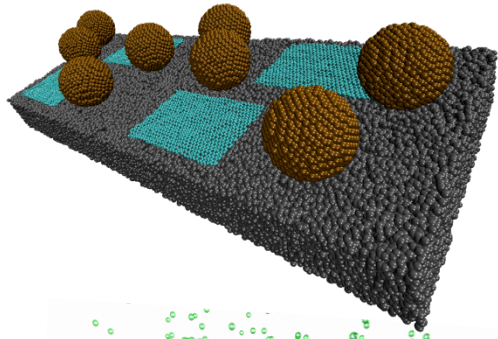
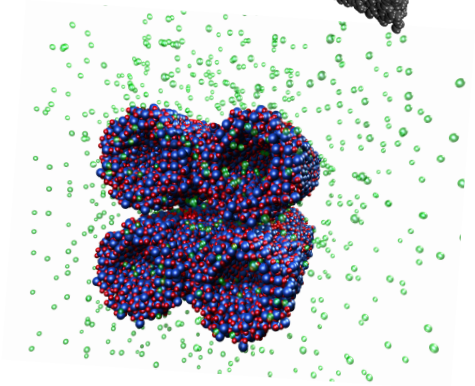
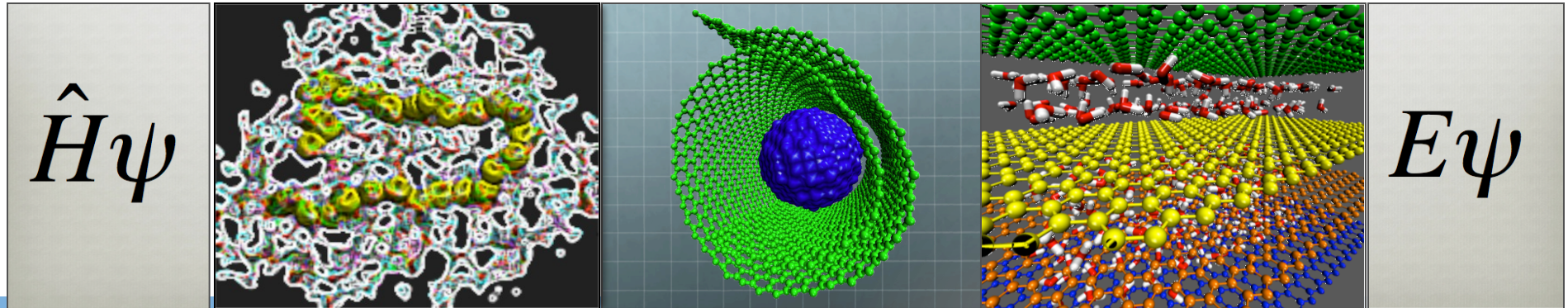
Pushing the Frontiers of Atomistic Modeling Towards Predictive Materials Design

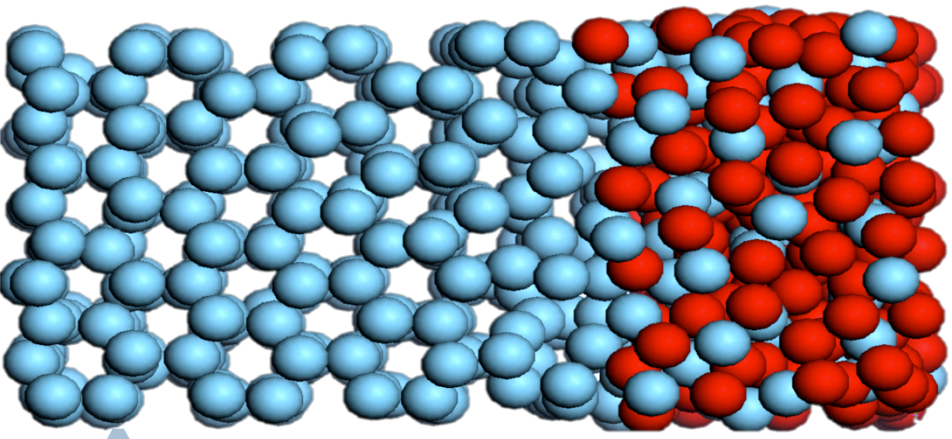




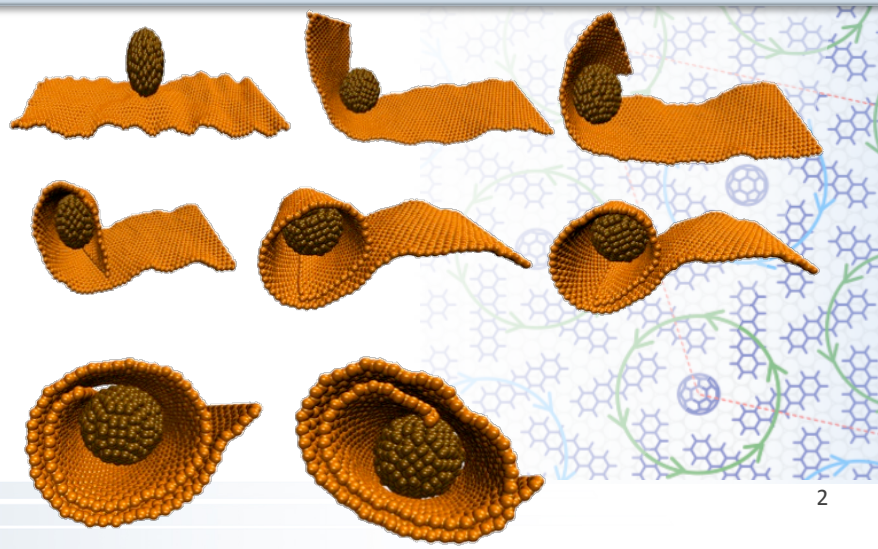
Subramanian K.R.S. Sankaranarayanan
Theory and Modeling Group
Center for Nanoscale Materials
Argonne National Laboratory



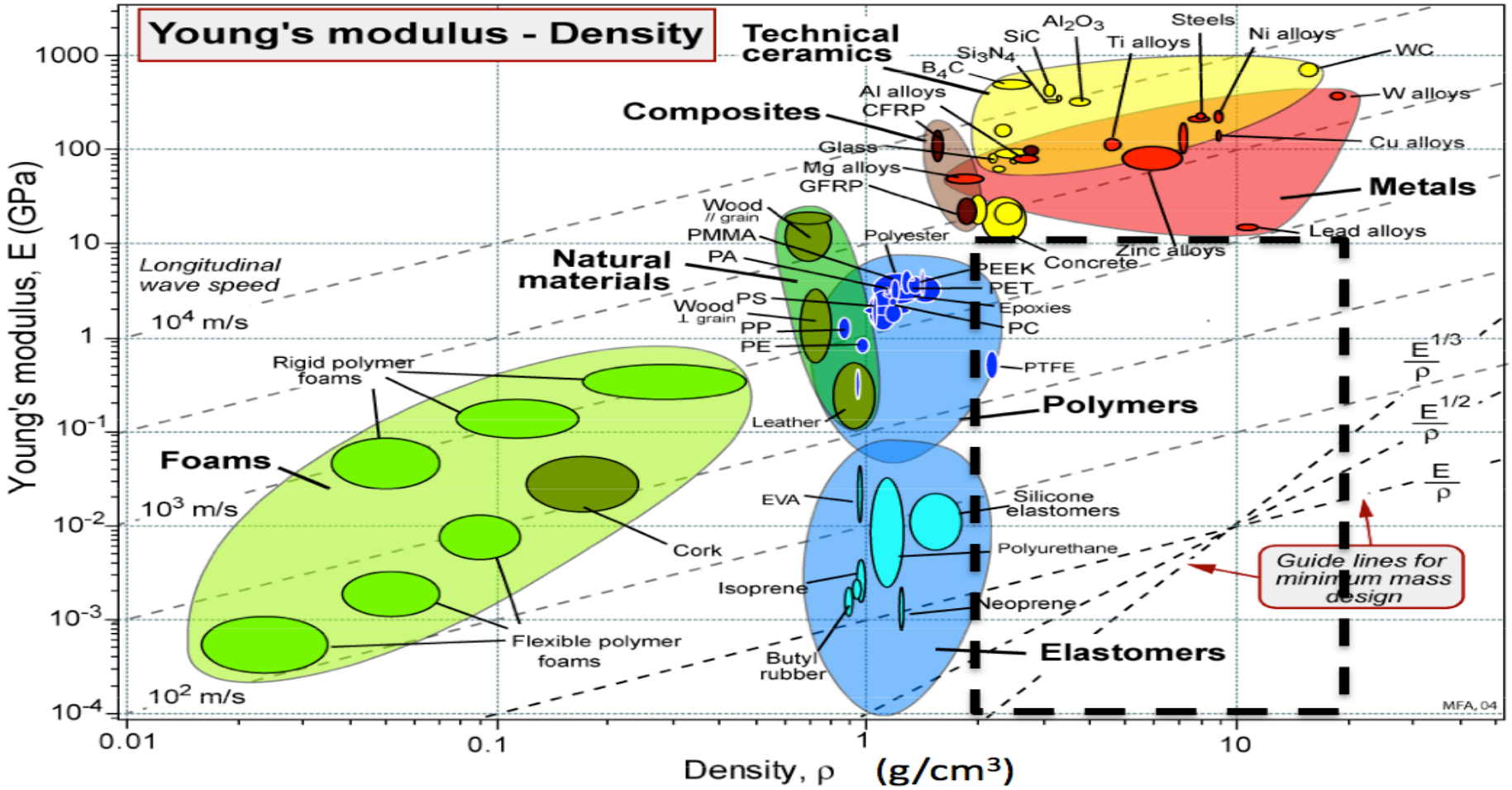
Molecular Conversion and Transport at Interfaces



Atomistic Origins of the Physical Properties of Nanoscale Materials



Materials By Design: Need for Hybrids

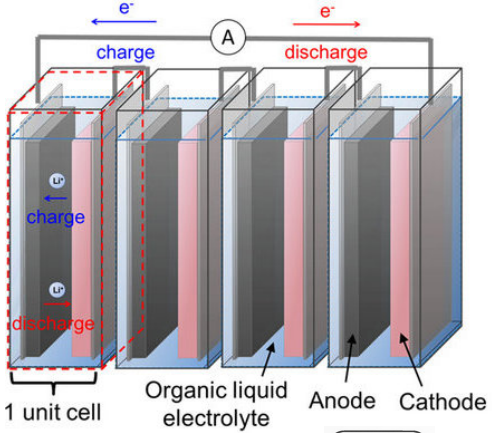


Different desired characteristics of materials are often in conflict in materials design ➔ HYBRID MATERIALS HOLD THE KEY

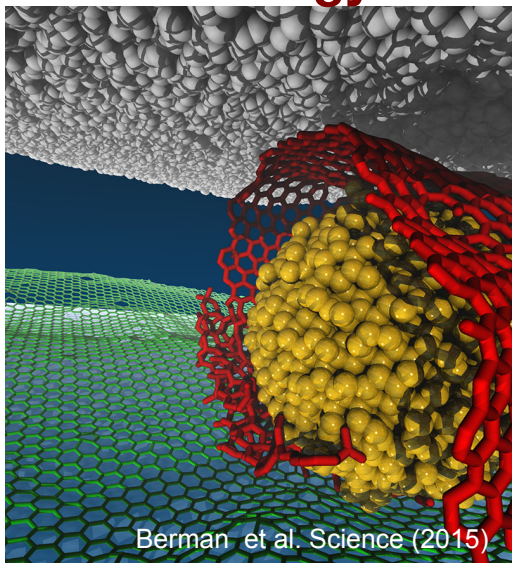
Ashby Materials selection in mechanical design (2005)

Reactive interfaces underpin energy technologies

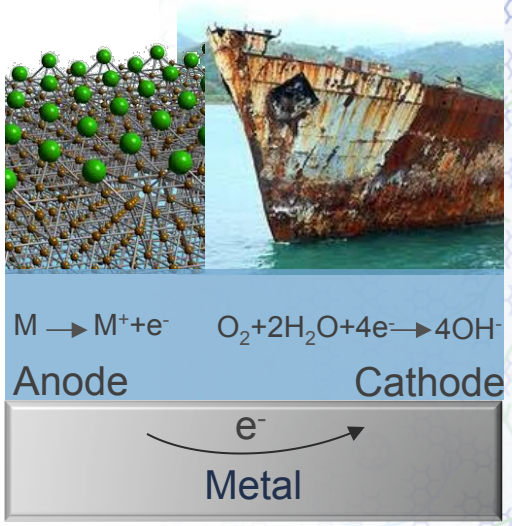
Li-ion batteries



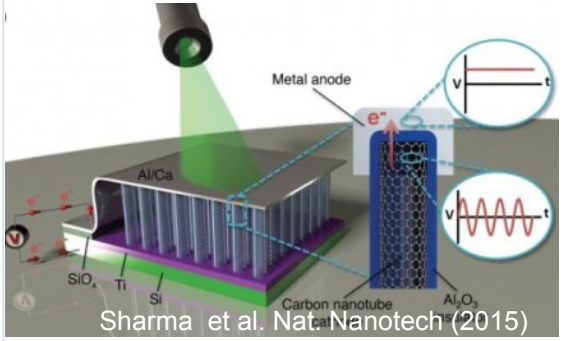
Tribology

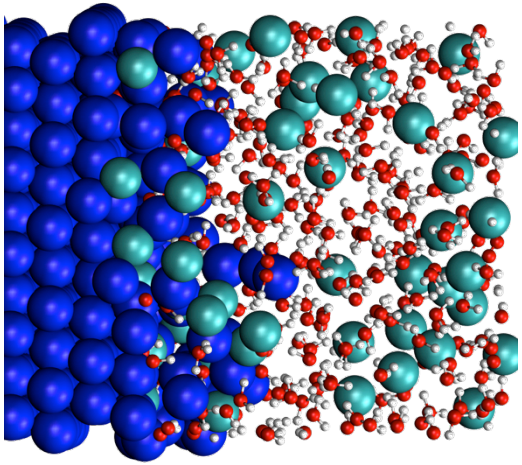


Marine corrosion

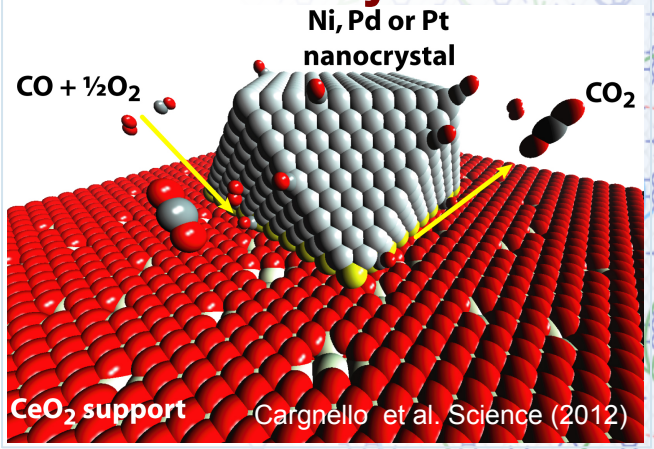


Photovoltaics





Catalysis

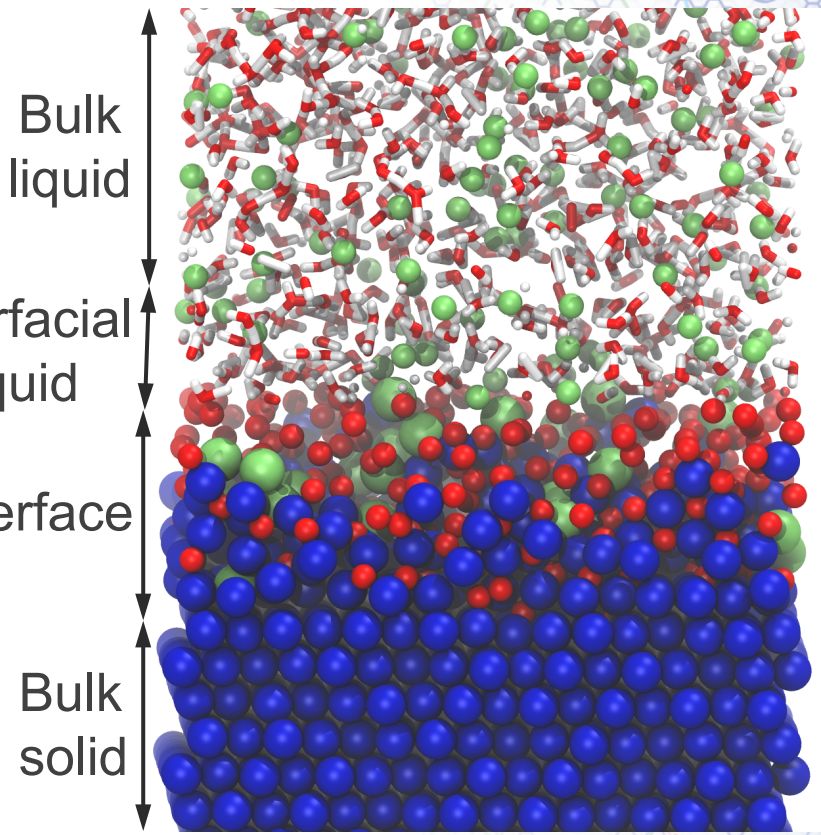


Solid-liquid interfaces

Interface

- Chemical reactions
- Adsorption

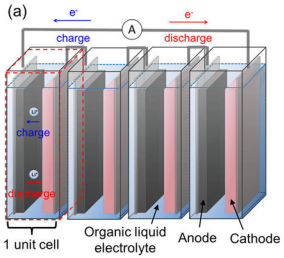
Structure (defects etc.), composition, and morphology of the interface strongly influence its functionality



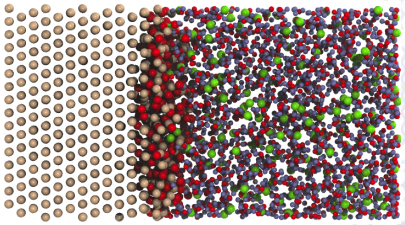
Interfacial water

- Solvation dynamics

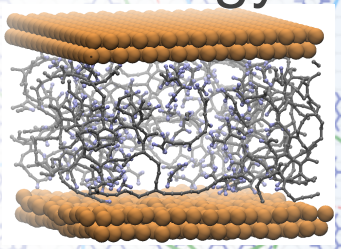
Batteries



Corrosion



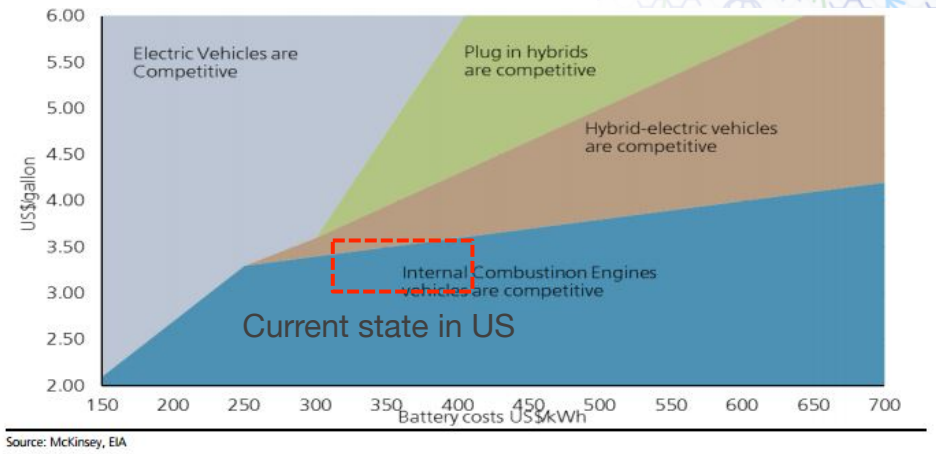
Tribology



Why should we understand reactive interfaces?

Energy Storage

Development of cost-effective, high energy density batteries is essential to enable complete electrification of road transport



Aqueous Corrosion

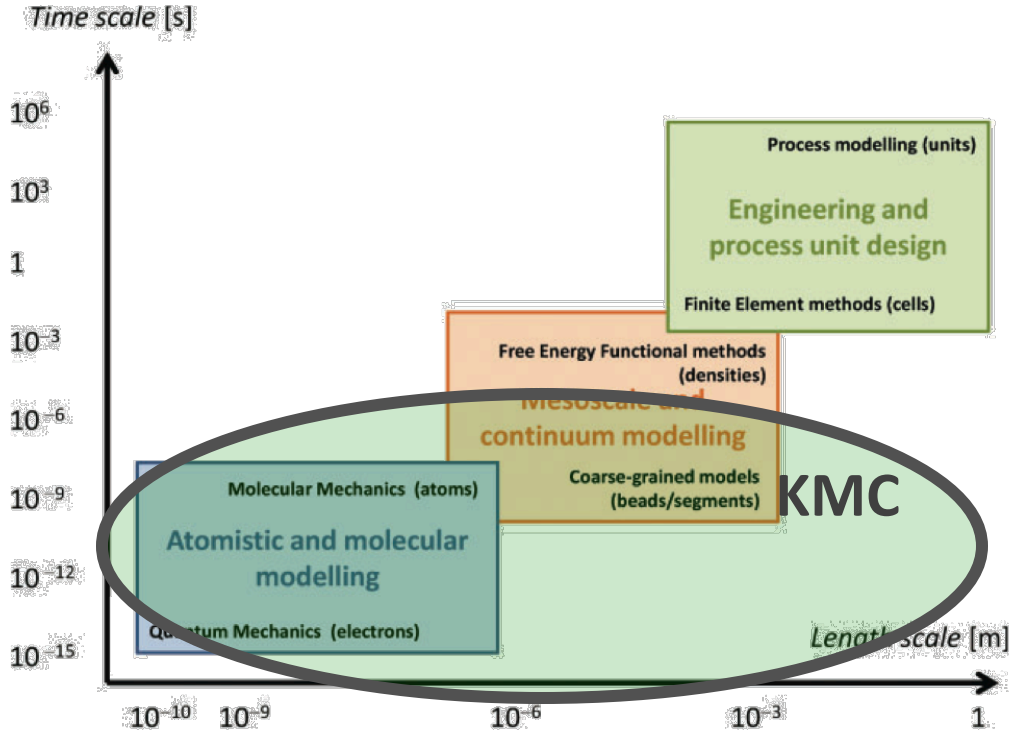
Corrosion cost the US economy 1.1 trillion dollars (5% GDP) in 2015
-NACE International (2015)

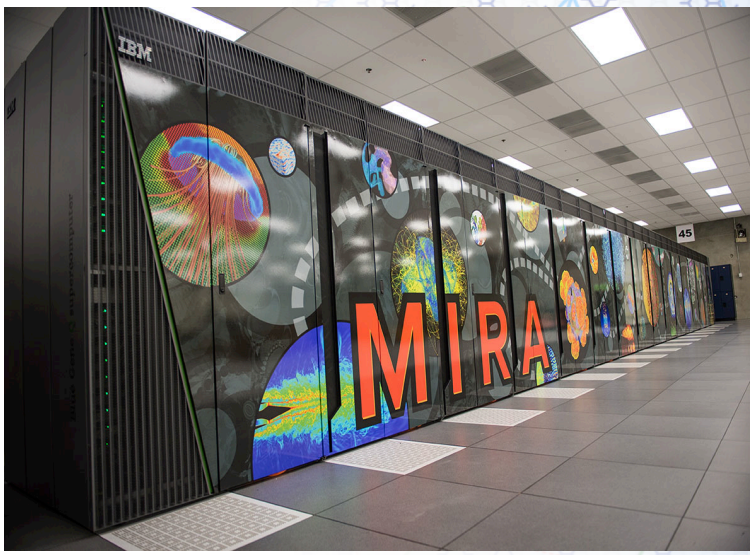
Tribology

1.2 million barrels of oil are spent everyday just to overcome friction in car engines
-Tribology International (2012)

A fundamental understanding of interactions at reactive interfaces is crucial to develop next-generation energy technologies

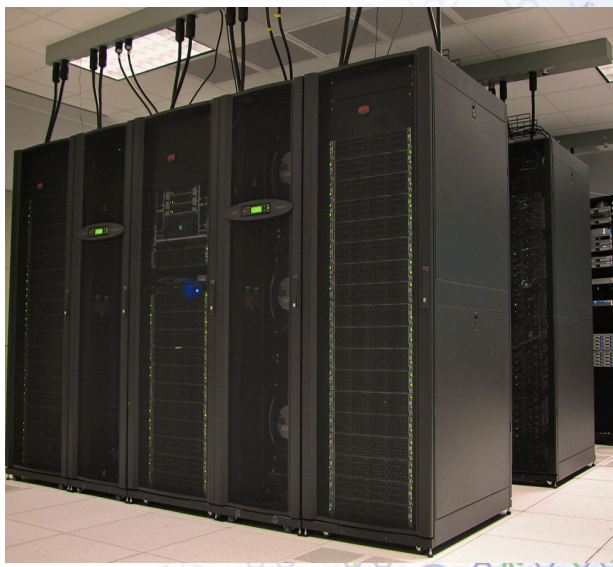
Our Approach





Method/Tool Development

- **Force Field Development:** Bridging the electronic and atomistic scales
- **Integrated imaging:** Ultrafast imaging with MD and data analysis
- **SIMOX for Reactive MD**



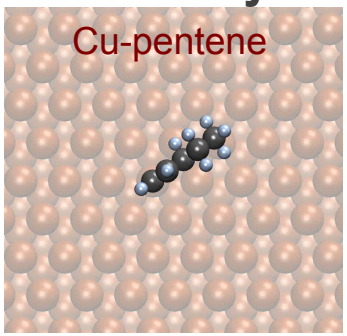
Carbon

International Materials Reviews 2011 **56** 207

Length and time-scales for materials modeling methods

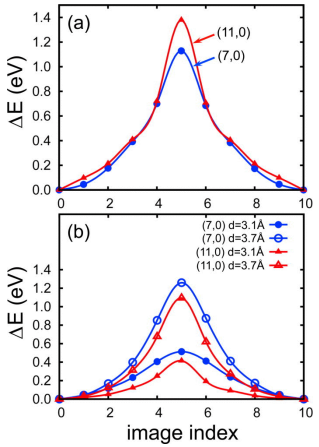
ps ns μs Time

Density functional theory



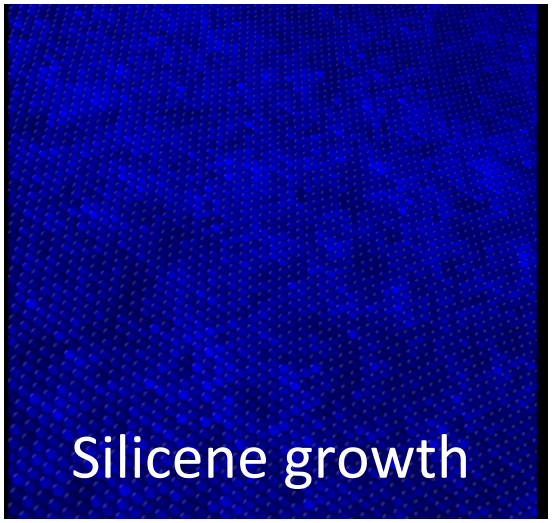
A.E, G. R., O.E., B. Narayanan et al . Nature (2016)

