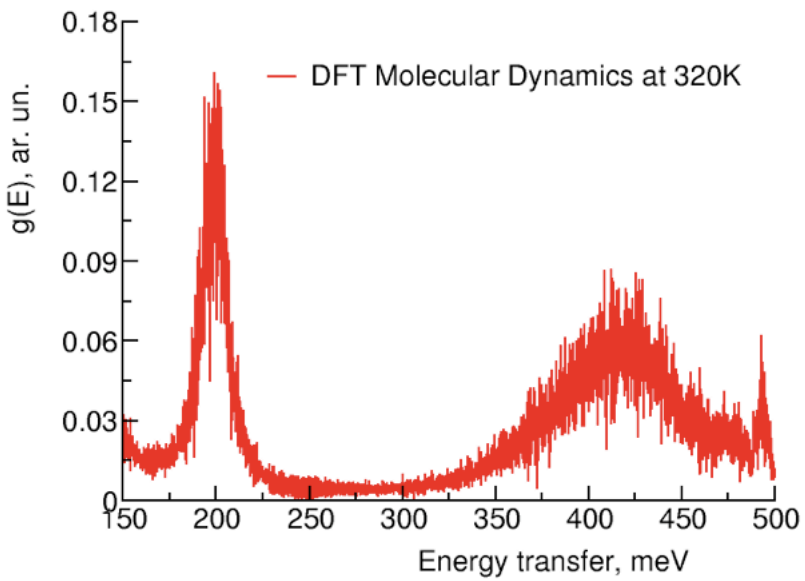
Vibrational density of states





- Near perfect agreement with experimental QENS results
- Enables cross-validation of theory and experiment
- Ability to deconvolve results, determine individual contributions to vibrational DOS
- Temperature dependent
- A general and promising technique: not specific to titania

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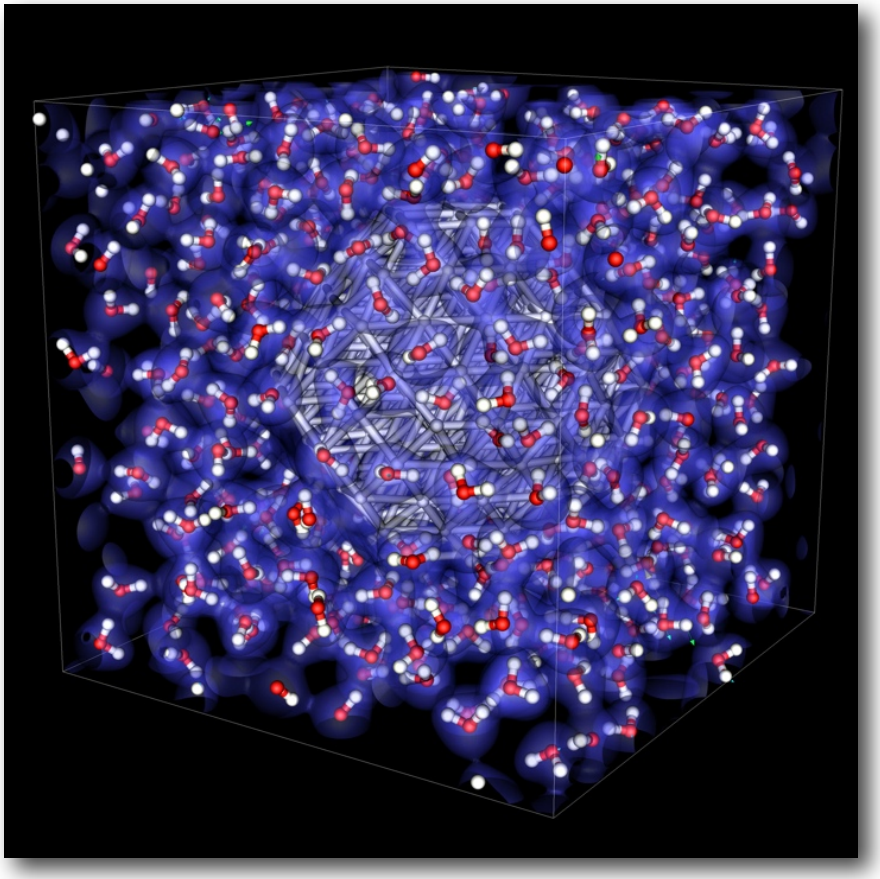
Nanocatalysis: Solvated Pt nanoparticle



Molecular dynamics
Pt_{201}+427H_2O 1482 atoms

- Aim to characterize change of atomic and electronic structure on solvation
- Follows rule of thumb: x1.5 speedup from 1024 to 2048 processors