Li-diffusion pathways

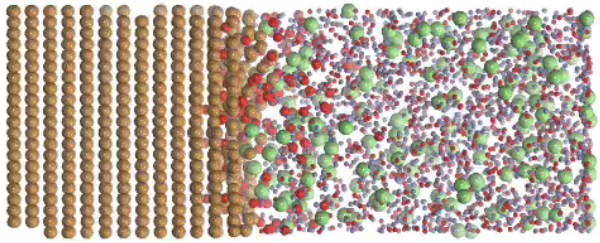


Yildirim et al. J. Phys Chem C. (2012)

Classical molecular dynamics



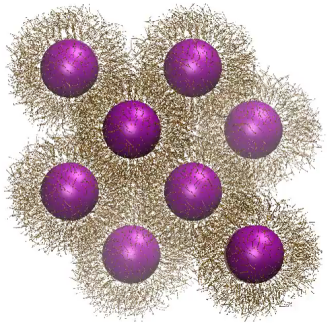
Aqueous Corrosion



(Invited) B. Narayanan et al., Electrochim Acta (2015)

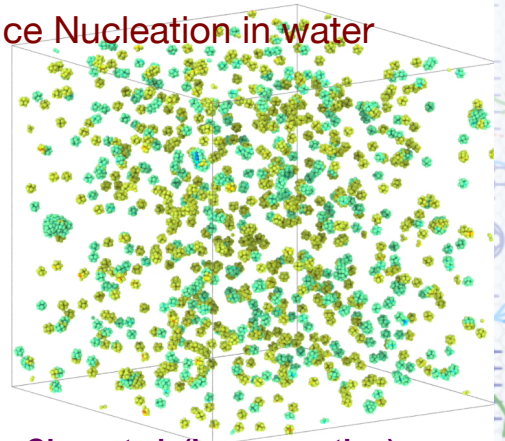
Coarse-grained models

Nanoparticle superlattices



B. Narayanan et al., In preparation (2016)

Ice Nucleation in water



Chan et al. (In preparation)

nm 100 nm μm Length

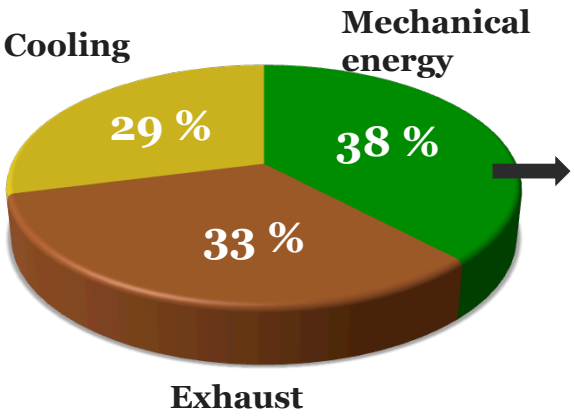
Overview:

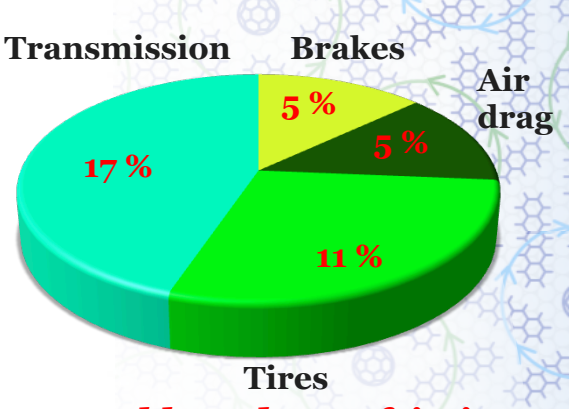
- **Extreme Computing and Big Data in Molecular Dynamics**
  - Size range (Few millions to billion atoms?)
  - Need for performing all atom simulations
  - Properties (Mechanical properties, phase transitions, Chemical dynamics etc)
- **Representative Examples:**
  - Friction laws at the mesoscale (Graphene-Diamond like carbon-Diamond)
  - Design of Clean Operando Lubricants for Anti-wear protection
- **Limitations of Existing Force-Fields:**
  - Energetics
  - Dynamics
- **Our Data Driven Agnostic approach towards Force-Field Development:**
  - Bridge the electronic and atomistic length-scales
    - Non-reactive
    - Reactive
- **Modeling, Analysis and Ultrafast Imaging**
- **Future Directions/Perspectives**

Reducing energy losses due to friction is a grand materials challenge

Fuel energy dissipation

60 km/h





Fuel loss due to friction

Reduce friction ⇒ ***Superlubricity*** *(Almost zero friction)*

- *Save energy*
- *Reduce emission of toxic gases*

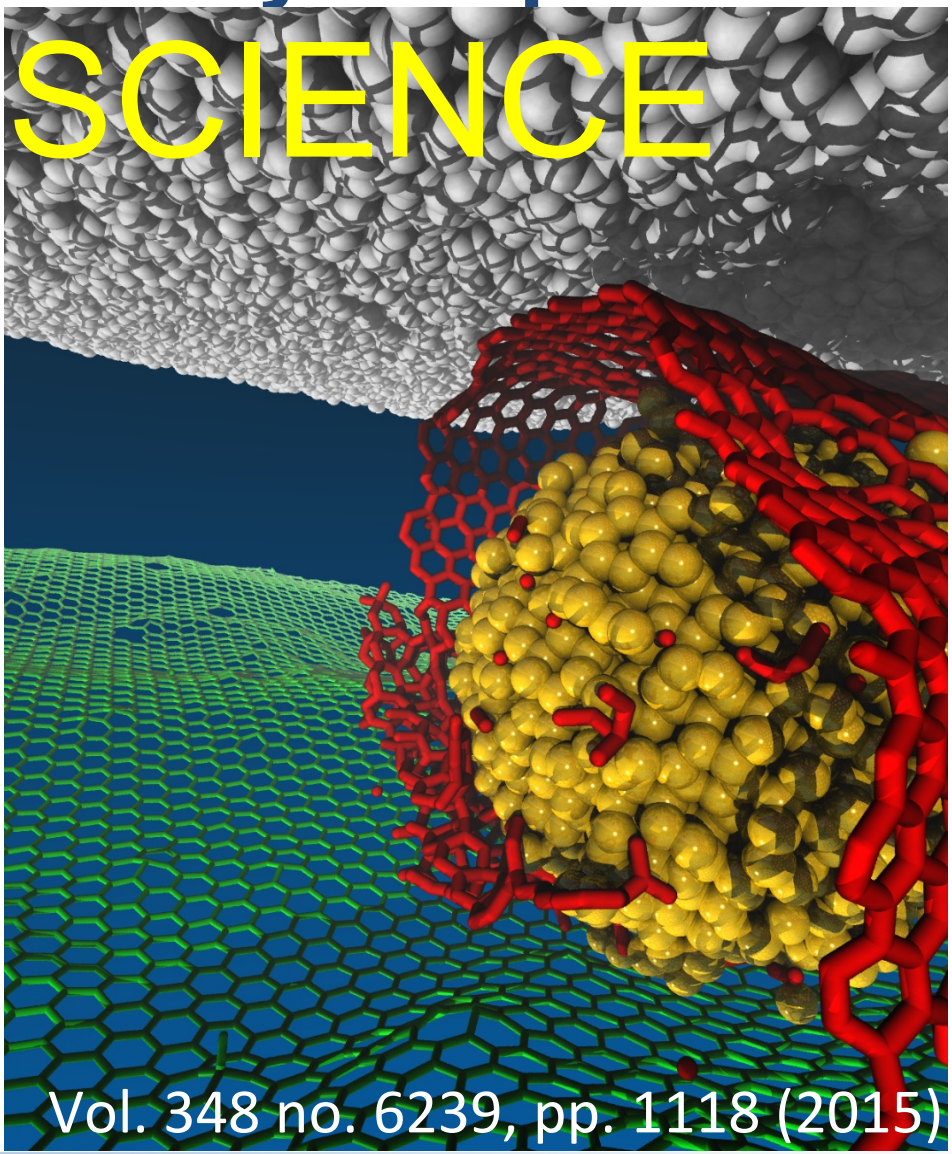
Atomic level understanding of friction

- Unified mechanism
- Mechanochemistry
- Surface chemistry

⇒ ***New materials***

Tribology International; 47, 221, 2012

Example 1: Macroscale Superlubricity Enabled by Graphene Nanoscrolls



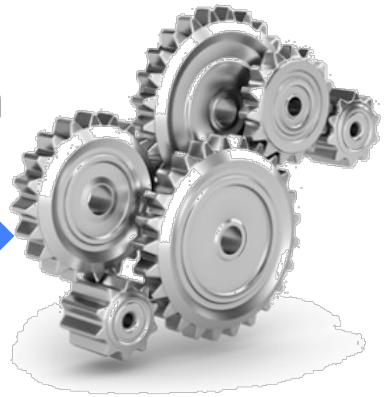
S. Deshmukh, D. Berman, A. Erdemier, A. V. Sumant and S. K.R.S Sankaranarayanan

Friction and Structural Superlubricity

After use

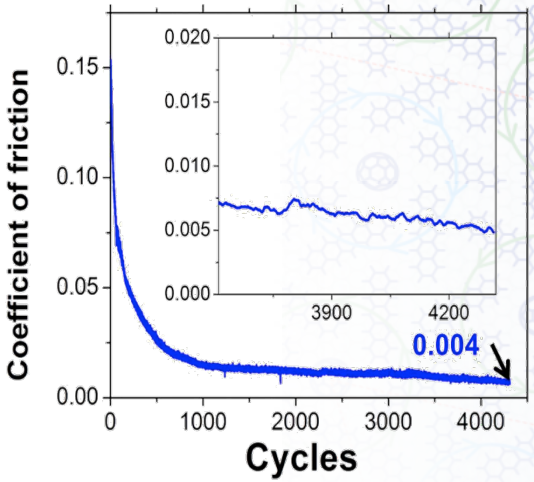
New lubricant ?

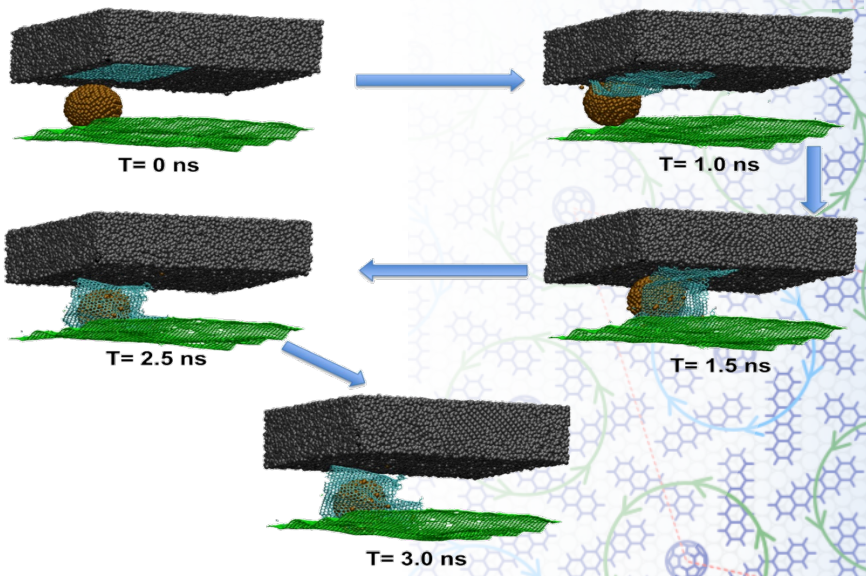
Million cycles



=> Friction reduction remains a major challenge

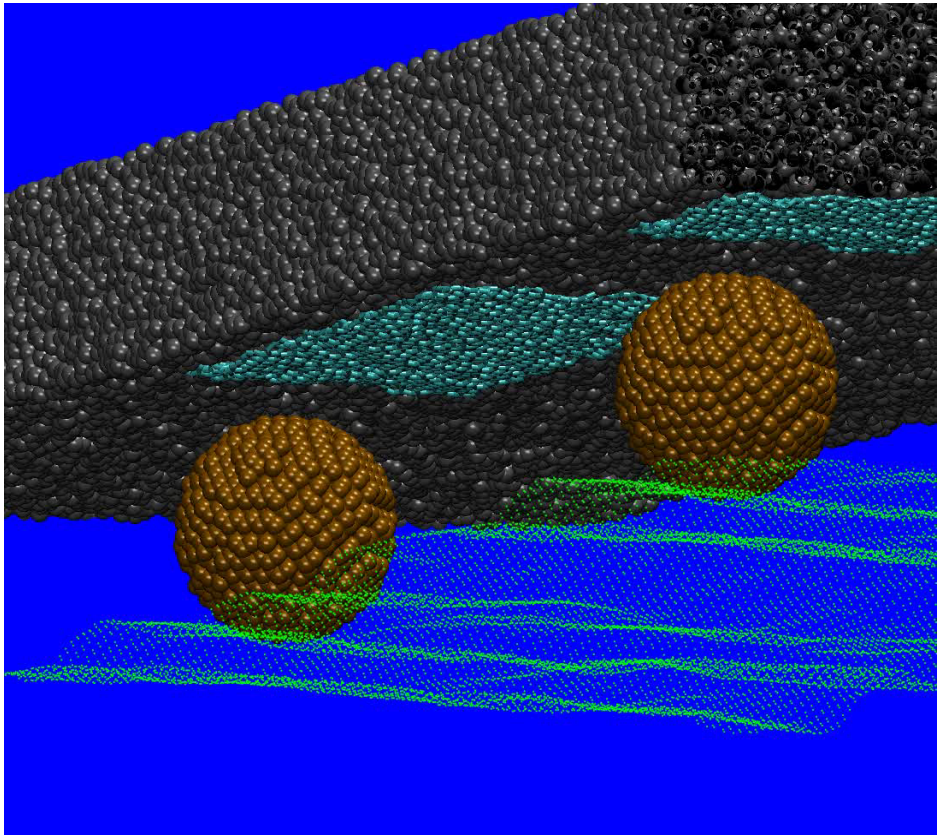
Our Solution



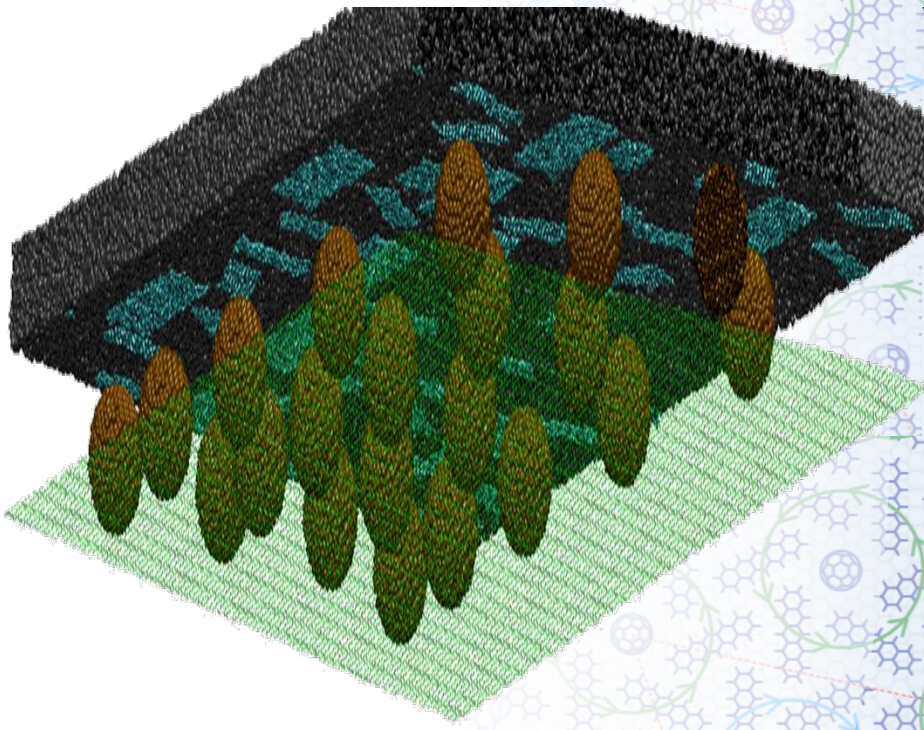


System Size ~ 100,000 atoms

Scrolling of graphene flakes over nanodiamond

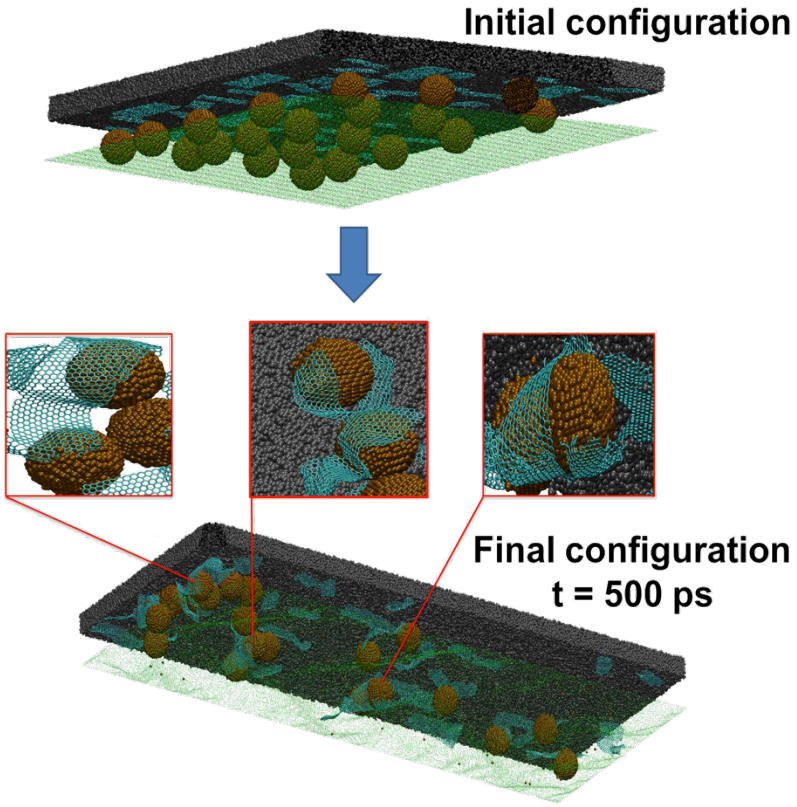


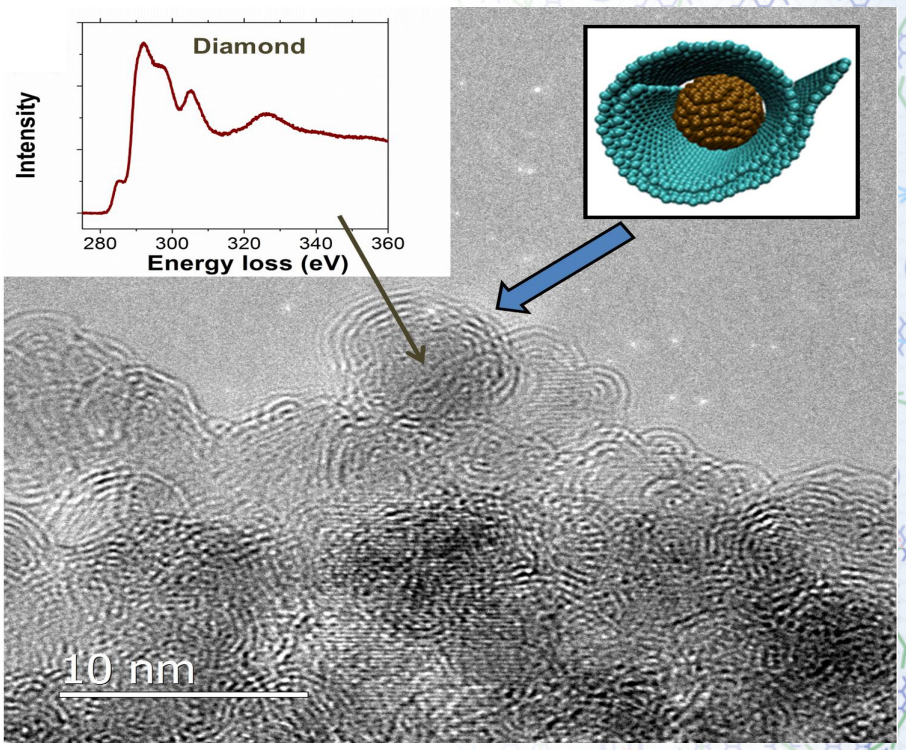
Nanoscale



Mesoscale

Mesoscale Link?

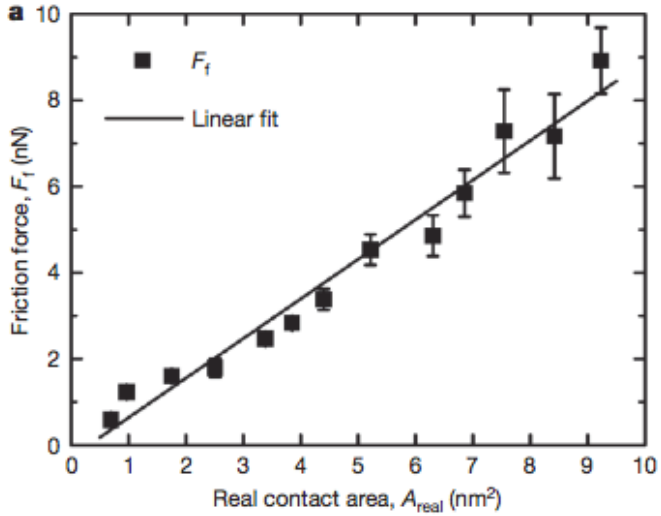


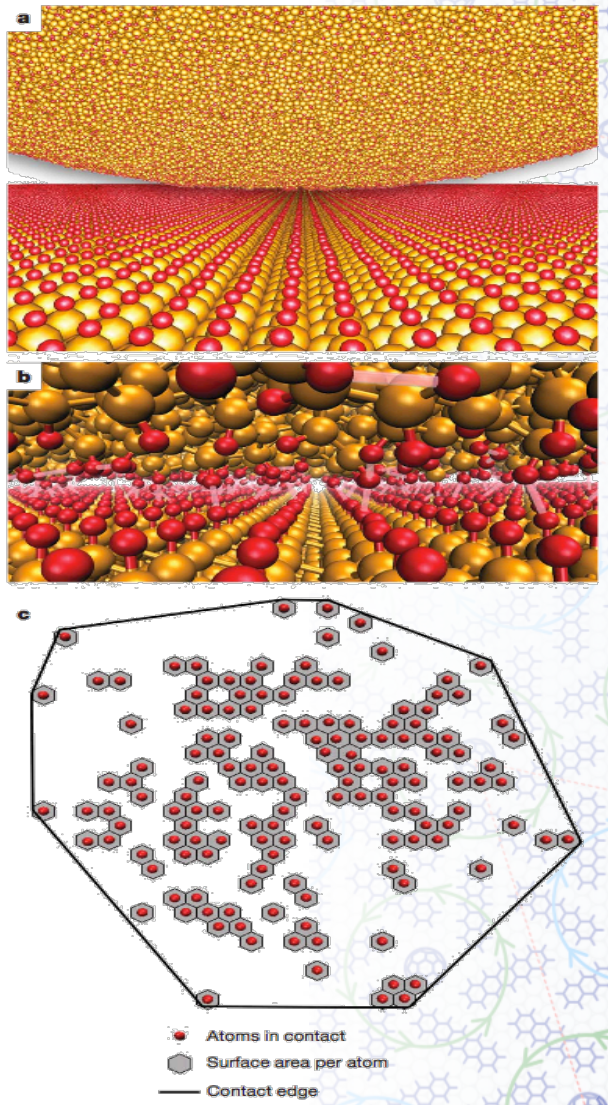


- Collective scrolling and tribological behavior evaluated at the mesoscale
- Scroll formation is consistent with TEM images of the wear debris

Friction laws at the nanoscale

- Single asperity theories break down at the nanoscale
  - Frictional force is not a linear function of the contact area of asperity
- Nanoscale contacts explained on the basis of the multiple asperity behavior
- Frictional force is proportional to the actual contact area (N_{atoms}*A_{atom})
- Contact area reduction can lead to superlubricity





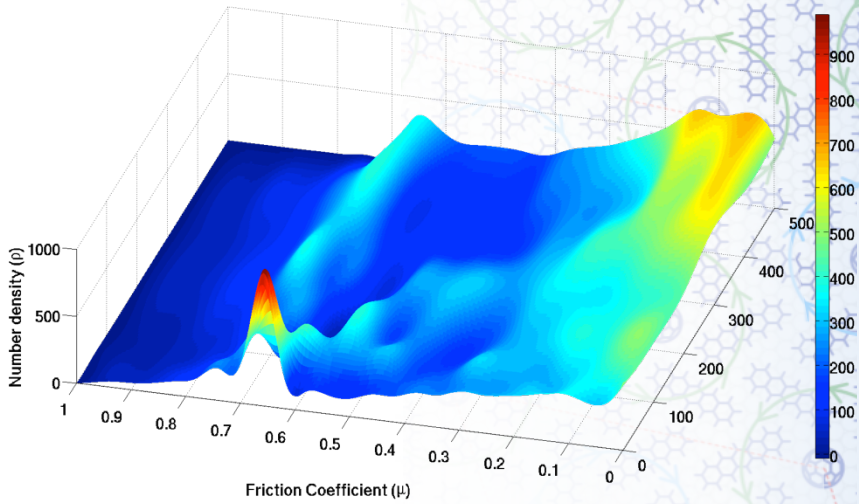
Szlufarska and co-workers, Nature, 457, 1116 (2009)

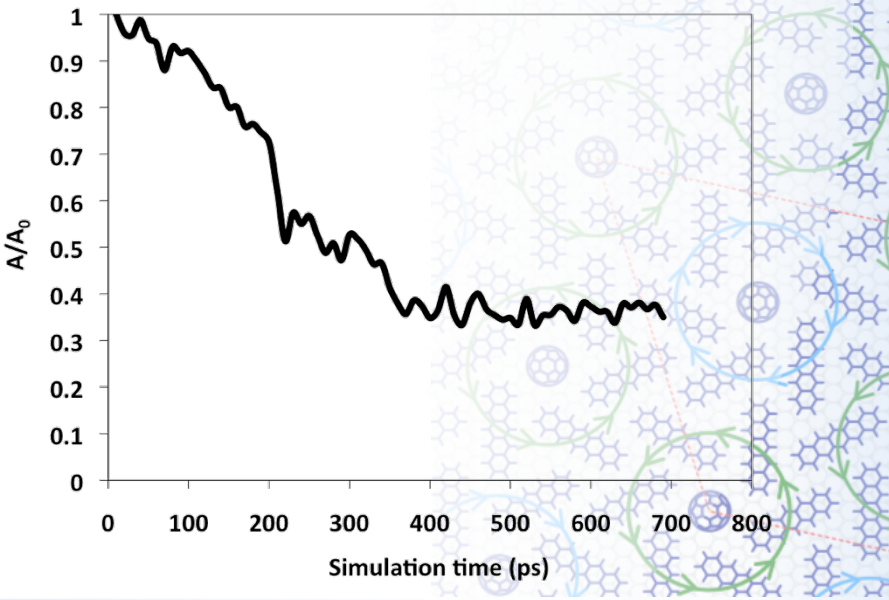
Friction laws at the mesoscale

- Initial distribution of the COF values shows most graphene patches are in a state of high friction
- Density profile shifts towards lower COFs
- Contact area reduction is significant at the later stages of sliding (~65-70%)

=> **Mesoscopic system is in a superlubric state**

- Friction mechanism at mesoscale is the same as nanoscale
- Tribological evolution from nanoscale to mesoscale is analogous to single vs. multiple asperity behavior





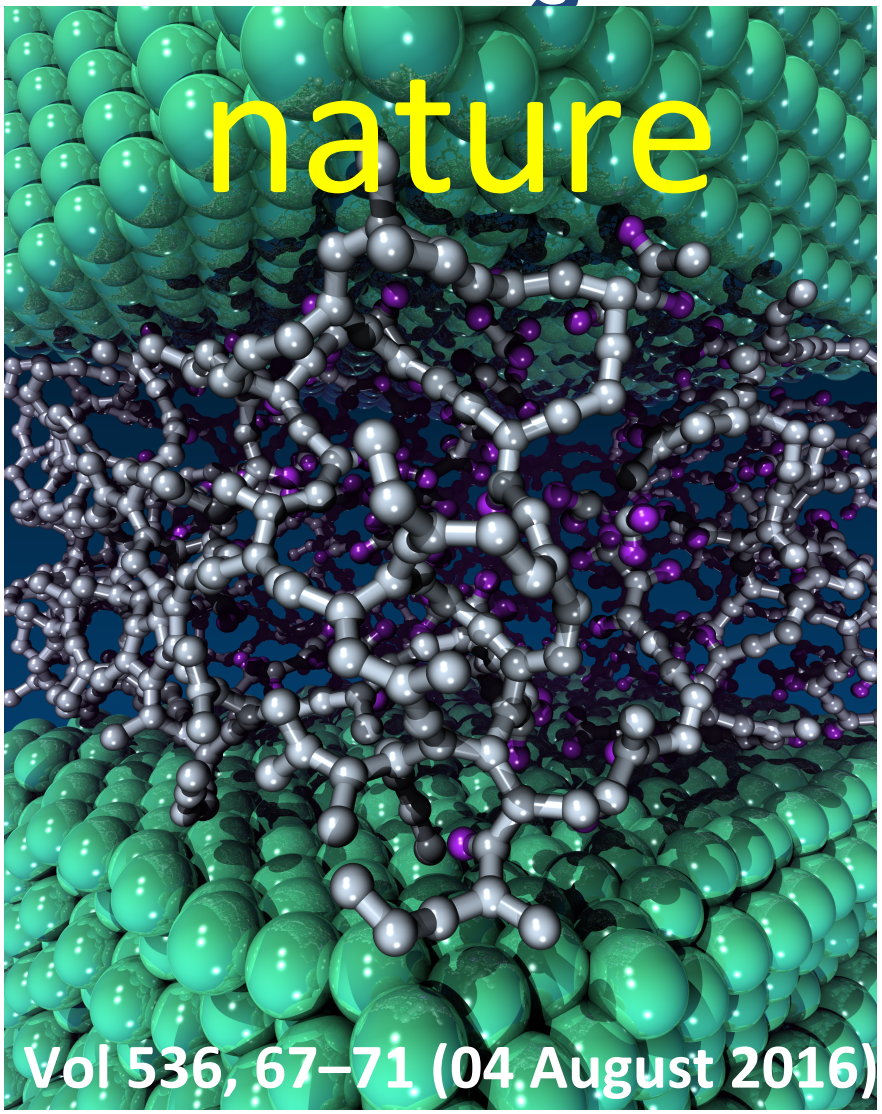
Summary

- First experimental demonstration of stable superlubricity at the engineering length scales
- Discovery of a new mechanism of nanoscale scroll formation that leads to macroscale superlubricity
- Diamond nanoparticles play a key role in stabilizing the superlubric regime
- Opens up new avenues for realizing macroscale superlubricity in real systems of industrial interest

Future directions:

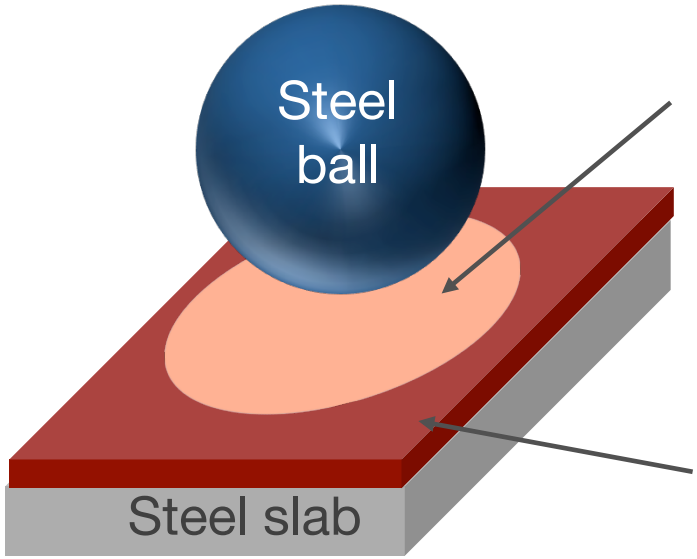
- Can we extend this to other 2-D materials like Boron Nitride, MoS_2 and others?
- Explore mesoscopic friction laws
- Explore materials phase space to identify other NPs candidates
- Study geometric effects of the nanoparticle (spheres, rods, tubes, facets)
- Can we realize superlubricity in humid environment?

Example 2: Tribocatalysis - Carbon based lubricating films from oil



B. Narayanan, G. Ramirez, A. Erdemier, and S. K.R.S Sankaranarayanan

Friction reduction is generally achieved by adding a lubricating film



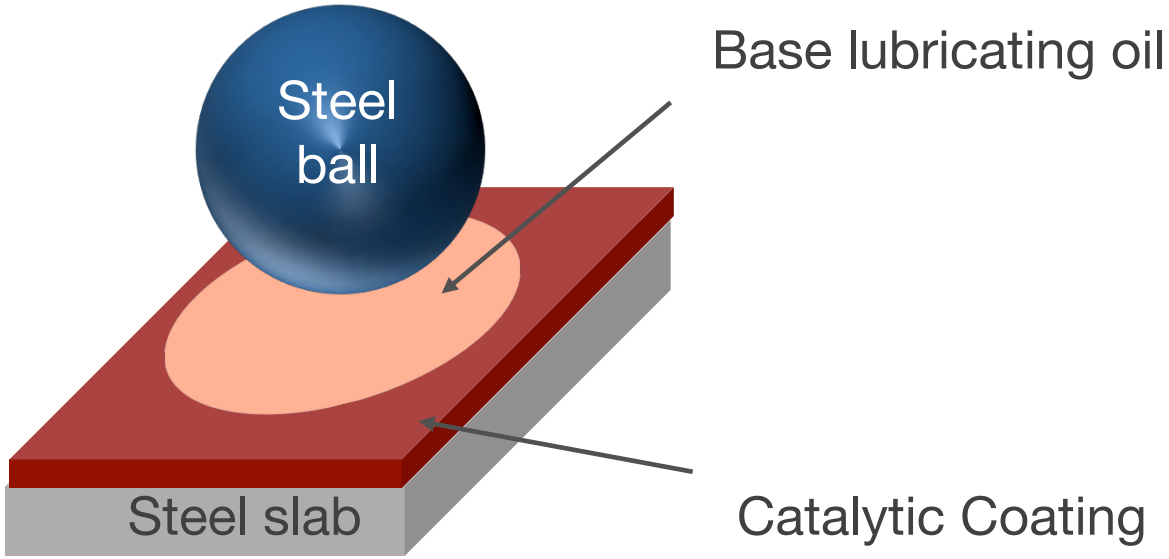
Liquid lubricants

- Reducing viscosity
- Additives (e.g., ZDDP, SAPS) environmental hazards

Low-friction coatings

- Adhesion problems

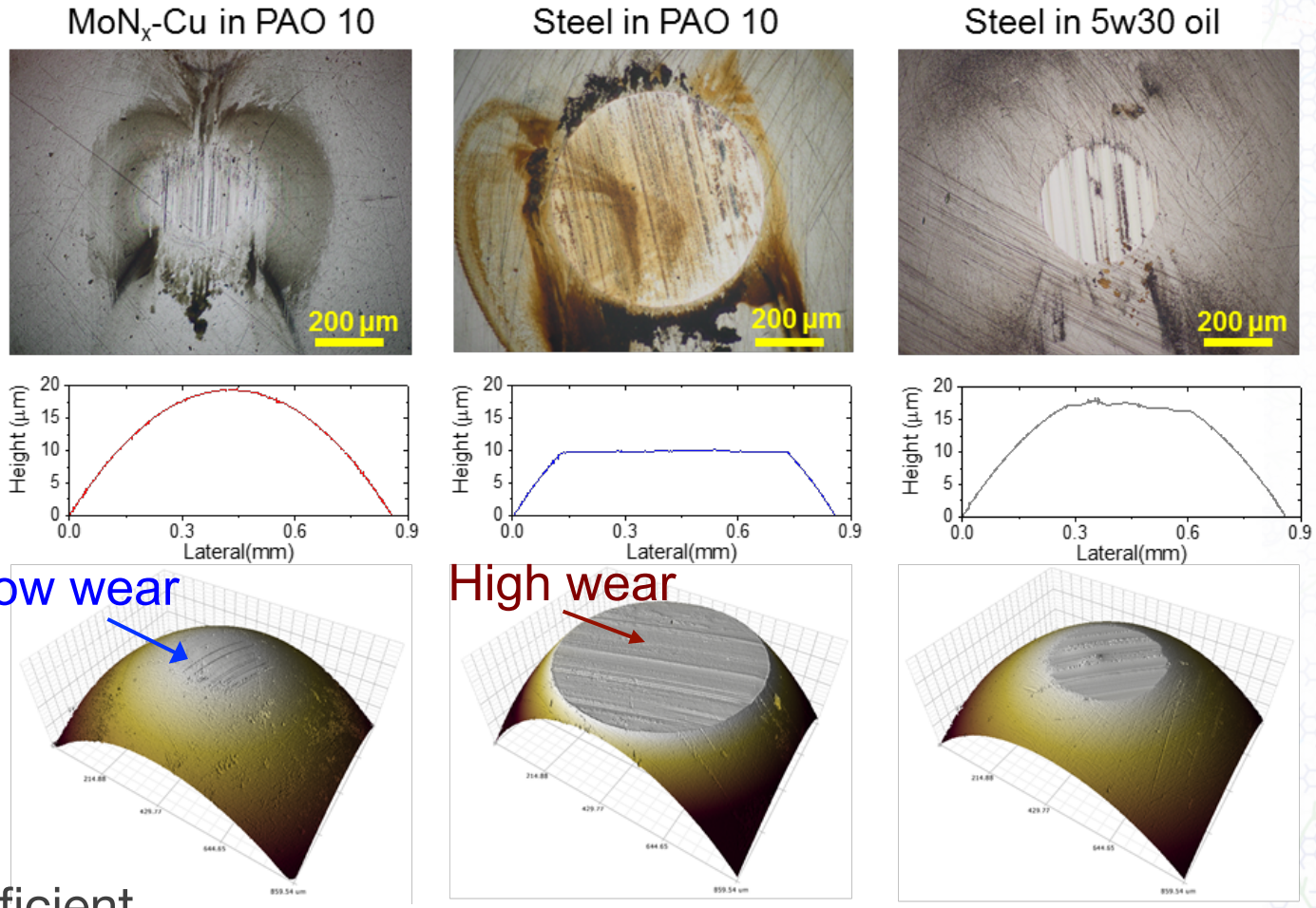
Tribology + Catalysis



Employ catalytic coatings to form solid low-frictions films from base lubricants during operation

A.Erdemir, G. Ramirez, O.Eryilmaz, B. Narayanan, G. Kamath, S. Sankaranarayanan, Nature, **536, 67–71** (2016)

MoN_x-Cu coating on steel shows minimal wear in base lubricating oil



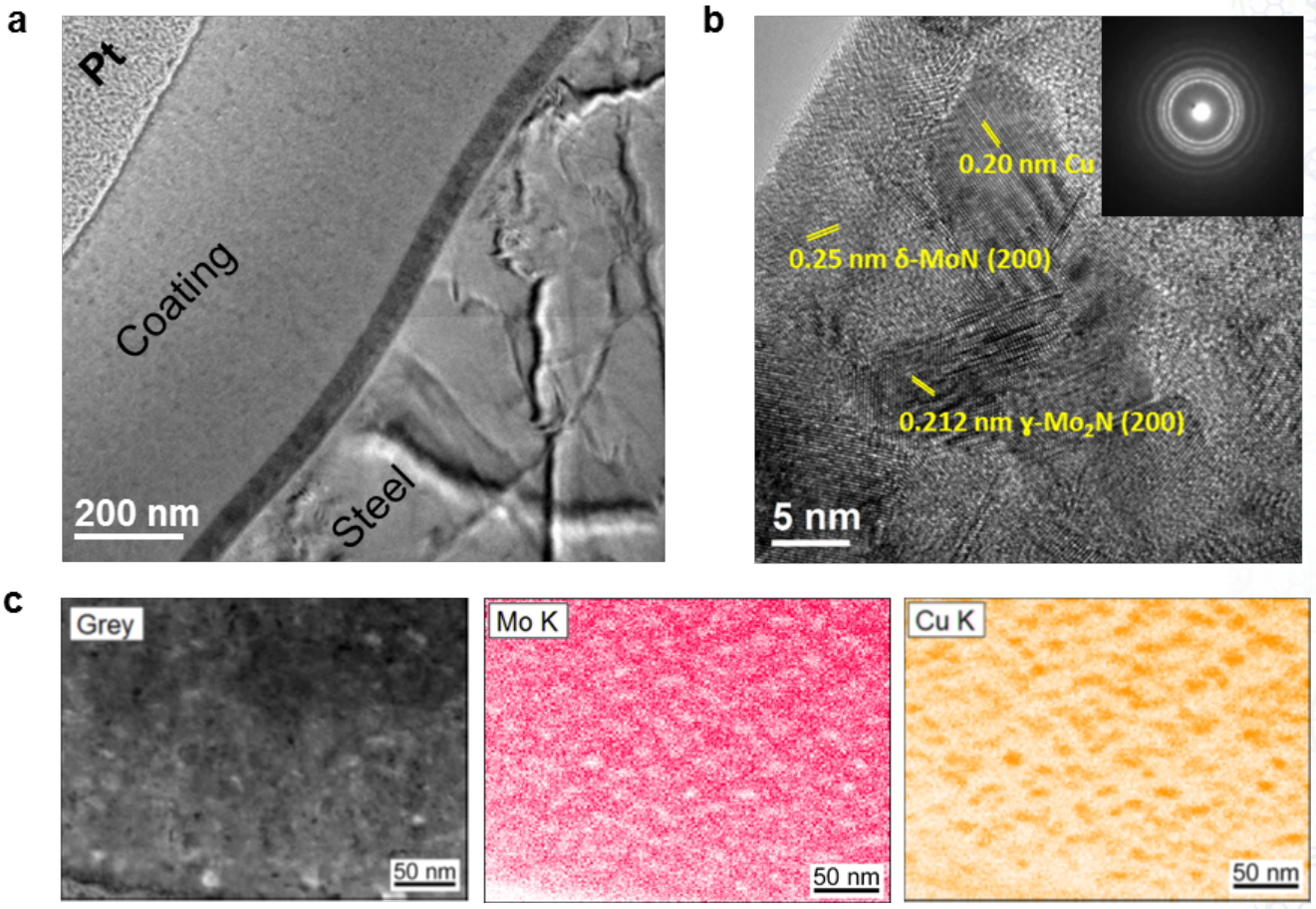
Optical Microscopy

2D/3D profilometry

Coefficient of friction

A.Erdemir, G. Ramirez, O.Eryilmaz, B. Narayanan, G. Kamath, S. Sankaranarayanan, Nature, **536, 67–71** (2016)

MoN_x-Cu coatings contain Cu rich regions



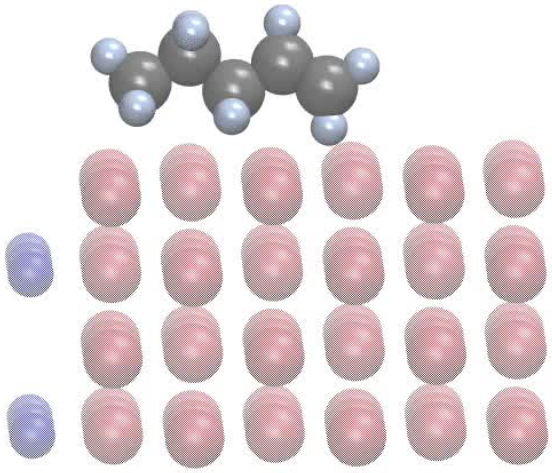
Atomic scale processes responsible for wear reduction are unclear

A.Erdemir, G. Ramirez, O.Eryilmaz, B. Narayanan, G. Kamath, S. Sankaranarayanan, Nature, **536, 67–71** (2016)

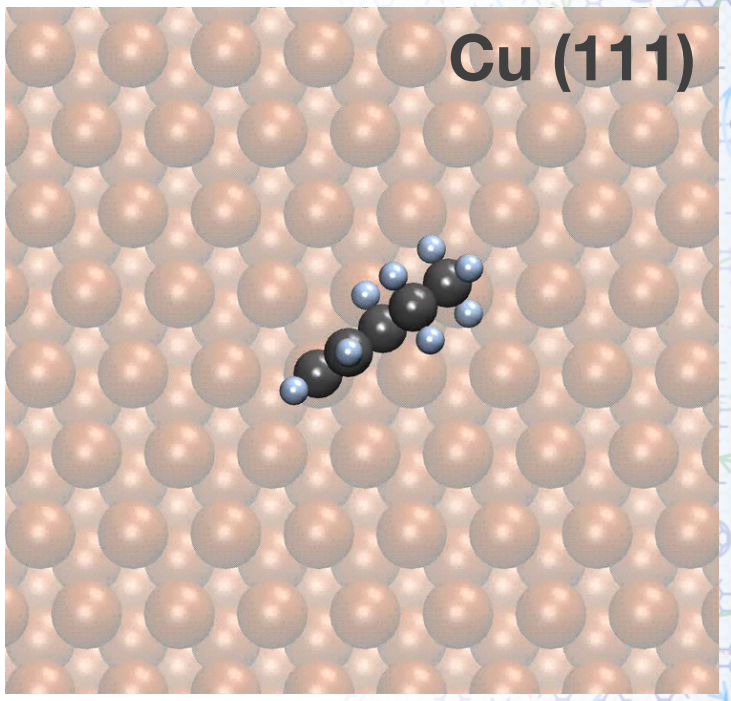
Ab initio molecular dynamics simulations

1-pentene on metal/metal-nitride surface for 10 ps at 1000 K

MoN (001)



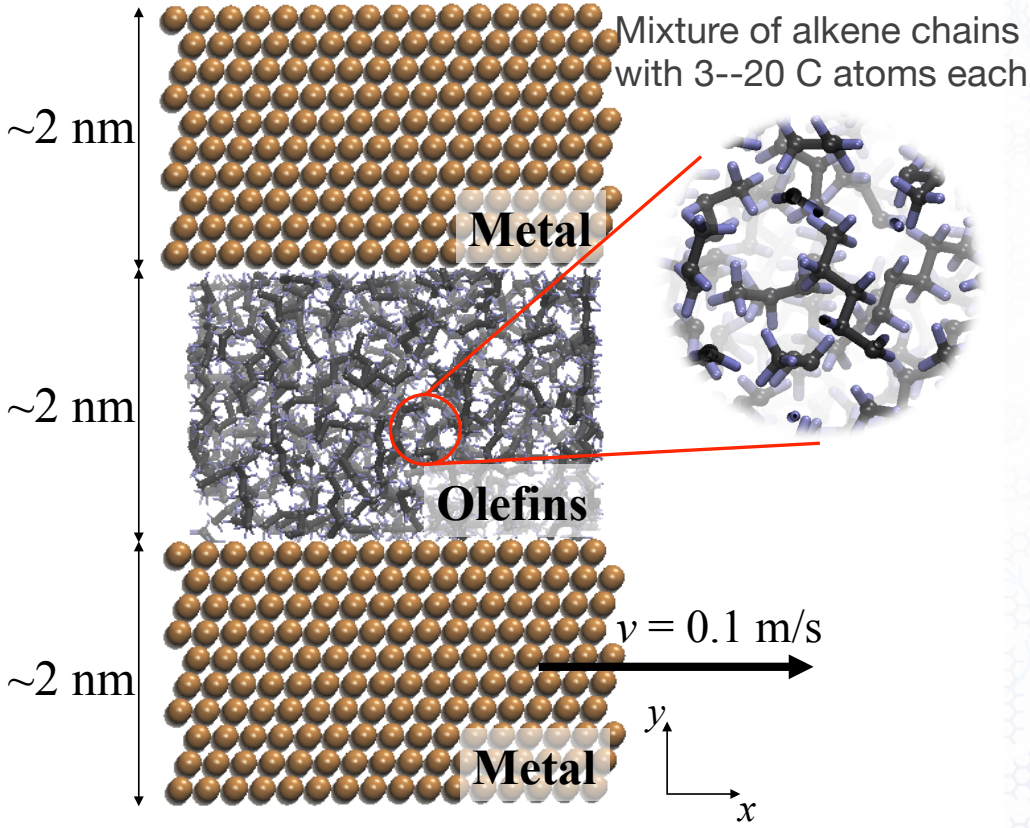
Cu (111)



- *MoN surfaces are inert*
- *Copper surfaces catalyze dissociation of olefins*

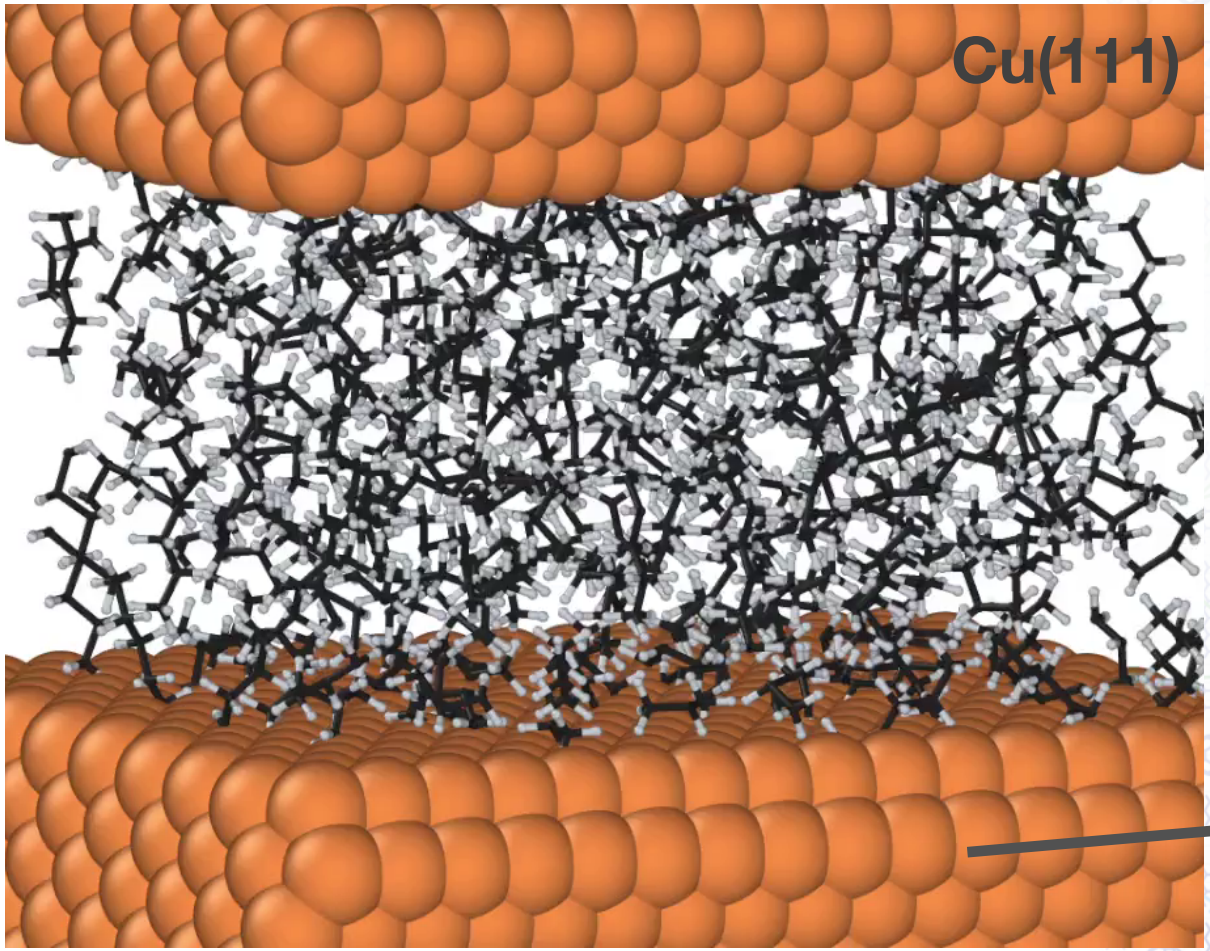
Reactive molecular dynamics simulations

ReaxFF based MD simulations of evolution of olefins between two sliding Cu surfaces for 2 ns at 1000 K