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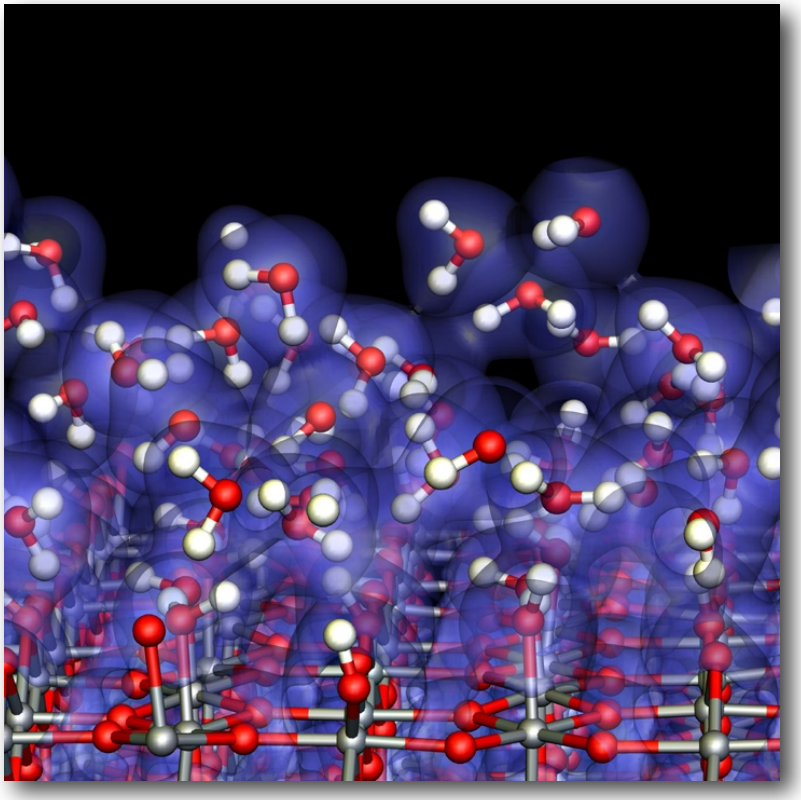
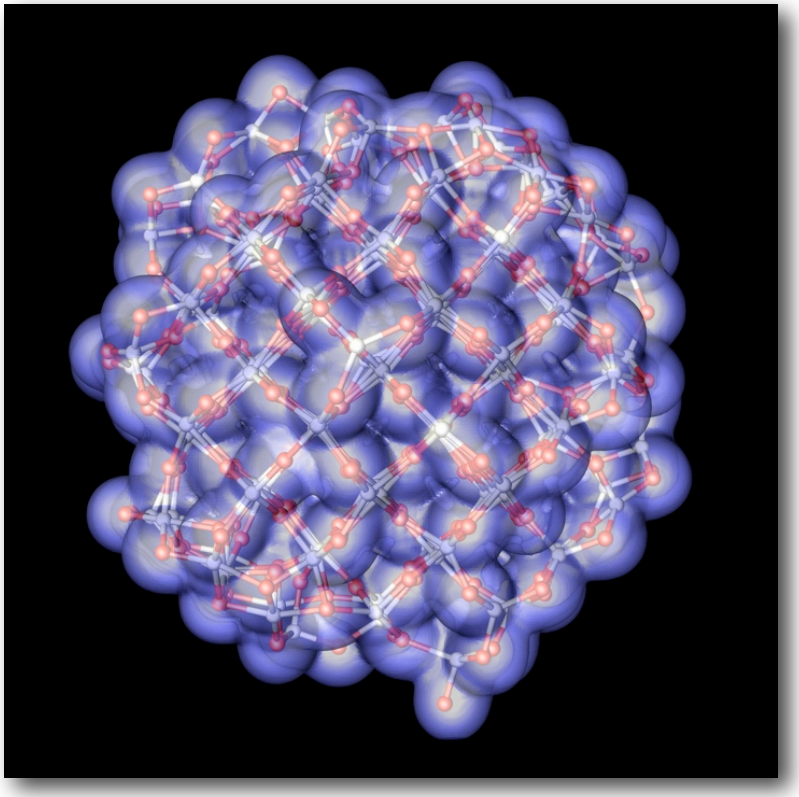
Short-term opportunity: nanoscale dynamics with DFT



Titania surface



Titania nanoparticle

Increased complexity of materials and geometries

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Challenges and opportunities

- The overlap of experimentally and computationally accessible length scales is a tremendous opportunity for science and technology
- Many opportunities to gain detailed knowledge of dynamic processes at the nanoscale, moving in the direction of designer materials
- Density functional investigations at the nanoscale - using existing methods - have large aggregate CPU demands (million CPUh) but will not generally run on 10^5+ processors. Productive science can be carried out - in the short term - at smaller scales

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