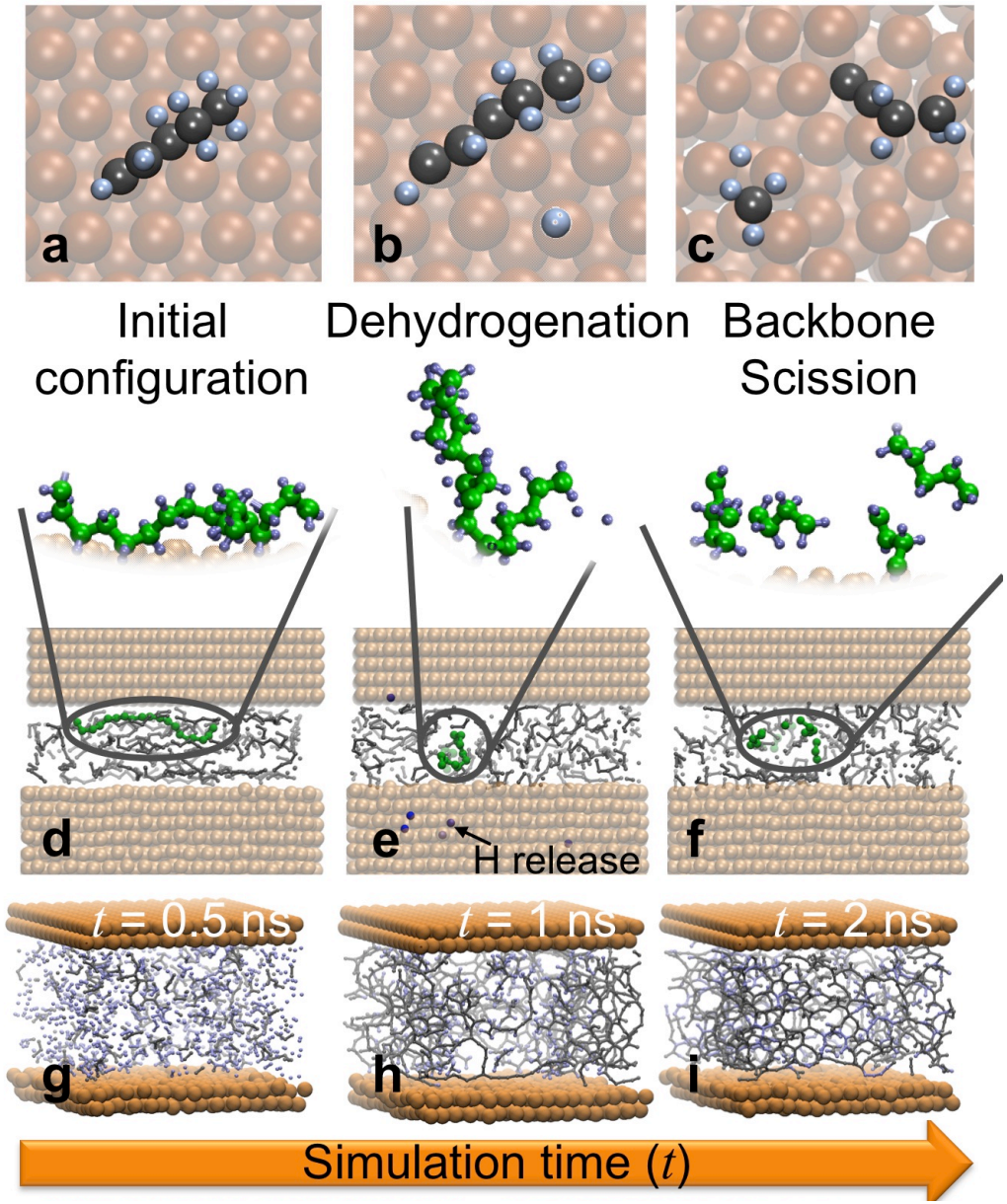
Reactive molecular dynamics simulations

ReaxFF based MD simulations of evolution of olefins between two sliding Cu surfaces for 2 ns at 1000 K



A.E, G. R, O.E, **B. Narayanan,** G. K, S. S, Nature, *Accepted for publication*, (2016)

Atomic scale processes governing carbon tribofilm growth

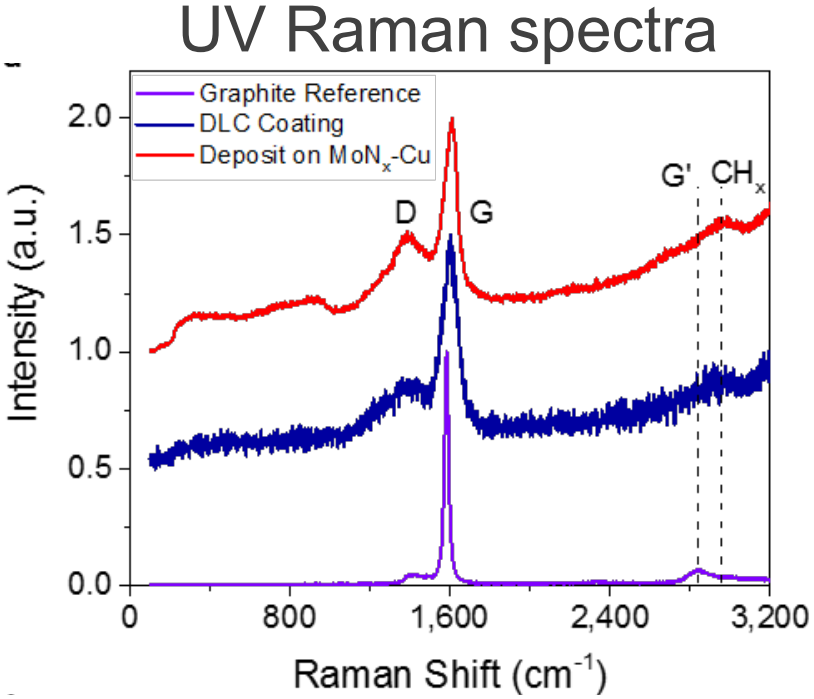


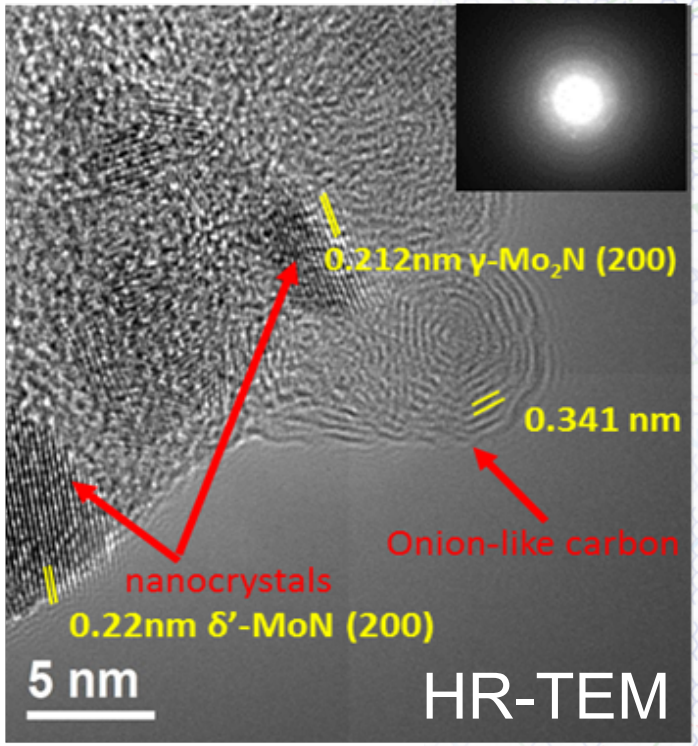
AIMD

Classical (Reactive) MD

A.E, G. R, O.E, **B. Narayanan,** G. K, S. S, Nature, *Accepted for publication*, (2016)

Experimental characterization of the formed tribofilms

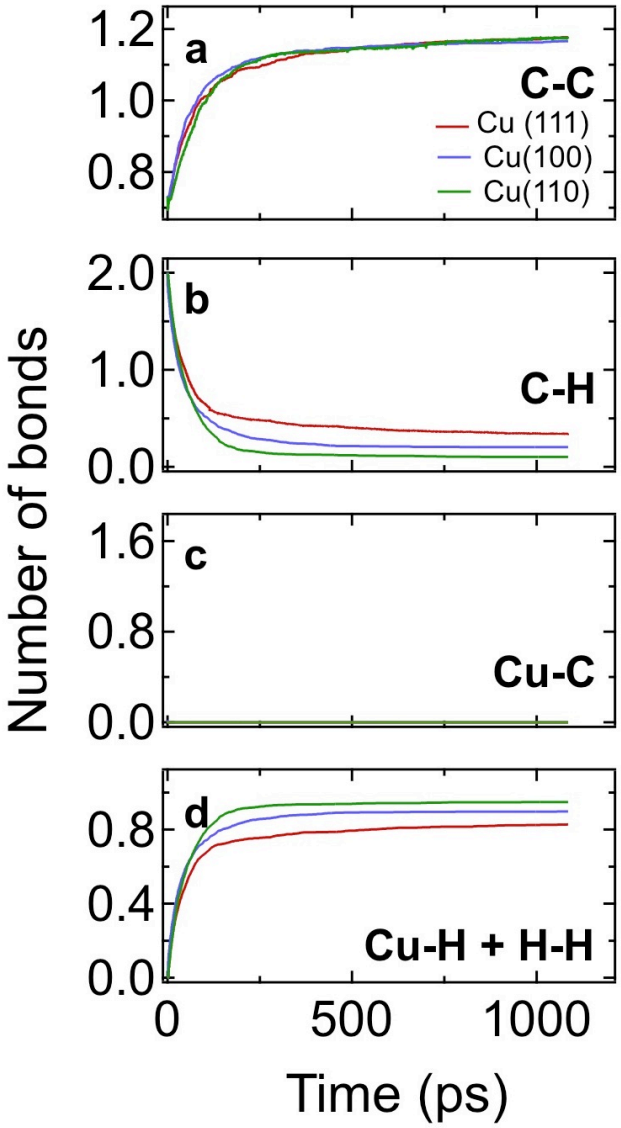


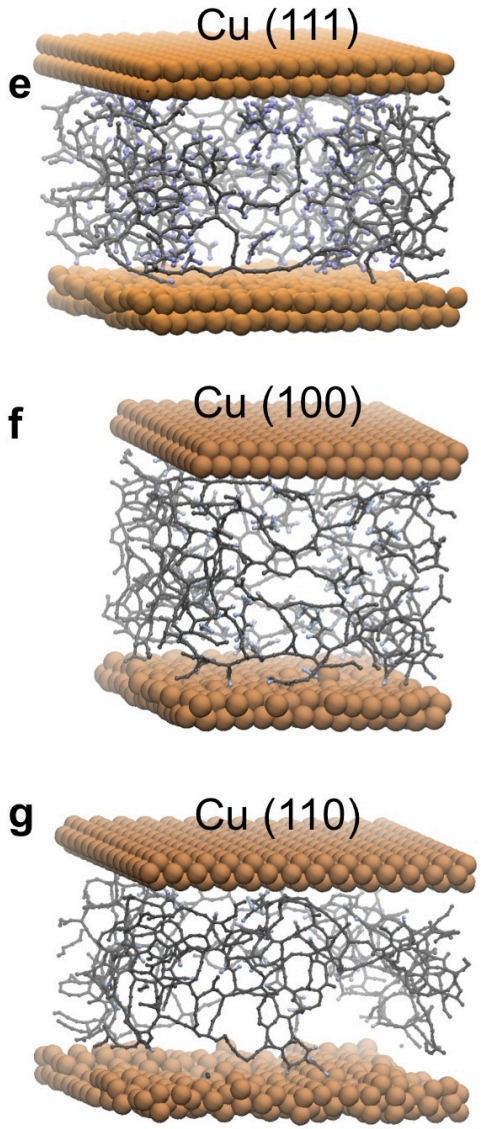


Experimental characterizations confirm that these tribo-films possess structure similar to diamond-like carbon with graphitic features.

A.E, G. R, O.E, **B. Narayanan,** G. K, S. S, Nature, *Accepted for publication*, (2016)

Effect of surface orientation

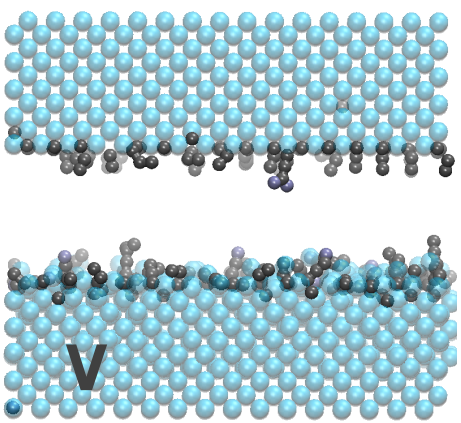


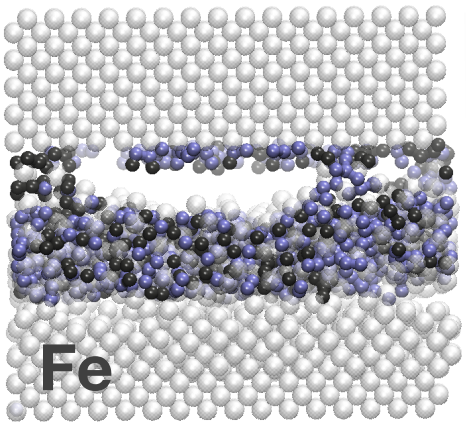


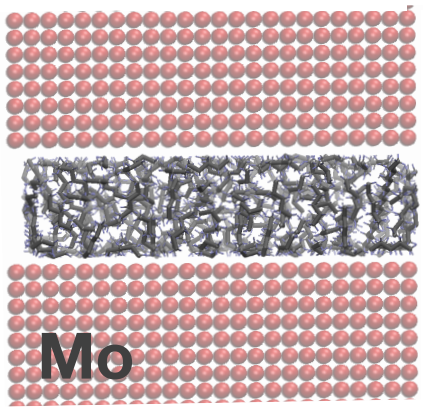
- DLC-like carbon tribofilm forms irrespective of surface orientation
- Dehydrogenation rate increases as surface stability decreases

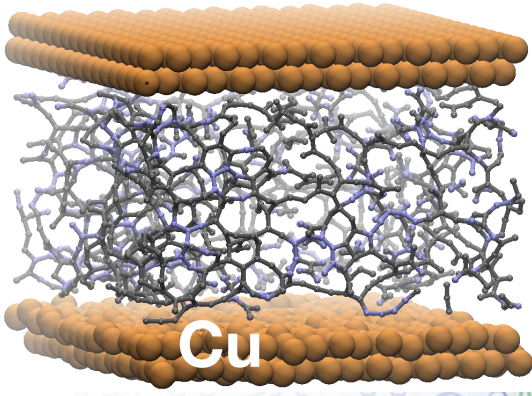
Effect of chemical nature of the surface

Formation of carbon tribofilm









Propensity to form carbide

A.Erdemir, G. Ramirez, O.Eryilmaz, B. Narayanan, G. Kamath, S. Sankaranarayanan, Nature, **536, 67–71** (2016)

Design of low-friction coatings

- MoN_x-Cu coatings reduce friction via formation of solid amorphous DLC-like carbon tribofilms from base lubricating oils

- Atomistic simulations (AIMD + ReaxFF-MD) show that Cu catalyze breakdown of PAO oils
  - Dehydrogenation
  - Chain scission
  - Polymerization of C atoms into an amorphous, hydrogenated DLC-like solid tribo-film

- The tribocatalyst employed should have a low propensity to form carbides.

Force-Field Development from the First Principles: Bridging the electronic and atomistic length-scales

S.K.R.S. Sankaranayanan
Maria Chan
Stephen Gray
Mike Davis
Benoit Roux

Molecular Dynamics

Potential Energy

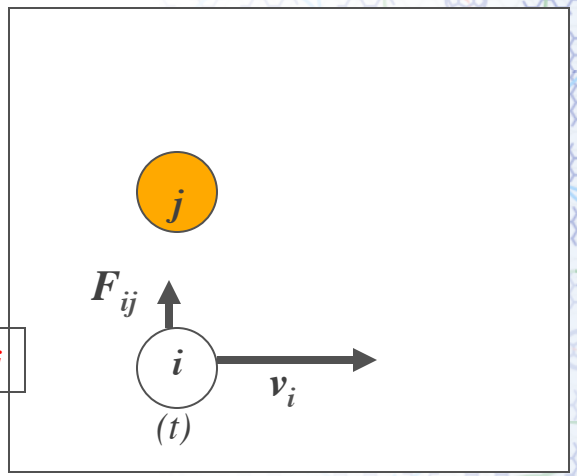
$$\sum_j \vec{F}_{ij} = m_i \frac{d\vec{v}_i}{dt} \qquad \vec{F}_{ij} = -\frac{\partial \phi_{ij}}{\partial r_{ij}} \hat{r}_{ij}$$

Force

Mass

Acceleration

Unit vector from i to j



Molecular Dynamics

- **Newton:** $\sum_j \vec{F}_{ij} = m_i \frac{d\vec{v}_i}{dt}$ $\vec{F}_{ij} = -\frac{\partial \phi_{ij}}{\partial r_{ij}} \hat{r}_{ij}$

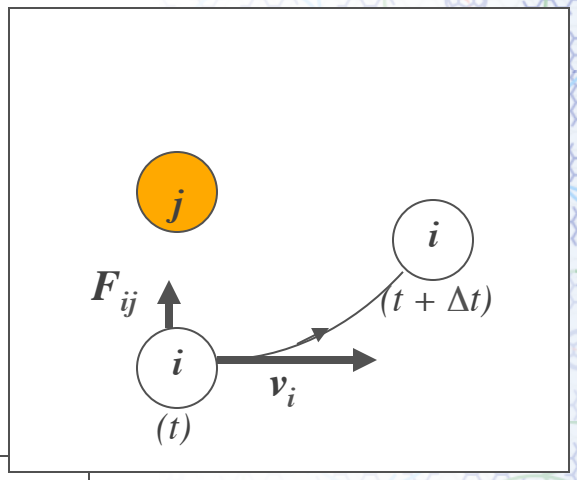
$$\vec{r}_i(t+\Delta t) = \vec{r}_i(t) + (\Delta t)\vec{v}_i(t) + \frac{1}{2}(\Delta t)^2 \frac{\vec{F}_i(t)}{m_i}$$

new position

old position

old velocity

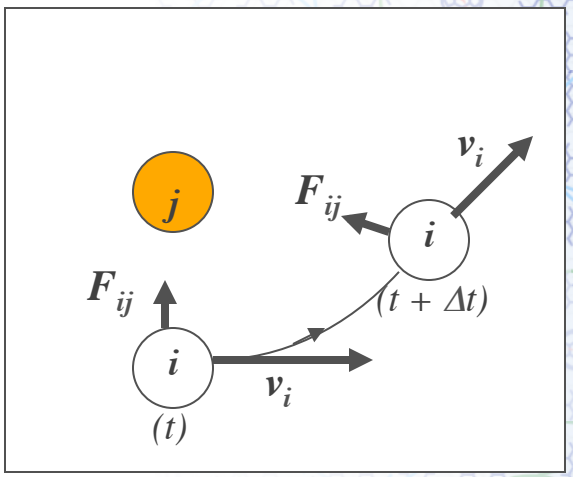
old force



Molecular Dynamics

- Newton: $\sum_j \vec{F}_{ij} = m_i \frac{d\vec{v}_i}{dt}$ $\vec{F}_{ij} = -\frac{\partial \phi_{ij}}{\partial r_{ij}} \hat{r}_{ij}$
- Updated Position:

$$\vec{r}_i(t+\Delta t) = \vec{r}_i(t) + (\Delta t)\vec{v}_i(t) + \frac{1}{2}(\Delta t)^2 \frac{\vec{F}_i(t)}{m_i}$$



$$\vec{v}_i\left(t+\frac{\Delta t}{2}\right) = \vec{v}_i(t) + \frac{1}{2}(\Delta t)\frac{\vec{F}_i(t)}{m_i}$$

first half of time step

$$\vec{v}_i(t+\Delta t) = \vec{v}_i\left(t+\frac{\Delta t}{2}\right) + \frac{1}{2}(\Delta t)\frac{F_i(t+\Delta t)}{m_i}$$

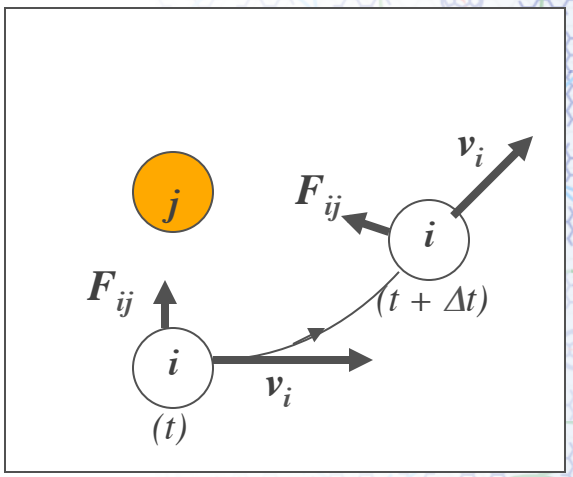
second half of time step

Verlet Algorithm

Molecular Dynamics

- Newton: $\sum_j \vec{F}_{ij} = m_i \frac{d\vec{v}_i}{dt}$ $\vec{F}_{ij} = -\frac{\partial \phi_{ij}}{\partial r_{ij}} \hat{r}_{ij}$
- Updated Position:

$$\vec{r}_i(t+\Delta t) = \vec{r}_i(t) + (\Delta t)\vec{v}_i(t) + \frac{1}{2}(\Delta t)^2 \frac{\vec{F}_i(t)}{m_i}$$



- Updated Velocity:

$$\vec{v}_i\left(t+\frac{\Delta t}{2}\right) = \vec{v}_i(t) + \frac{1}{2}(\Delta t)\frac{\vec{F}_i(t)}{m_i}$$

$$\vec{v}_i(t+\Delta t) = \vec{v}_i\left(t+\frac{\Delta t}{2}\right) + \frac{1}{2}(\Delta t)\frac{F_i(t+\Delta t)}{m_i}$$

$$\sum_{i=1}^{N} \frac{1}{2} m v_i^2 = \frac{3}{2} N k_B T \Rightarrow T = \frac{1}{3Nk_B} \sum_{i=1}^{N} m_i v_i^2$$

system kinetic energy

equipartition theorem

Force Field or Inter-atomic Potential

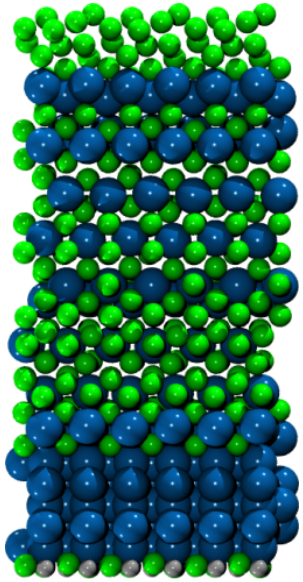
- The heart of any molecular dynamics scheme is the force model used to analytically describe the atomistic interactions
- Regardless of the merits of the other algorithms in the simulation code (integrators, pressure and temperature controls etc.), whether or not your simulation produces realistic results depends ultimately on the force model
- Force models are also the computationally most intensive parts of a molecular dynamics simulation code, taking up to 95% of the total simulation time

Force models / potential functions / force fields / ...

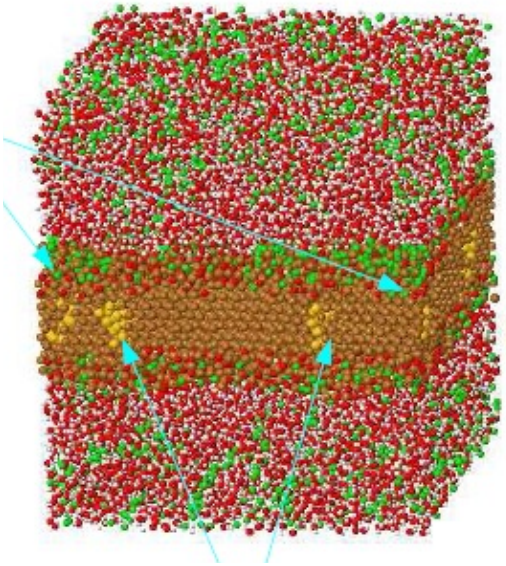
- **Empirical form or based on QM approximations**
- **Parametrized with *ab-initio* or experimental data**
- **Can be reactive (changes in chemical bonds) and polarizable**

Bridging the electronic and atomistic length scales

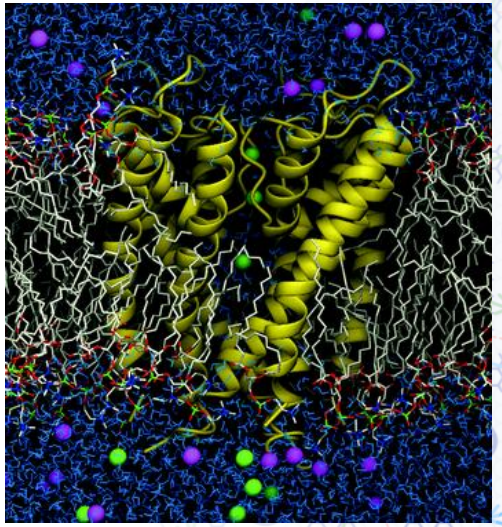
The broad ranges of time and spatial scales relevant to reactive interfaces and processes of interest cannot be met without a new generation of highly accurate and robust, yet computationally efficient, potentials.



energy
(battery)

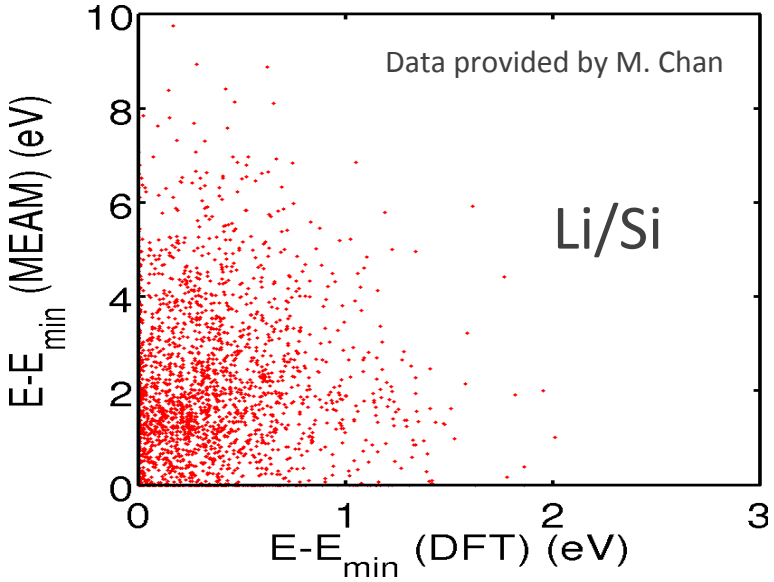


materials
(corrosion)

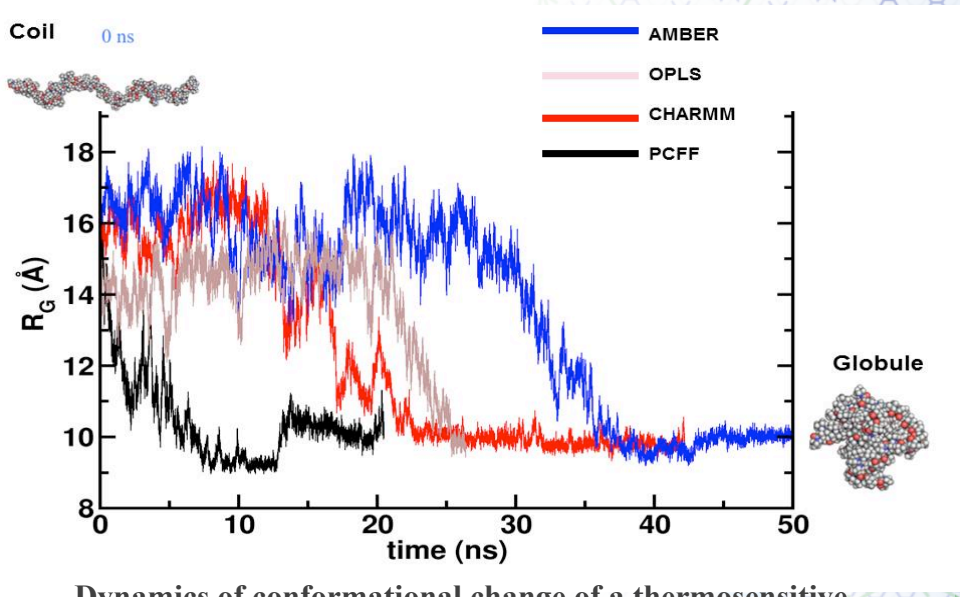


biology
(ion channels)

Existing Force Fields:



Energies relative to the minimum at each composition for the modified embedded atom potential (MEAM) vs. DFT. Note that the MEAM errors are very large.



Dynamics of conformational change of a thermosensitive polymer predicted by four popular force fields is seen to be widely different.

Limitations

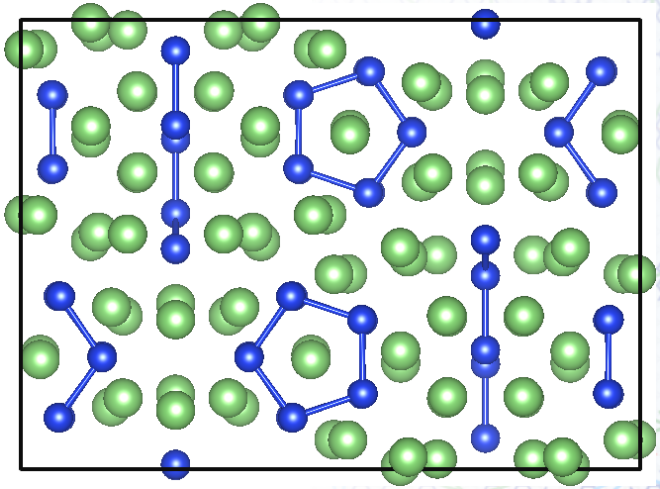
- Pre-defined function forms limit the flexibility of the potential model
- Forces are not included in the fitting
- Lack of adequate training datasets (overemphasis on equilibrium structures)
- Use of least square fitting (prone to difficulty of convergence and overfitting)
- Lack of quantitative cross-validation using data not used in fitting procedure

Goal

The flexibility and accuracy **of electronic structure calculations with the *speed* of classical potentials**

152 atoms, 512 electrons:
0.2 core-second classical (Buckingham)
2800 core-second DFT (PBE)

4 orders of magnitude in compute time
+ scaling advantage



Approach

Data-driven model selection:

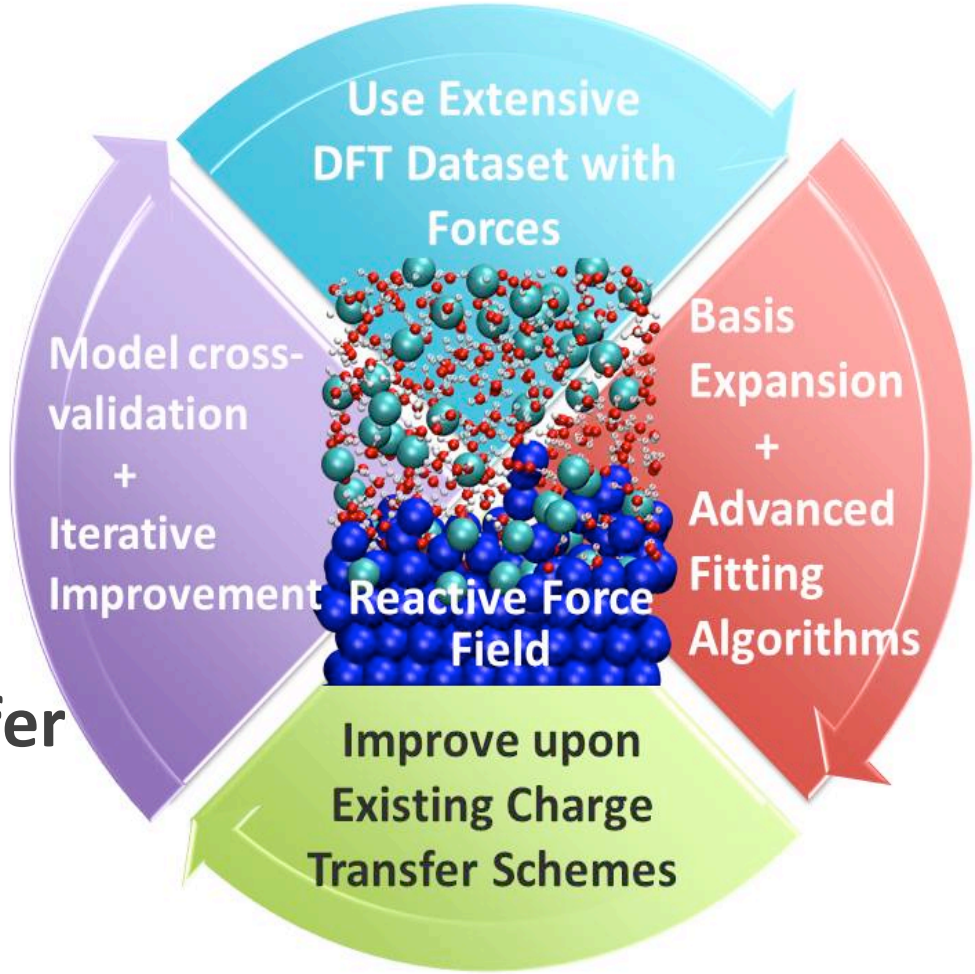
- linear fitting,
- beyond least square
- e.g. Sparse regression (compressive sensing)

Uncertainty quantification:

- sensitivity analysis
- sampling effects
- cross validation

Charge polarization/transfer dynamics:

- multipole moments
- charge equilibration
- boot-strap species-by-species fitting



Force Field Development from first princples

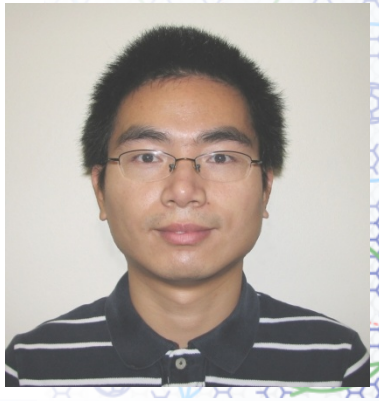
Test Systems

- Dimensionality effects in Gold Clusters
- Reactive force field for IrO_x
- Organic Molecules in Solvents

Badri Narayanan

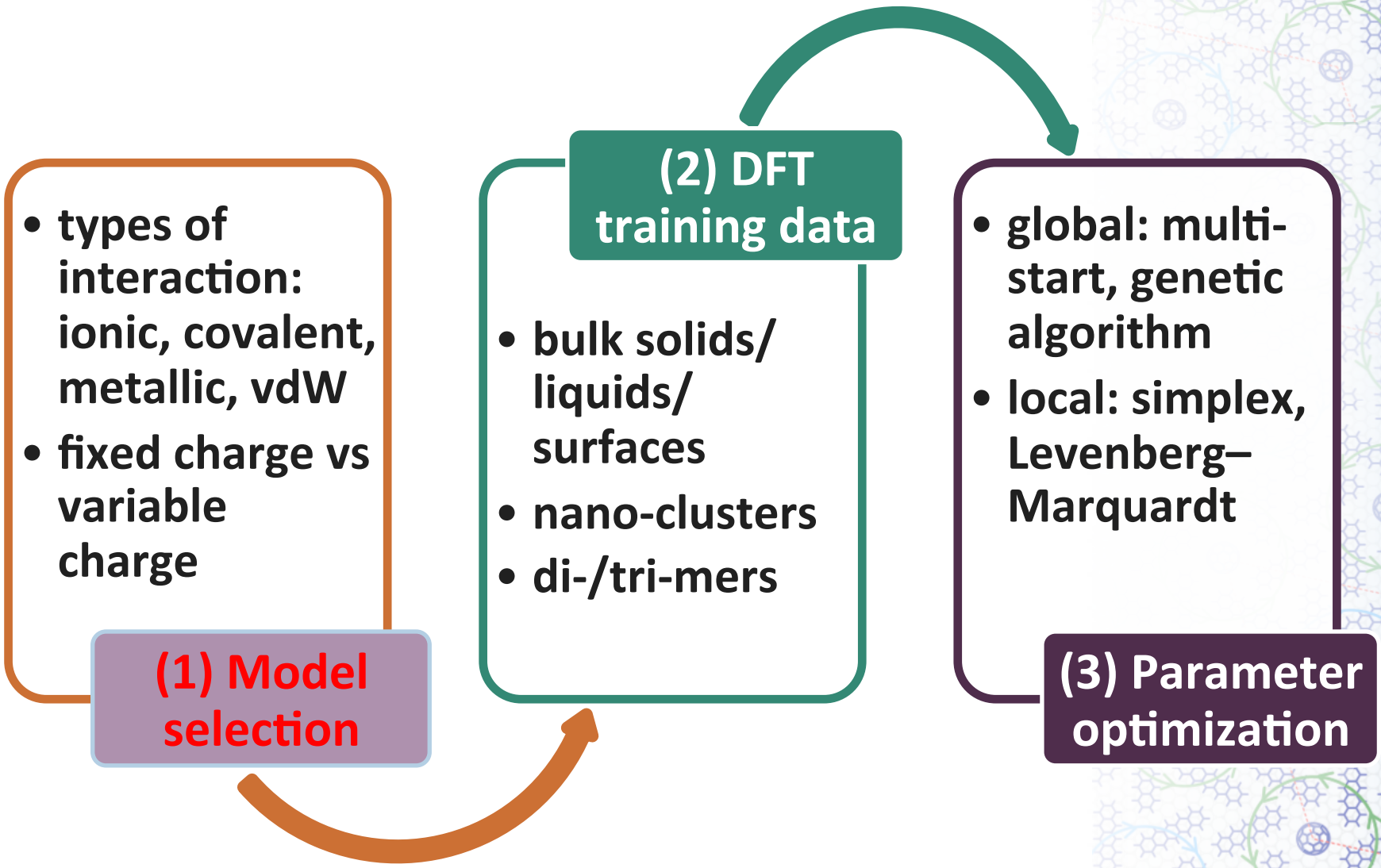
Alper Kinaci

Fatih Sen



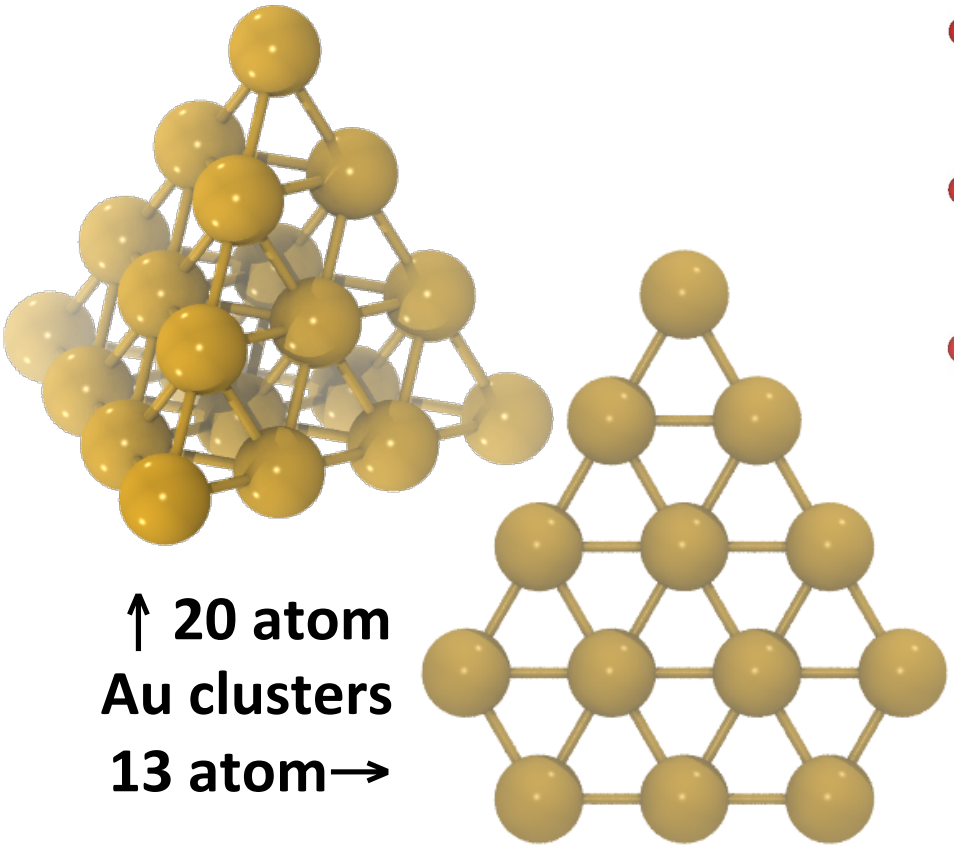
Lei Huang

Force Field Fitting Framework

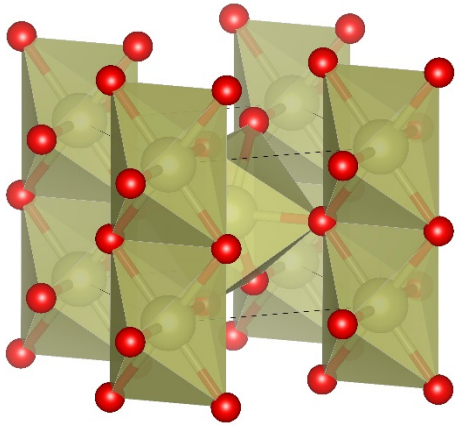


Test systems

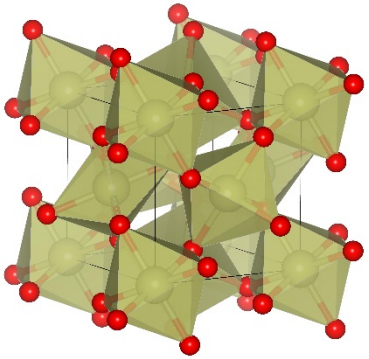
- Au & IrO_2: relevant for catalysis/photocatalysis



↑ 20 atom
Au clusters
13 atom→

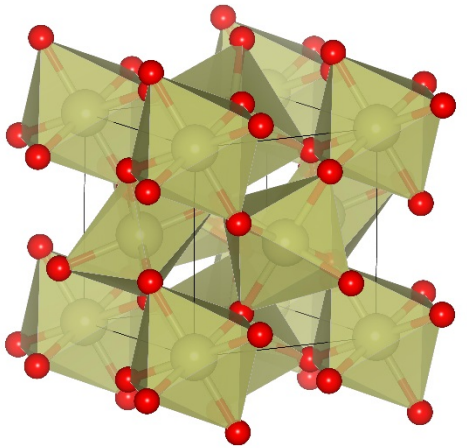


Rutile IrO_2



Pyrite IrO_2

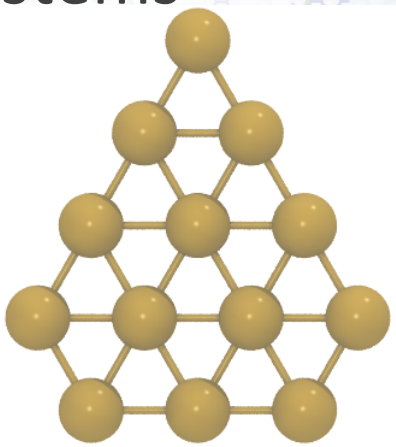
(1) Model selection $E=f(\{\vec{r}_i\})$



- IrO_2: no existing force fields
  - ionic system, Morse + electrostatics

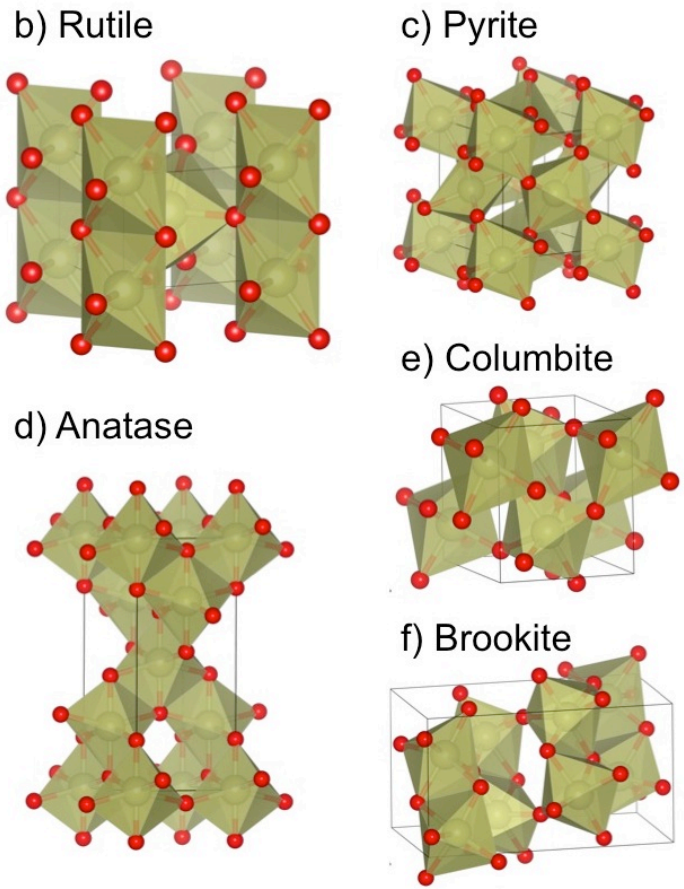
$$E(r_{ij}) = D_e\left([1-\exp(-a(r_{ij}-r_0))]^2 - 1\right) + \frac{q_i q_j}{r}$$

  - Variable charge scheme using QEq
- Au: several existing force fields for bulk systems – Sutton-Chen, Gupta, ReaxFF, Tersoff
  - do they work for nanoclusters?

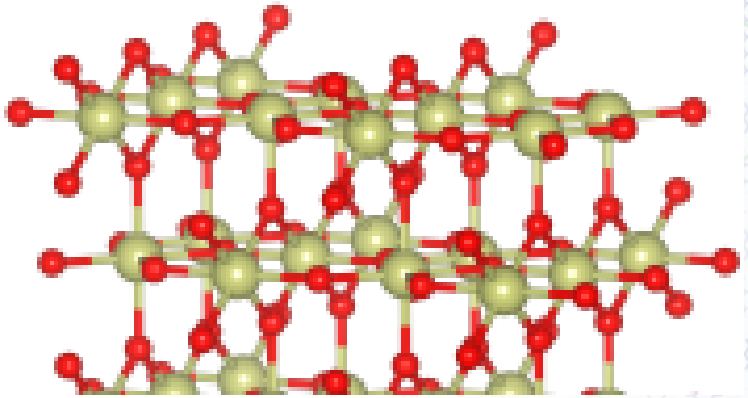


(2) DFT training data - bulk

- lattice parameters, formation energies, elastic constants, surface energies, internal coordinates



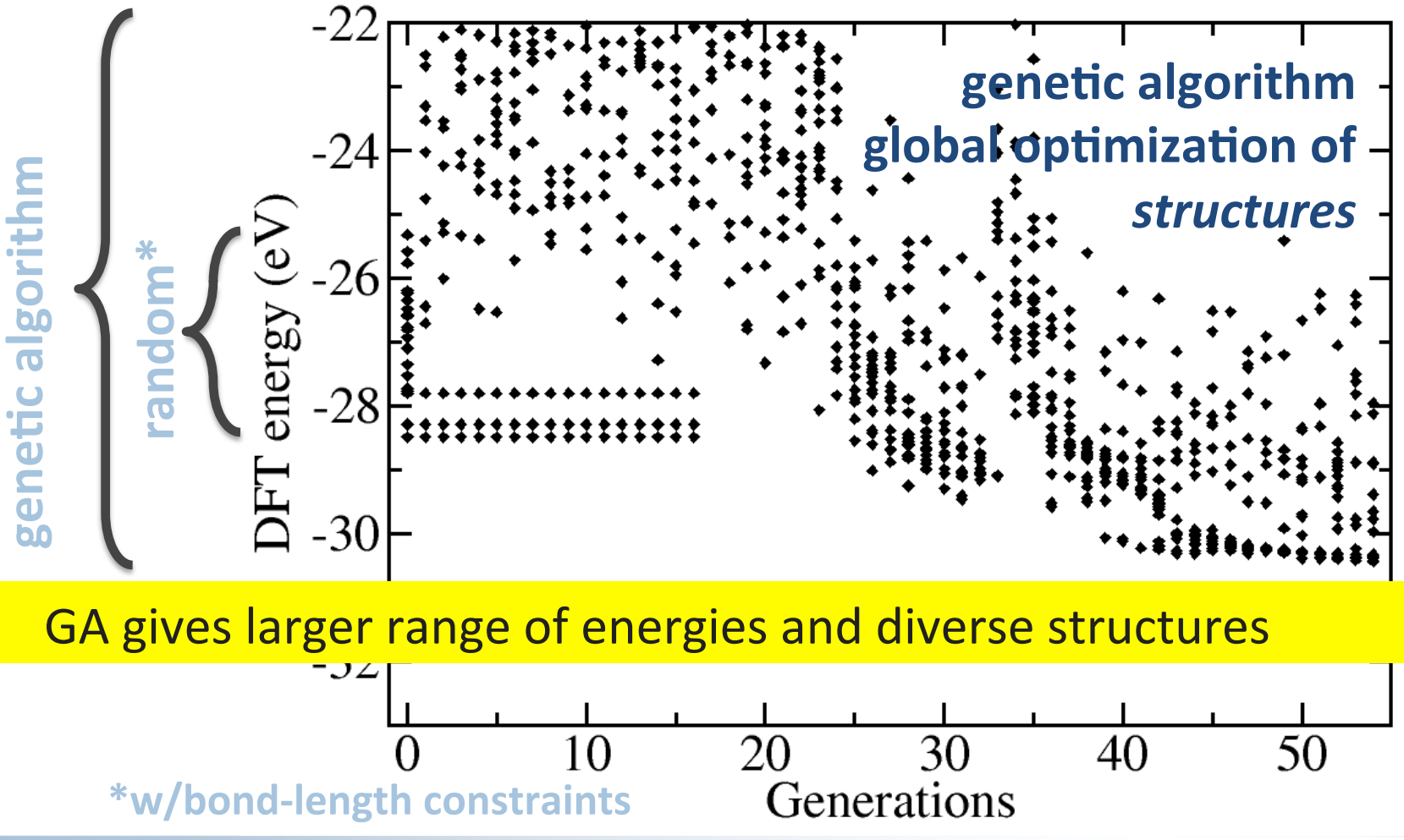
Bulk phases



Surfaces

(2) DFT training data - nano

- dimers and trimers: straightforward
- larger clusters?

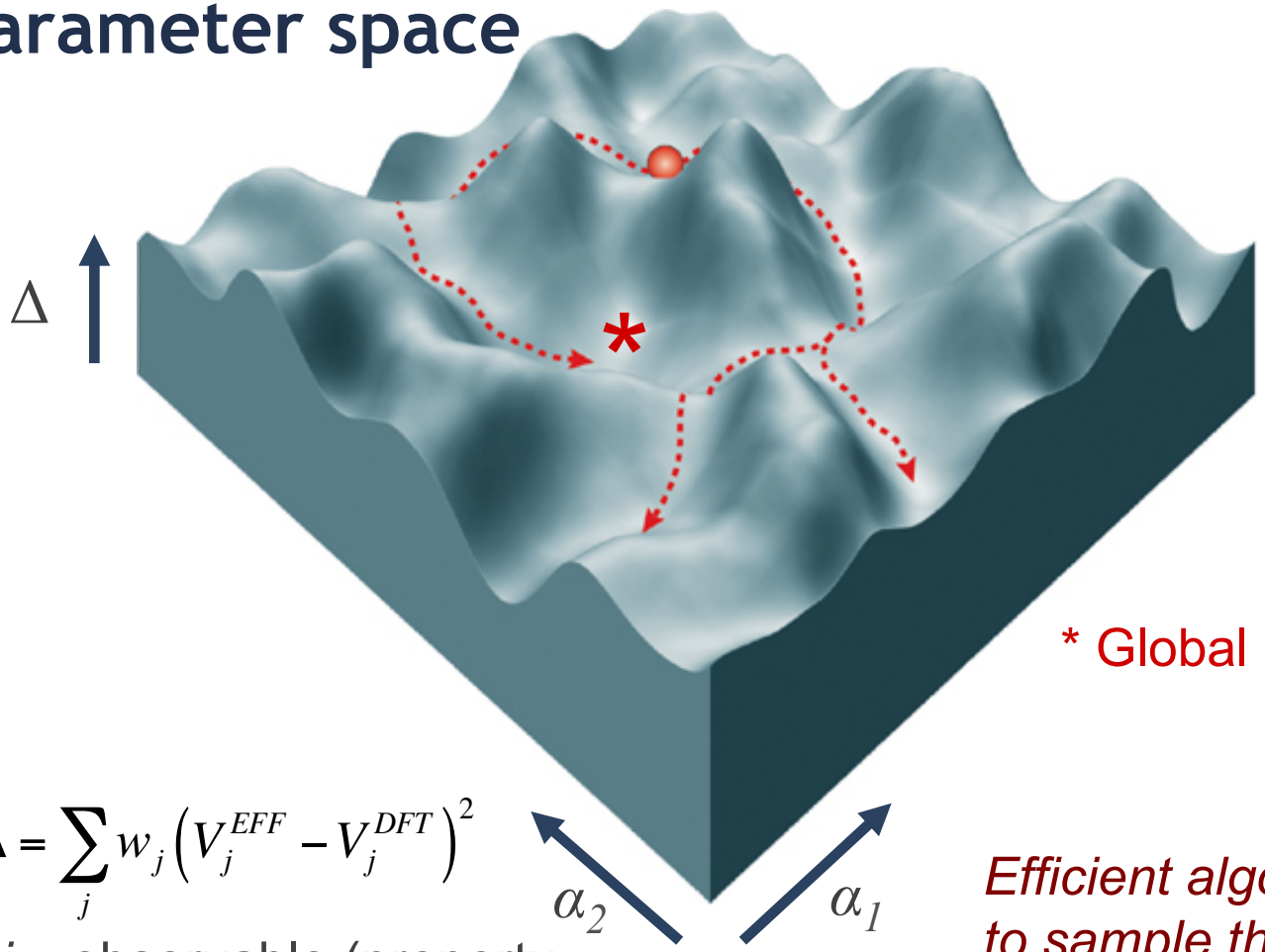


GA gives larger range of energies and diverse structures

*w/bond-length constraints

(3) Genetic algorithm (GA) Optimization

Objective Function landscape in multidimensional parameter space



* Global minimum

$$\Delta = \sum_j w_j \left(V_j^{EFF} - V_j^{DFT} \right)^2$$

j – observable (property used for training)

Efficient algorithms are necessary to sample this complex landscape to find the lowest point (the best parameter set)

(3) Genetic algorithm (GA) Optimization

DFT training set (energies, structures)

Random population of possible parameter sets

Objective function values

$$\sum_i w_i \left(X_{DFT} - X_{Pot} \right)^2$$

for parameters

Select *N* fittest solutions

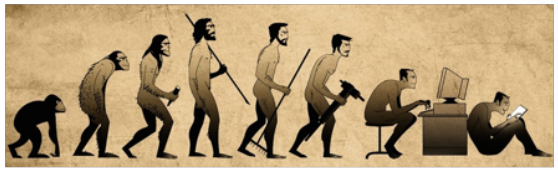
Perform genetic operations

Convergence reached?

No

Yes

Optimized parameter set



www.commonsenseevaluation.com

(3) GA + Local Optimization

DFT training set (energies, structures)

Random population of possible parameter sets

Objective function values

$$\sum_i w_i \left(X_{DFT} - X_{Pot} \right)^2$$

Local minimization

Select *N* fittest solutions

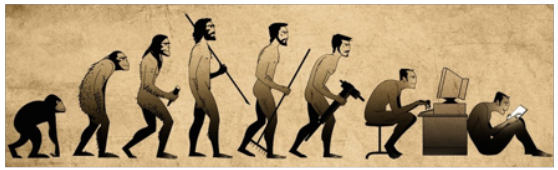
Perform genetic operations

Convergence reached?

No

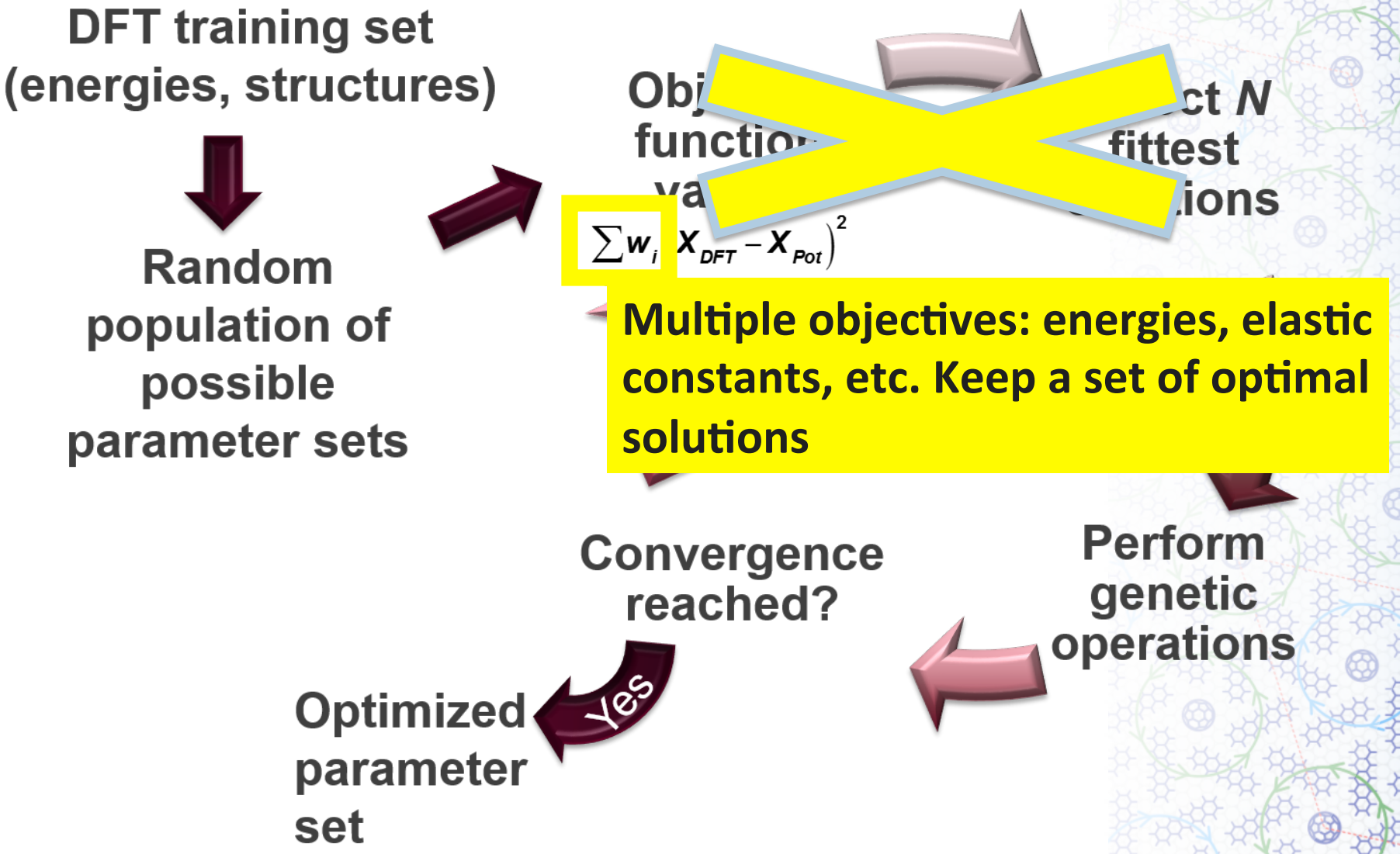
Yes

Optimized parameter set



www.commonsenseevaluation.com

(3) Multi-objective GA Optimization



Force Field Fitting Framework

- types of interaction: ionic, covalent, metallic, vdW
- fixed charge vs variable charge

(2) DFT training data

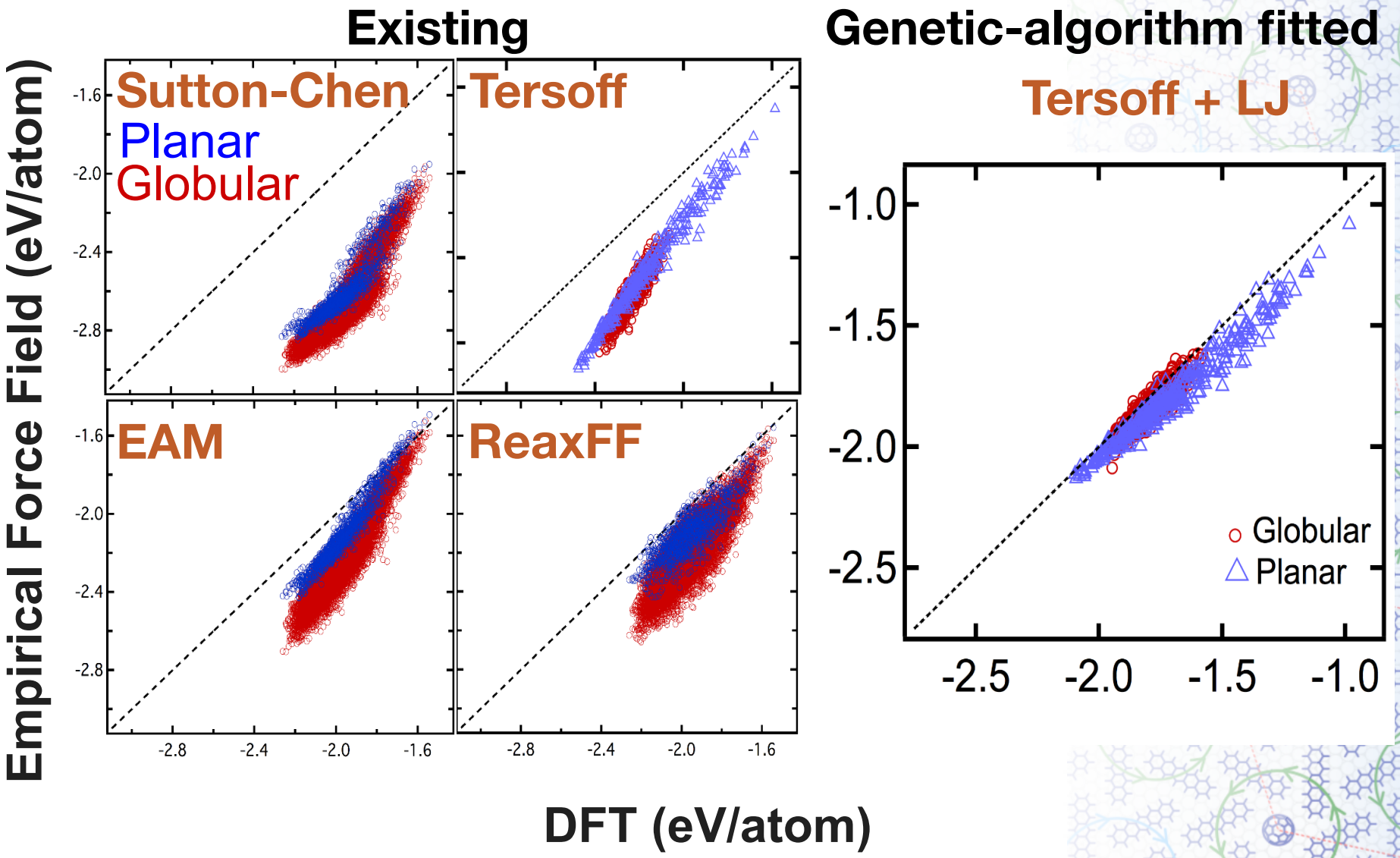
- bulk solids/ liquids/ surfaces
- nano-clusters
- di-/tri-mers

- global: multi-start, genetic algorithm
- local: simplex, Levenberg–Marquardt
- cross validation

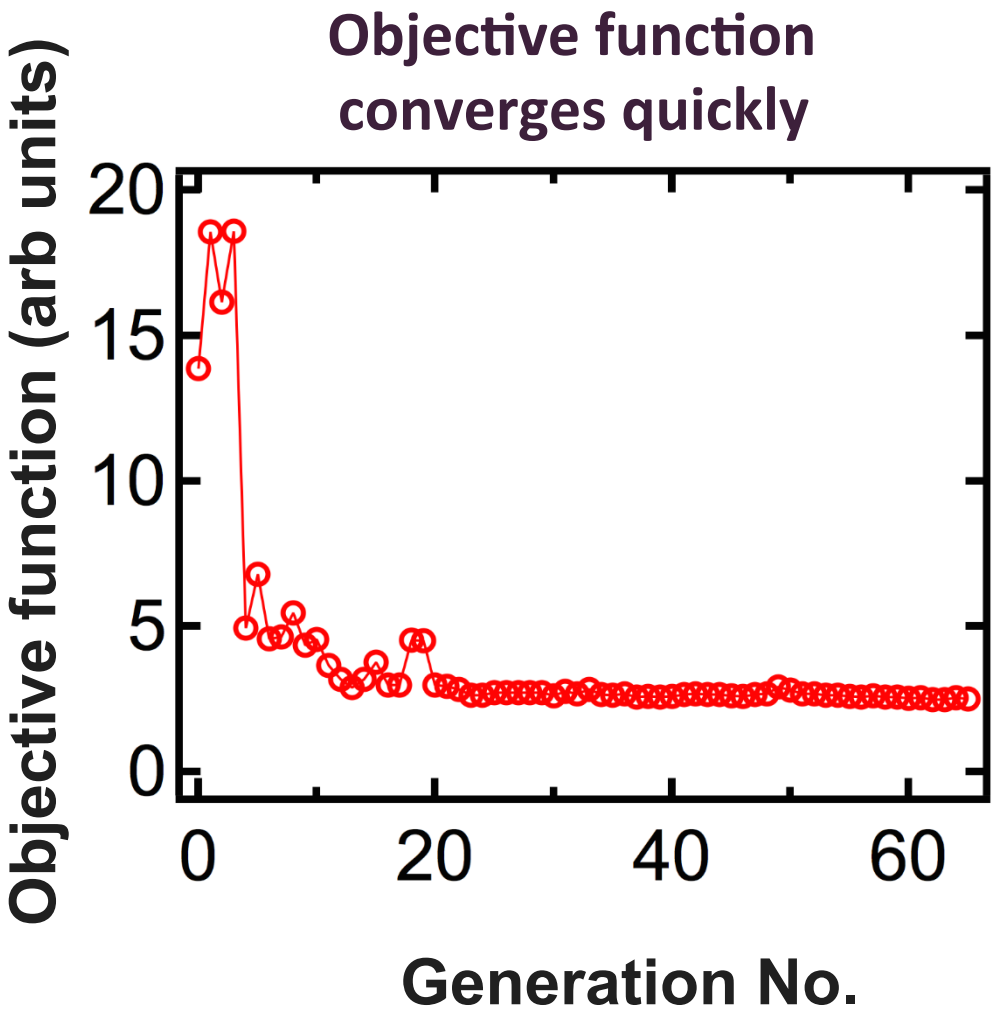
(3) Parameter optimization

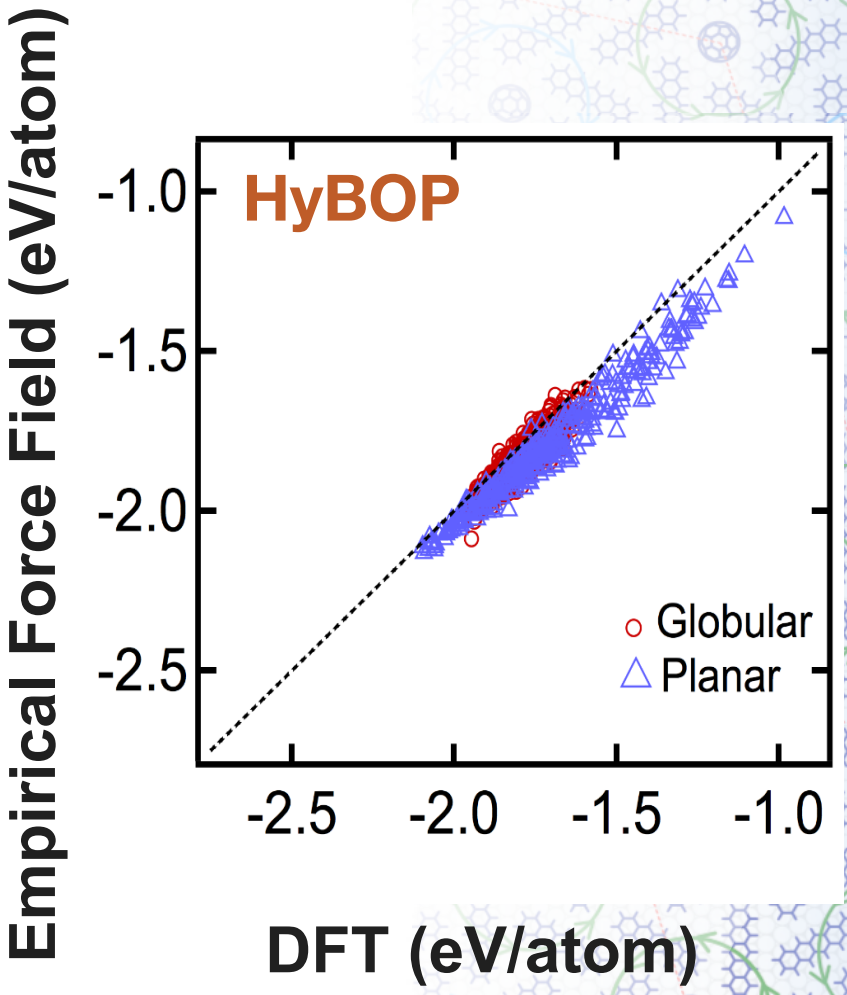
Results →

Au nanoclusters: model selection



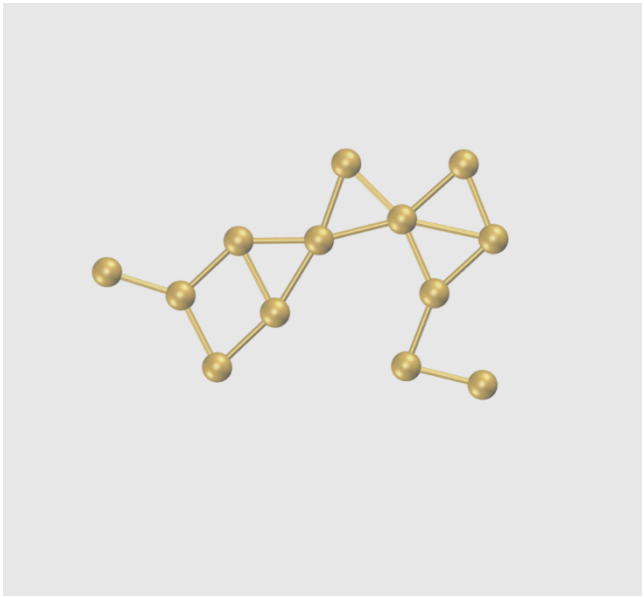
Au nanoclusters: GA+simplex optimization





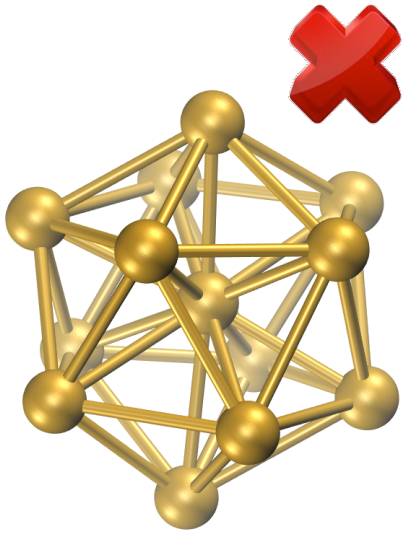
Fitted force field correctly predicts 2D-3D transition for Au

Electronic

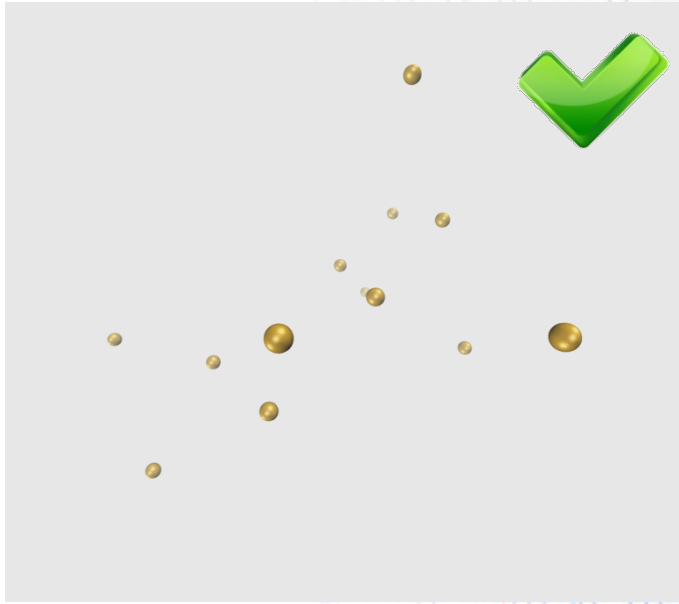


DFT genetic algorithm optimization of Au_{13}

Atomistic



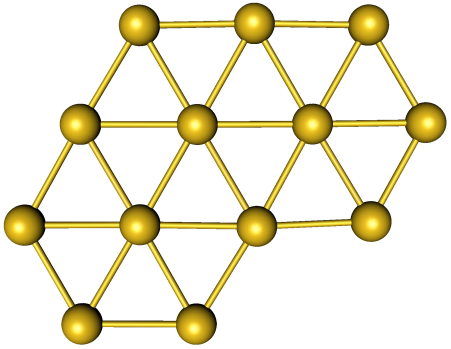
Existing force fields: wrong structure



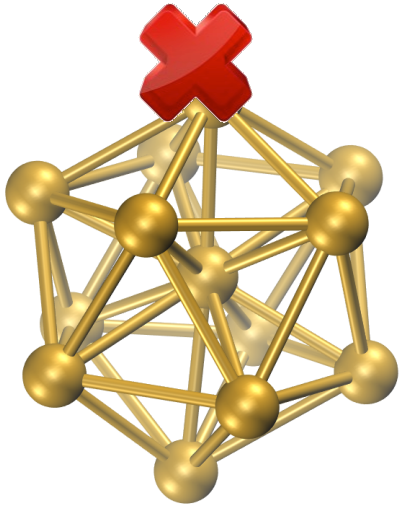
Re-parameterized force field: isolated Au coming together to form correct (DFT) structure

Fitted force field correctly predicts 2D-3D transition for Au

DFT



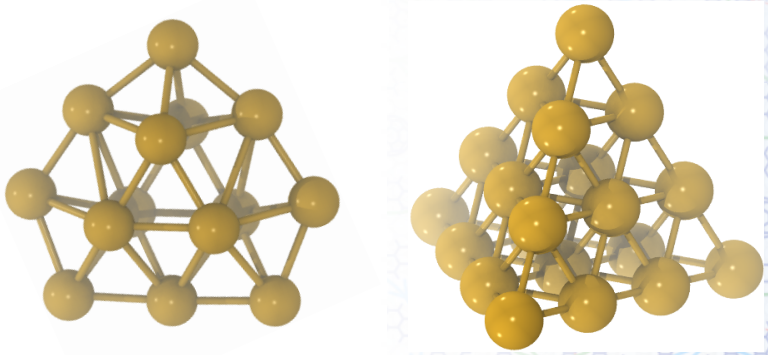
DFT genetic algorithm optimization of Au_{13}



Existing force fields: wrong structure for Au_{13}

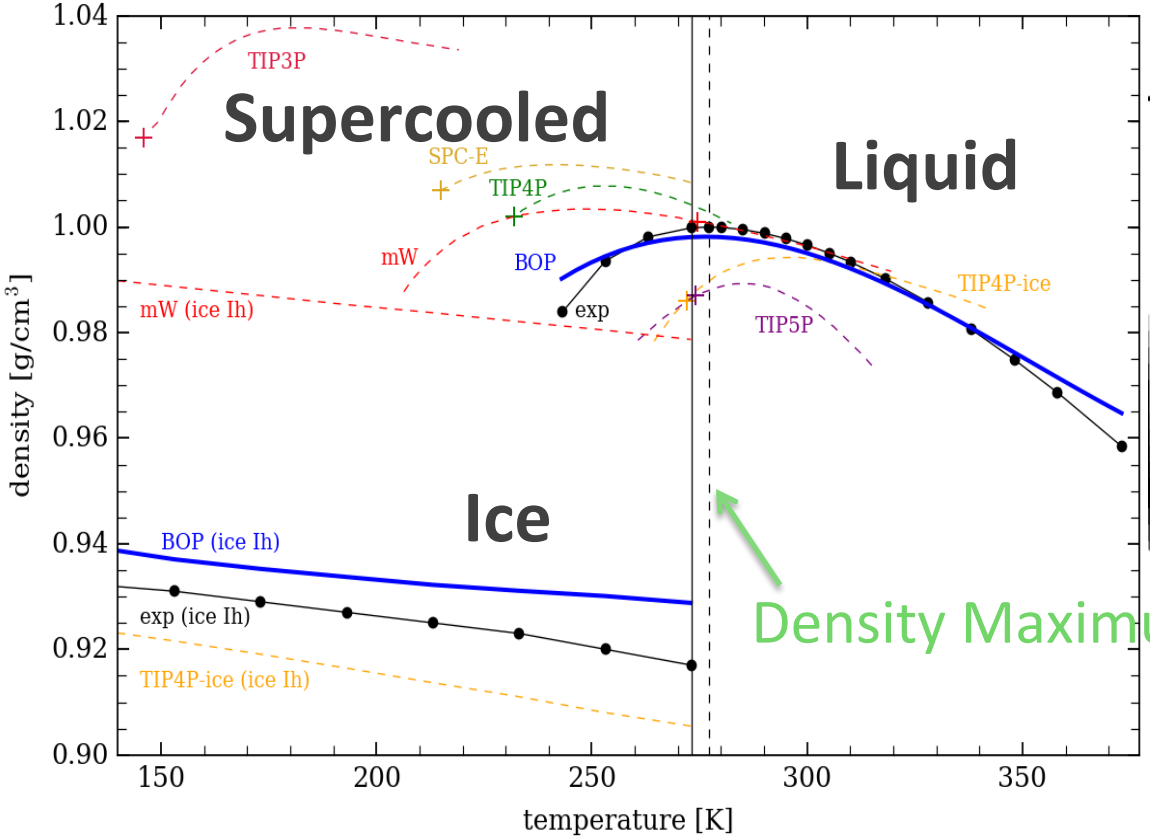
Force fields

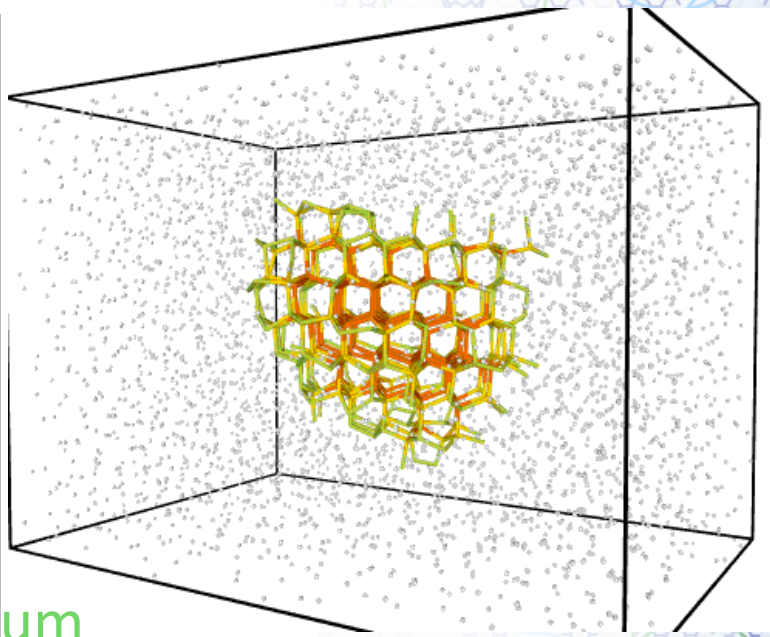
Re-parameterized force field: correct planar structure for Au_{13}



Correct 3d structure for Au_{14} and Au_{20}

Example: machine-learning based force fields and molecular dynamics analysis: an efficient and accurate model for water

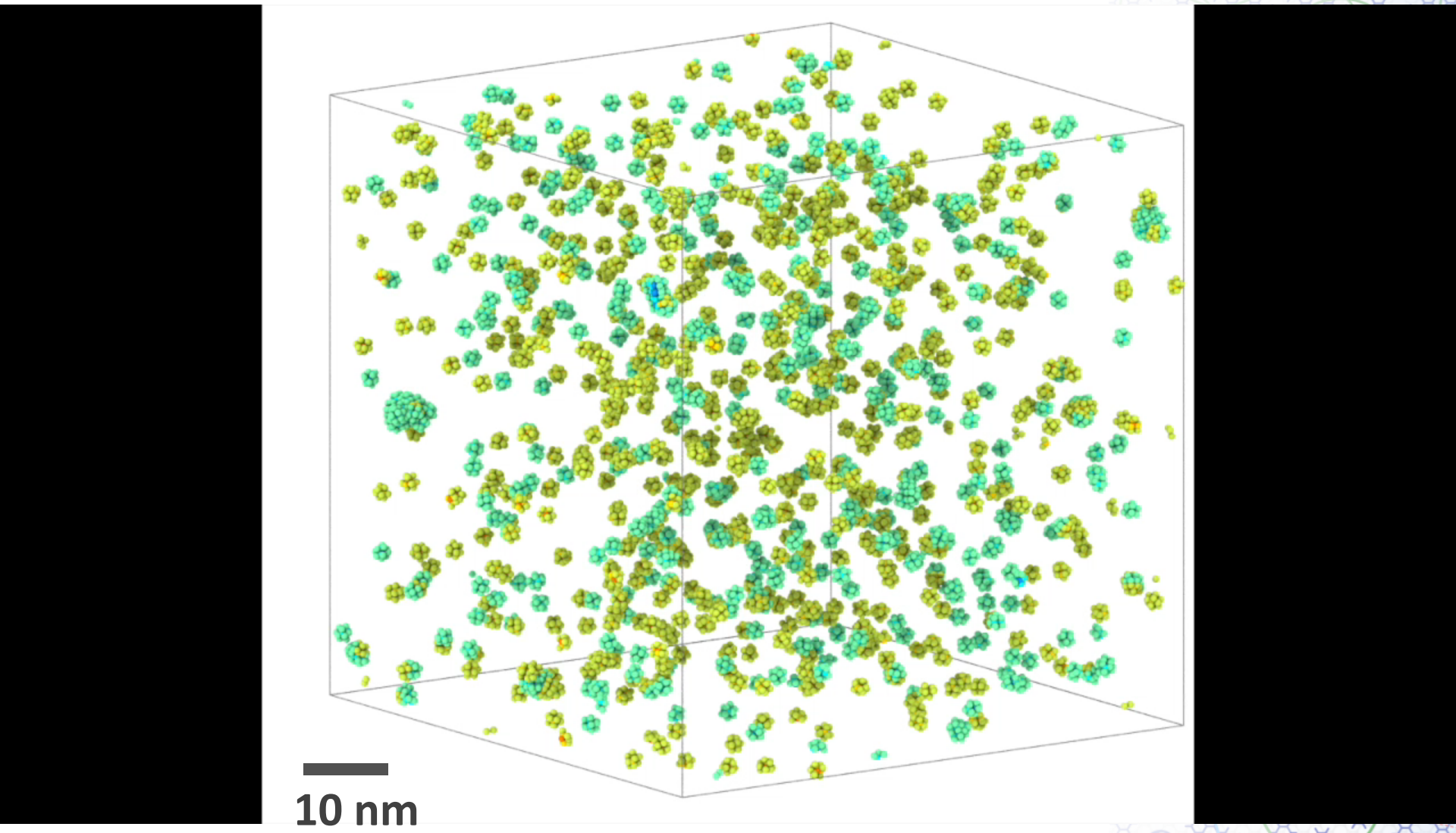




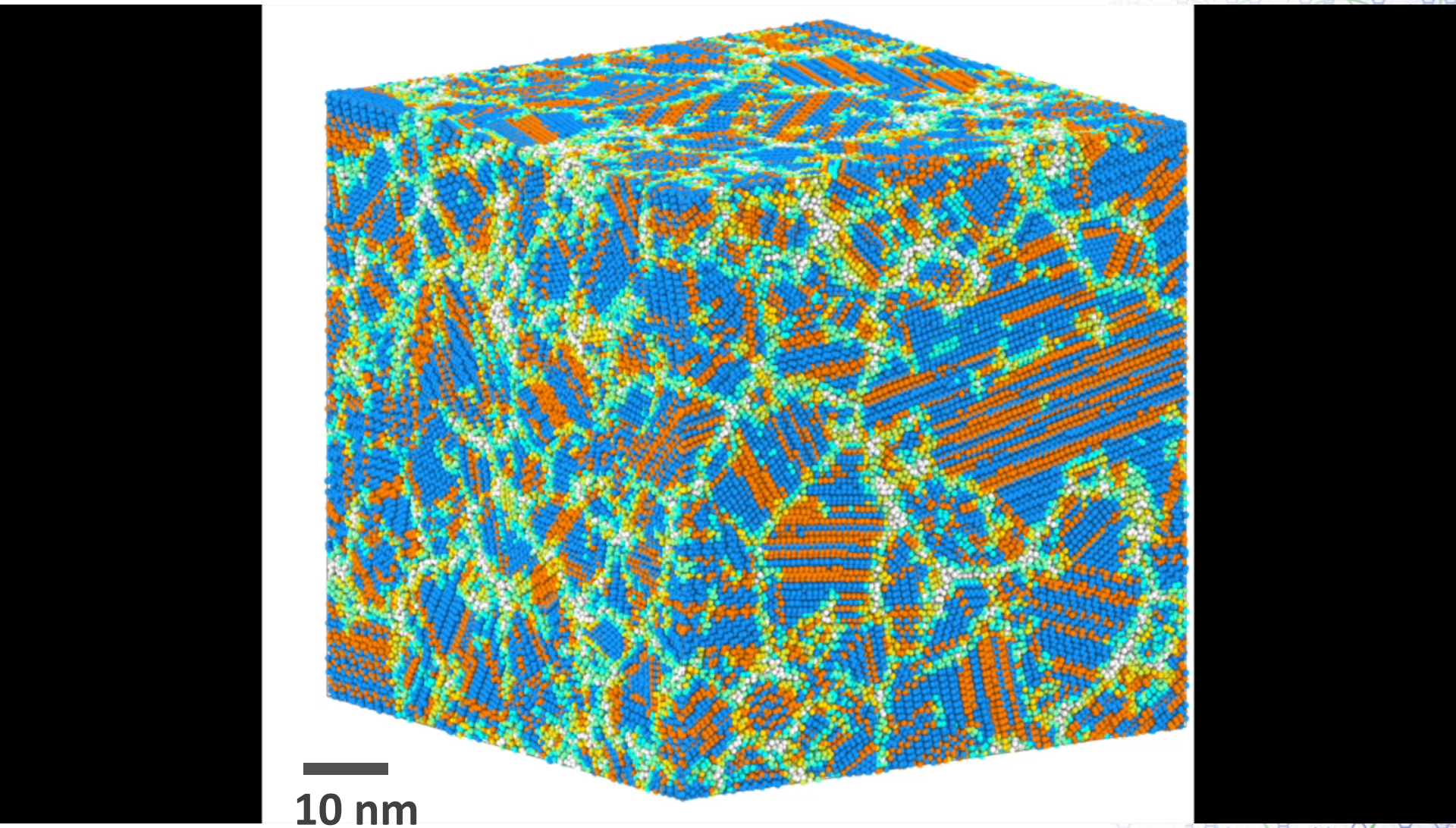
- New force field ("BOP") trained against best atomistic and empirical data describes water density anomaly and ice better than all existing models and is 1000 times faster.

- Large-scale (microseconds, billions of molecules) MD simulations using BOP to discover key aspects of nucleation, etc., which will be enabled by clustering, tracking.

Formation and growth of grains of ice

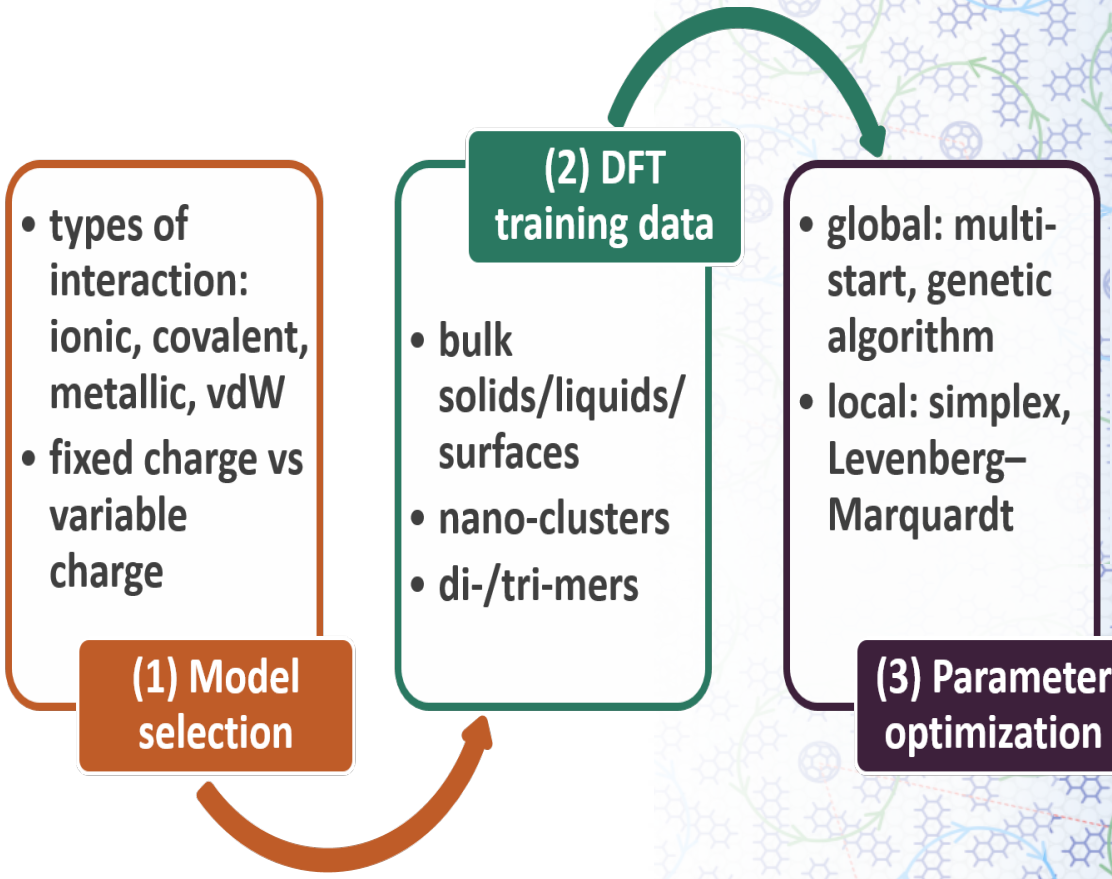


Formation and growth of grains of ice



Summary

- Developed framework to fit force fields from first principles
- Systematic DFT data generation
  - Use genetic algorithm global structural optimization for clusters
- Genetic algorithm approach for parameter fitting gives good results for test cases
  - multi-objective allows control of tradeoffs between competing objectives
  - maintaining appropriate population size important



Our New Approach to Force Field Fitting:

Potential function in terms of a superposition of a linearly-independent basis set of functions

$$E_{\text{total}} = \sum_m \alpha_m \sum_{<ij>} f_m(r_{ij}) + \sum_n \beta_n \sum_{<ijk>} g_n(r_{ijk}) + \ldots$$

E_{total} is the total energy, f_m and g_n are **over-complete sets** of basis functions (e.g. Bessel functions, Hermite polynomials, exponentials, trigonometric functions, or combinations thereof), r_{ij} and r_{ijk} are rotationally-invariant combinations of the atomic coordinates $\mathbf{r}_i$

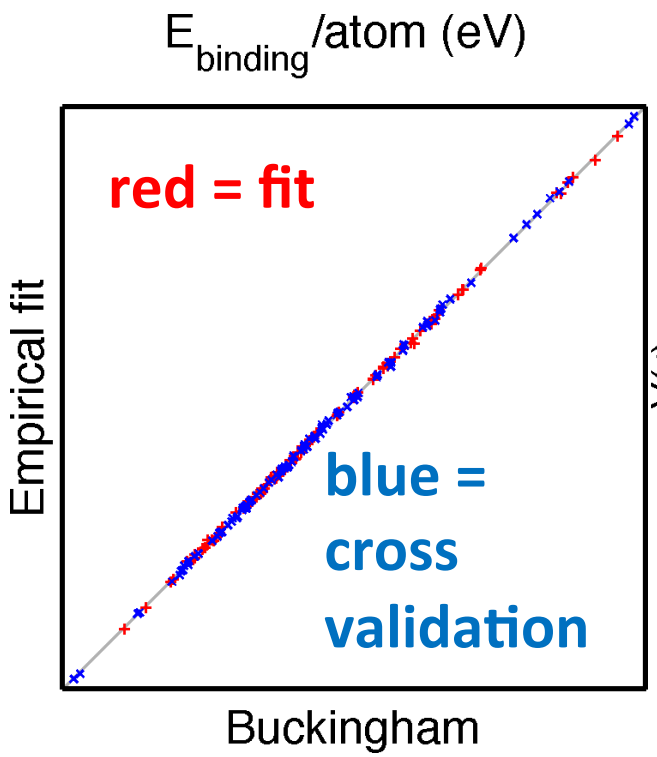
α's and β's are expansion coefficients to be determined by fitting

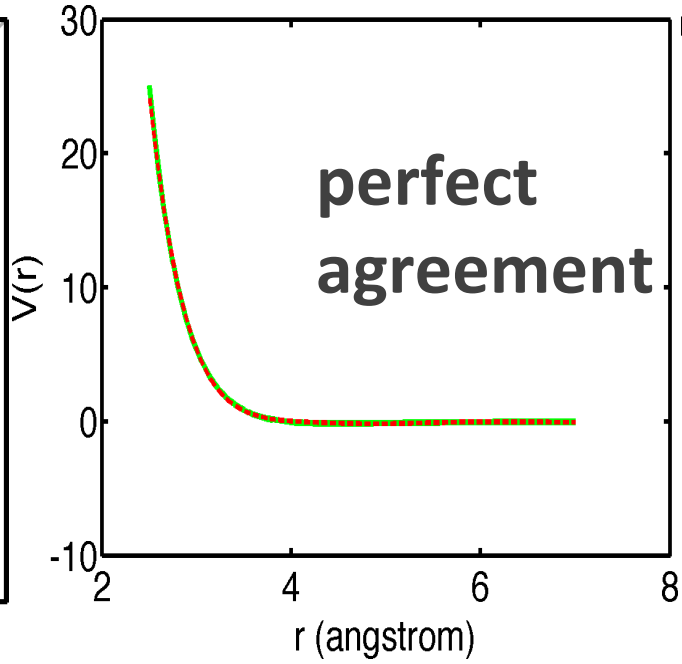
Compressive sensing to perform fitting to the ***large DFT dataset of forces and energies***

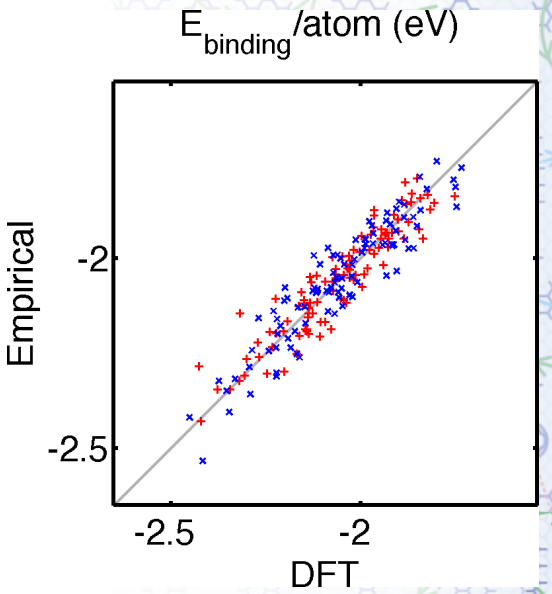
Test Case: nanoclusters

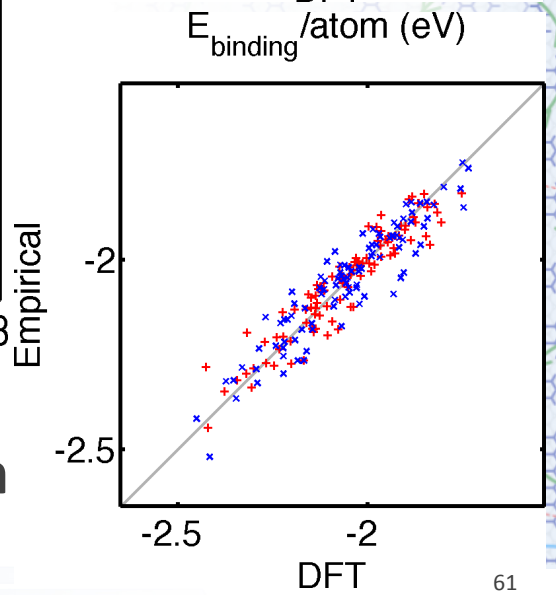
Work in progress

Gold: Fit total energy from Buckingham potential to linear expansion of Bessel fcns, exponentials, Gaussians, cosines, etc









Agnostic data-based approach to model selection

Choice of EFF Functional Form

Input training configurations

VASP input

DFT Material property, X_1

DFT Material property, X_2

DFT Material property, X_n

$X_1, X_2, \ldots X_n$: *Lattice parameters, Cohesive energies, elastic constants, surface energies*

First principles calculated training set using VASP

TRAINING DATA SET

Global optimization of EFF parameters

Population of *N* parameter sets for EFF

EFF predictions using LAMMPS

LAMMPS input

EFF Material property, X_1

EFF Material property, X_2

EFF Material property, X_n

Evaluate errors in prediction for each set $\Delta_1, \Delta_2, \ldots \Delta_N$

Converged ?

Yes

No

GA evolution to obtain next generation of N parameter set

Local or Derivative Free Optimization

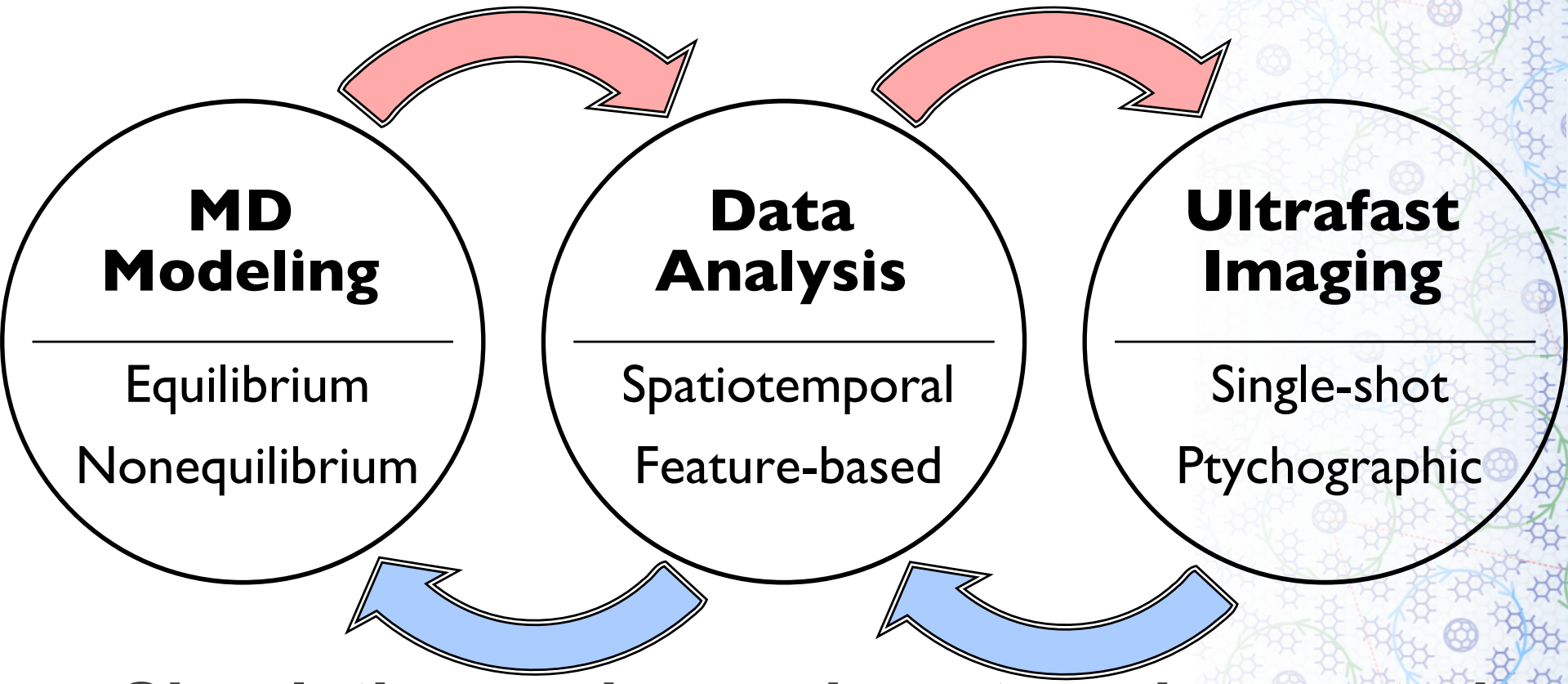
SIMPLEX, Levenberg-Marquardt, POUNDERS, ORBIT etc

Optimized EFF for MD simulations of Tribo-systems and Materials

Modeling, Analysis and Ultrafast Imaging

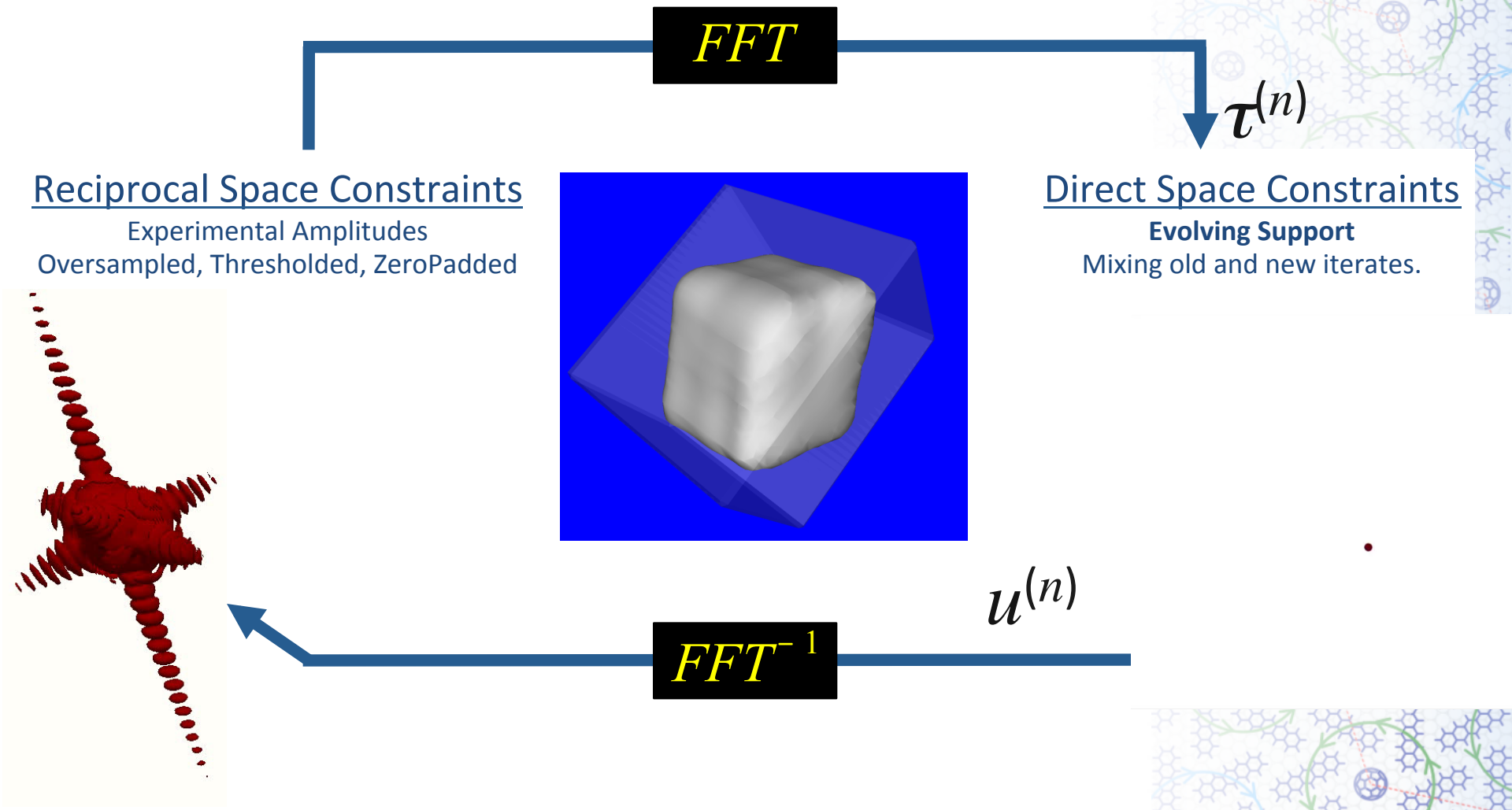
Tom Peterka
S.K.R.S. Sankaranarayanan
Ian McNulty
Ross Harder
Haidan Wen
Todd Munson

Modeling, Analysis and Ultrafast Imaging



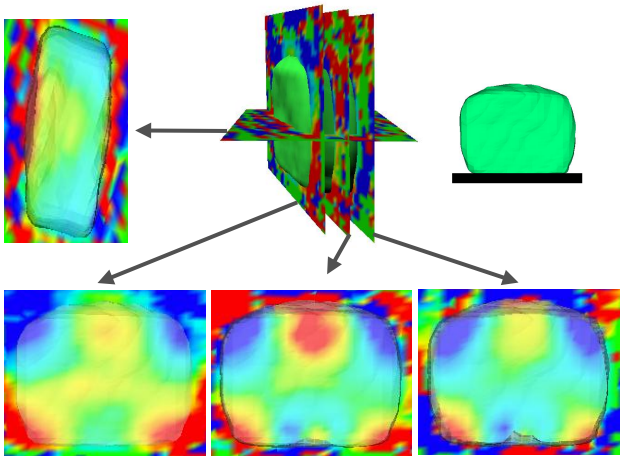
Simulation and experiment are integrated through a common language of data analysis.

Phase Retrieval: Recover real space structure and strains

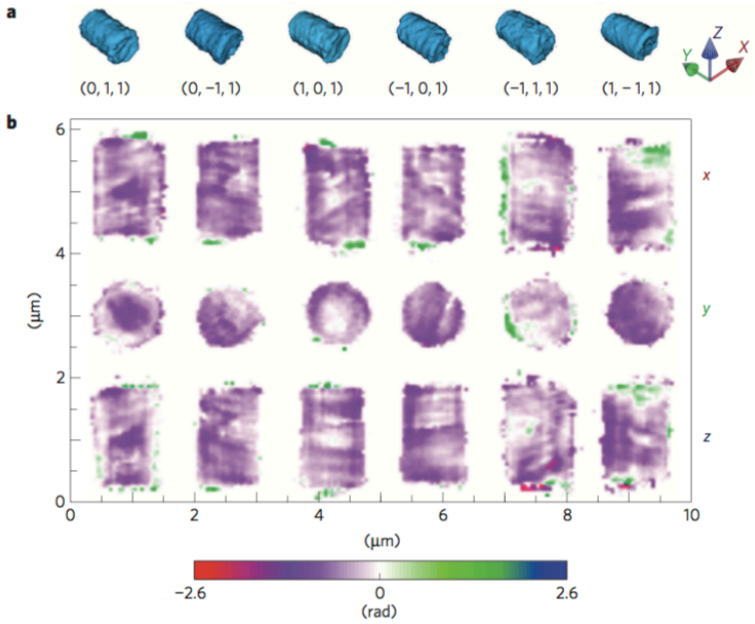


1000's of iterations and multiple random starts required for high fidelity data!

CDI and MD: not so strange bedfellows



Cha, et. al Nat Mater 12 (8) 729–34.



Newton, Leake, Harder, Robinson, Nat. Mat 9 120-124

CDI

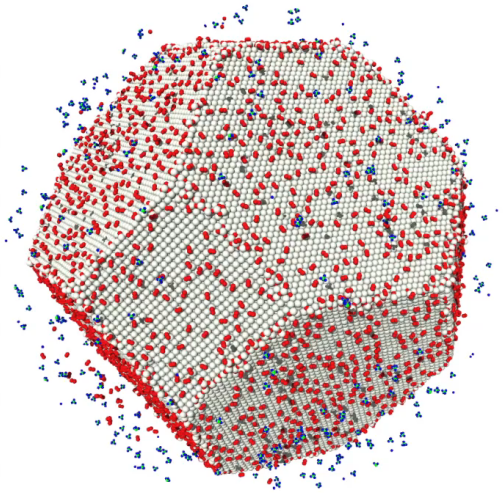
- ~10 nm spatial resolution
  - Lot that happens at smaller length scales
- Can only image crystalline structures.
  - Not chemical reactants and products
  - Not solvent or ions

MD

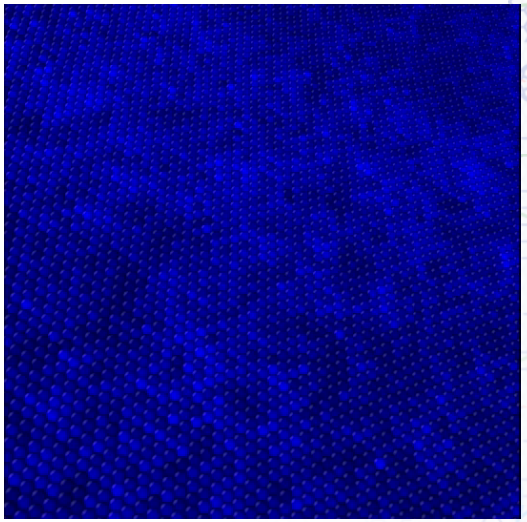
- Full atomistic information
  - Can do up to 100's of nm.
- Can provide a picture of all aspects of the system.
  - Ions, ligands, gas phase

MD is *the ideal technique* to complement CDI

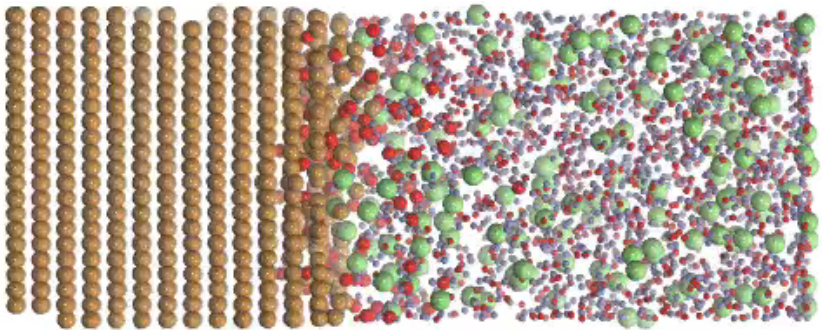
Catalysis



Interface chemistry

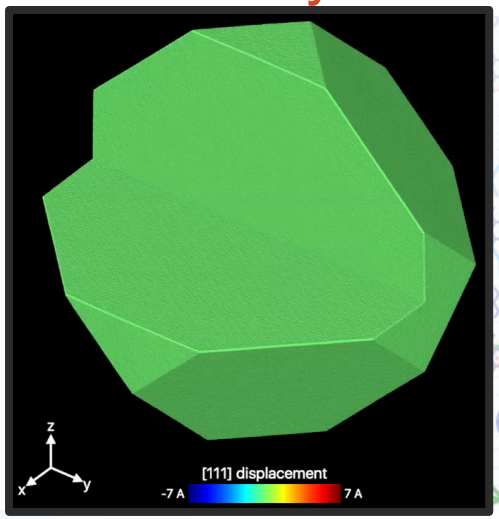


Electrochemistry

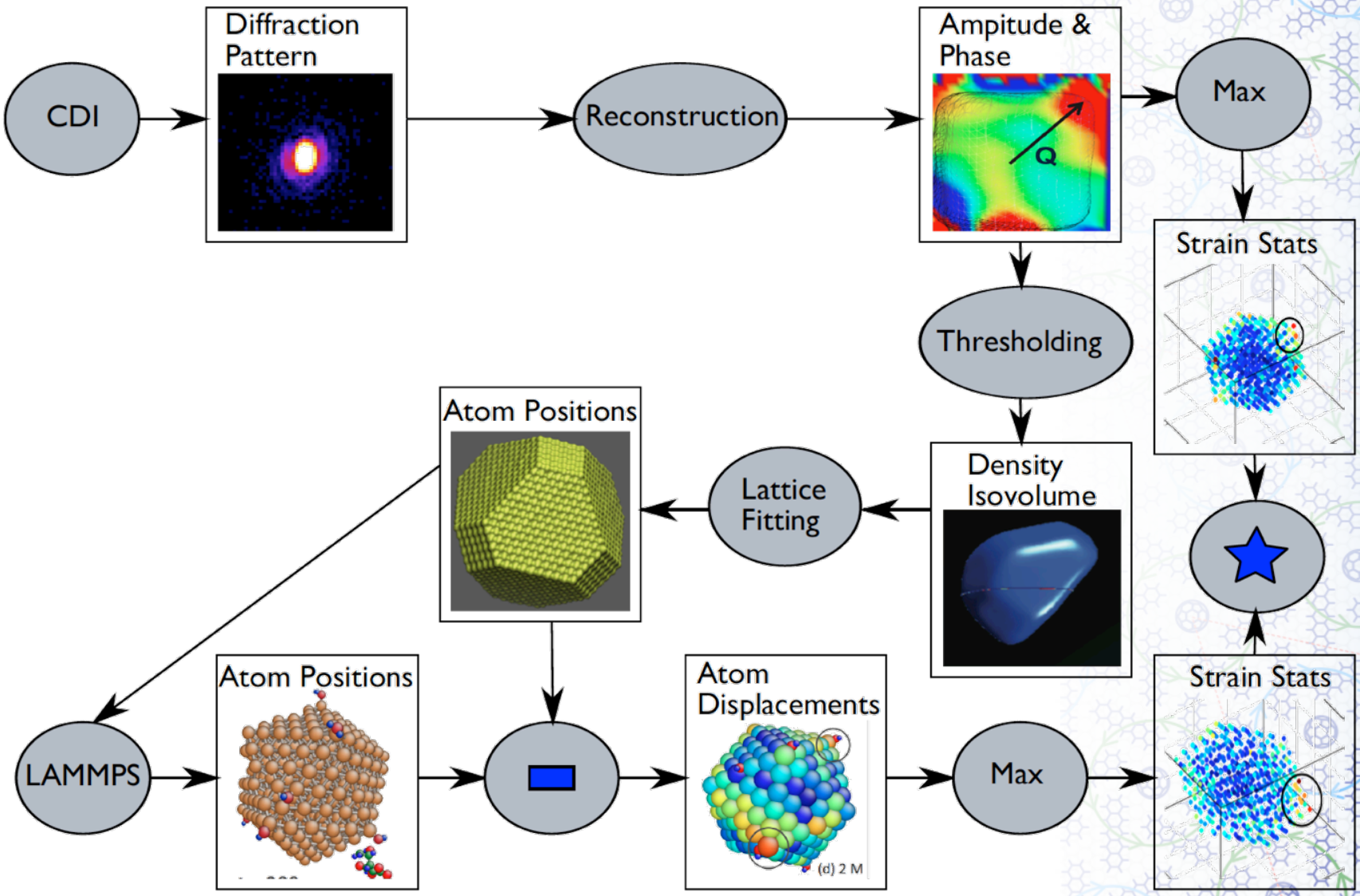


Source: B. Narayanan, S. Deshmukh, S. Sankaranarayanan

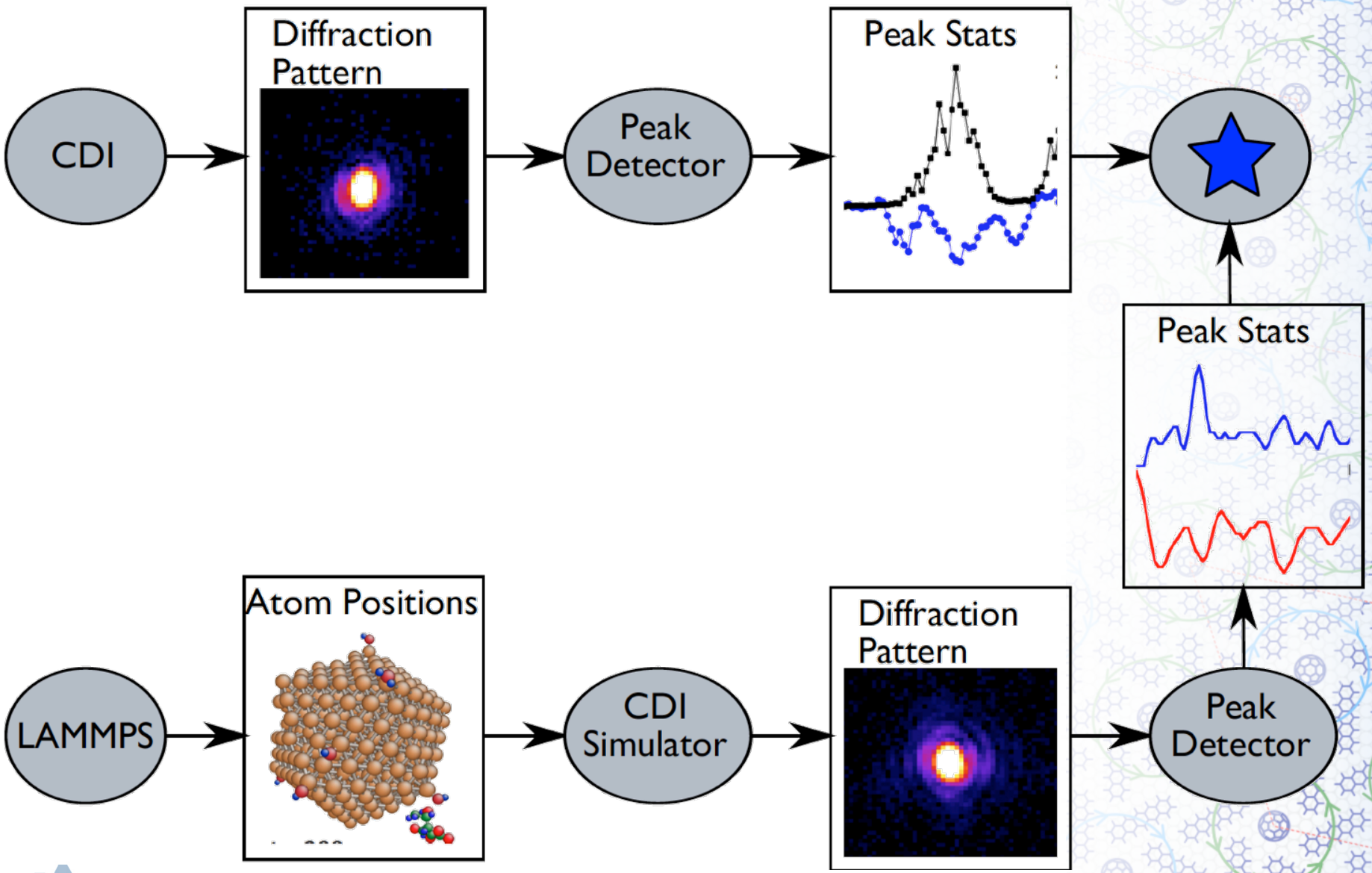
Structural dynamics



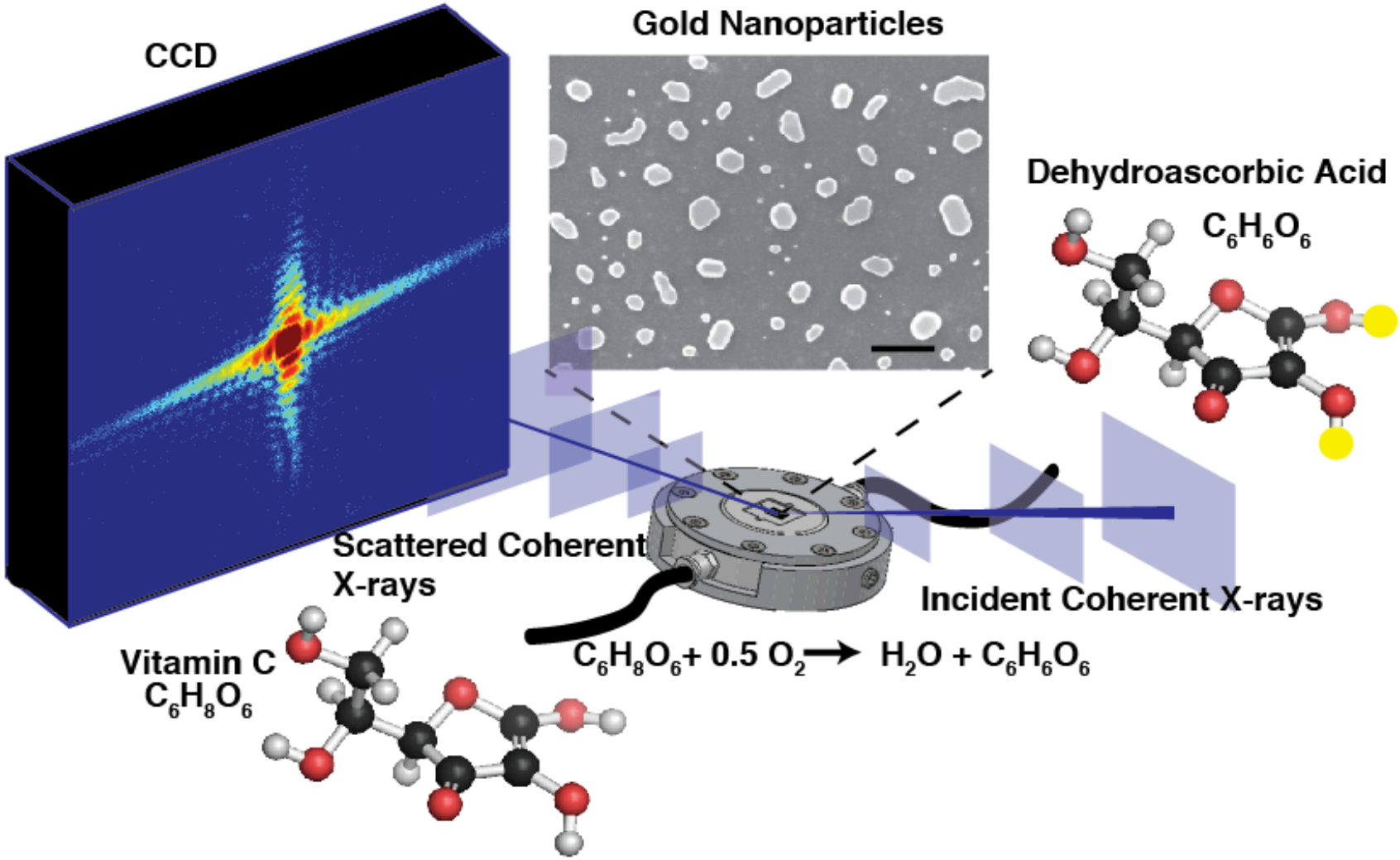
Forward Problem Full Workflow



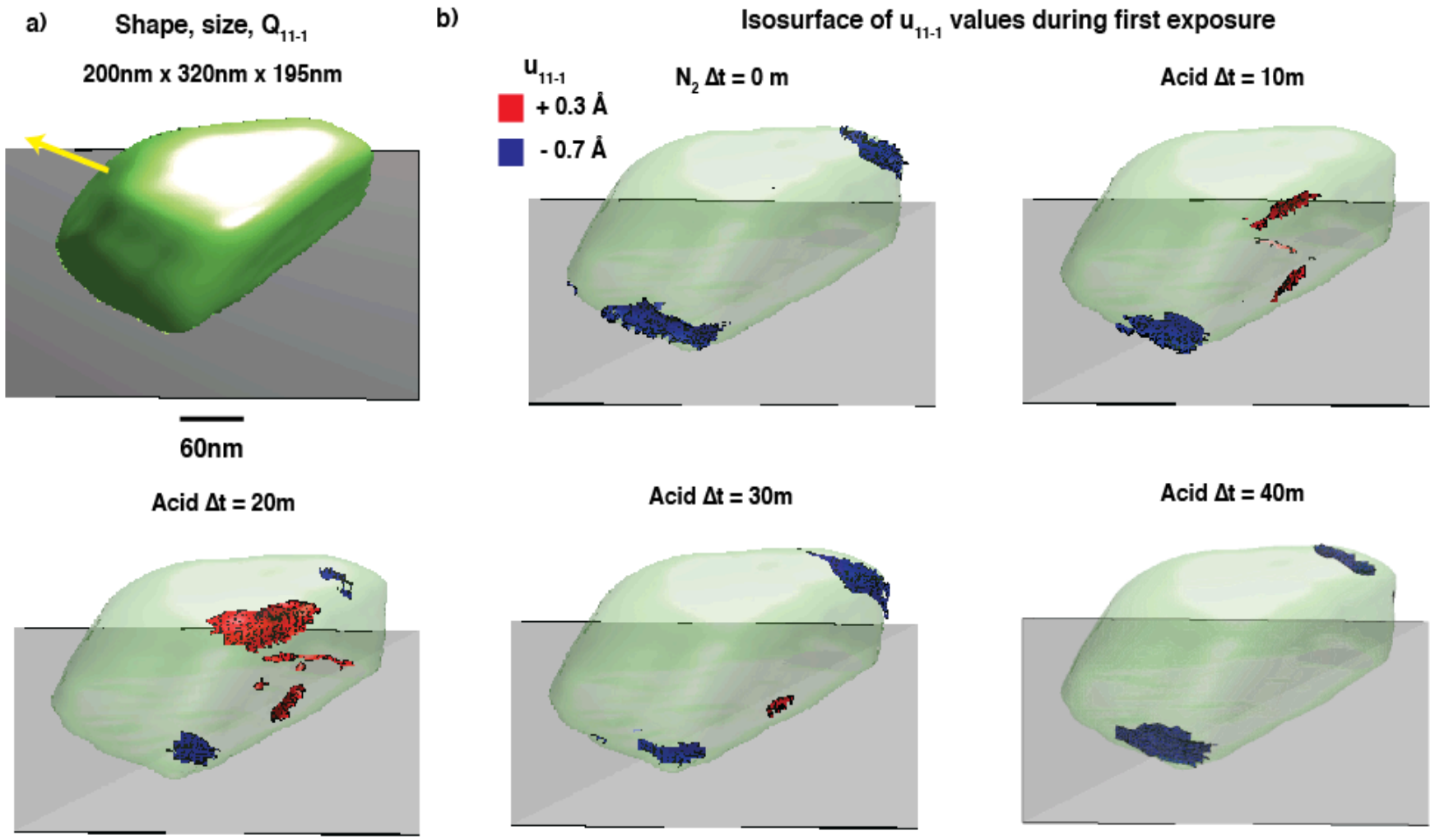
Reverse Problem Full Workflow



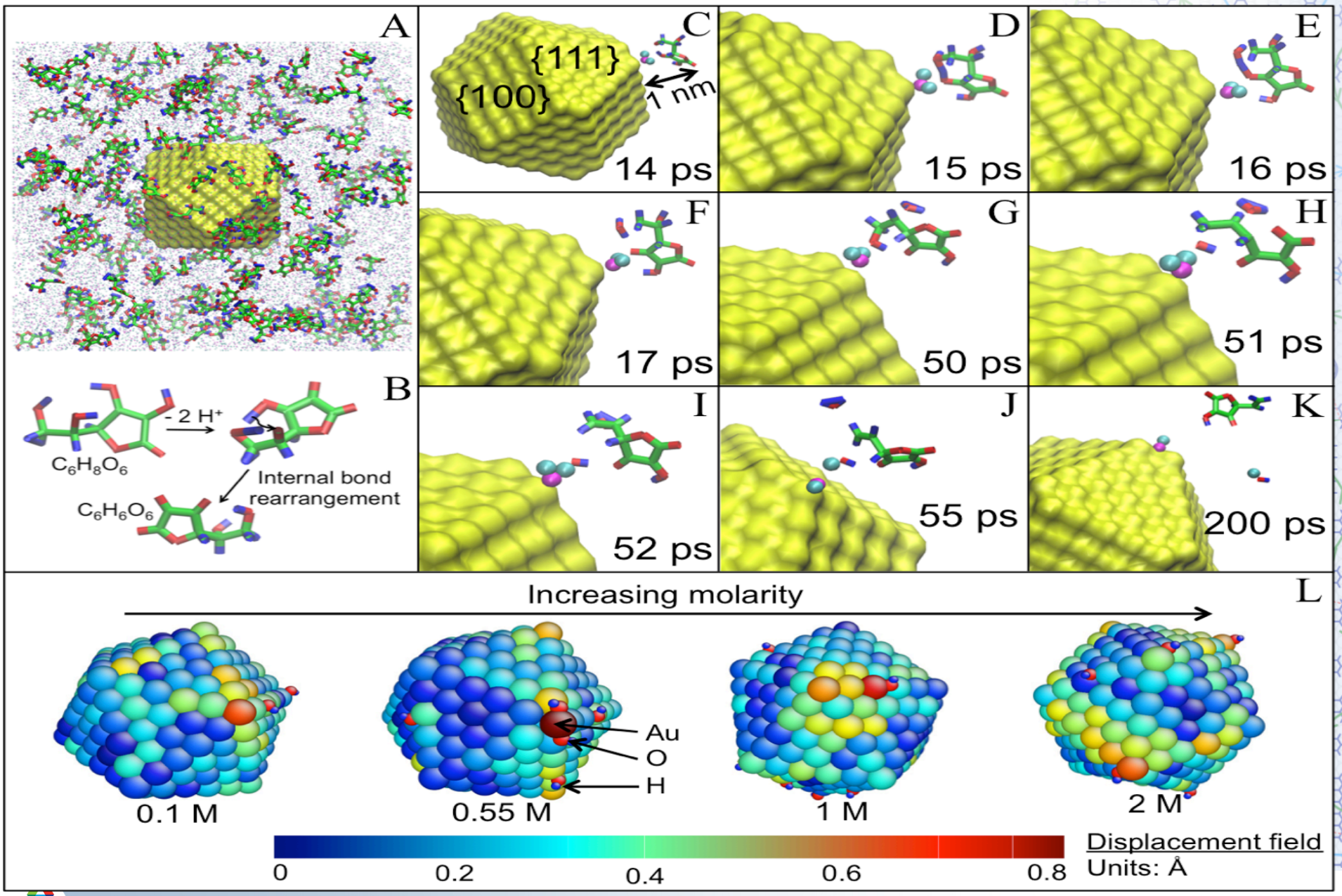
Nanocatalytic activity under *operando* conditions



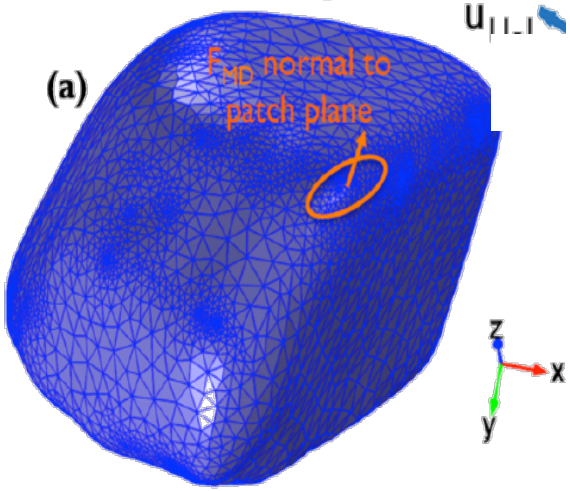
Displacement field dynamics during the first ascorbic acid exposure



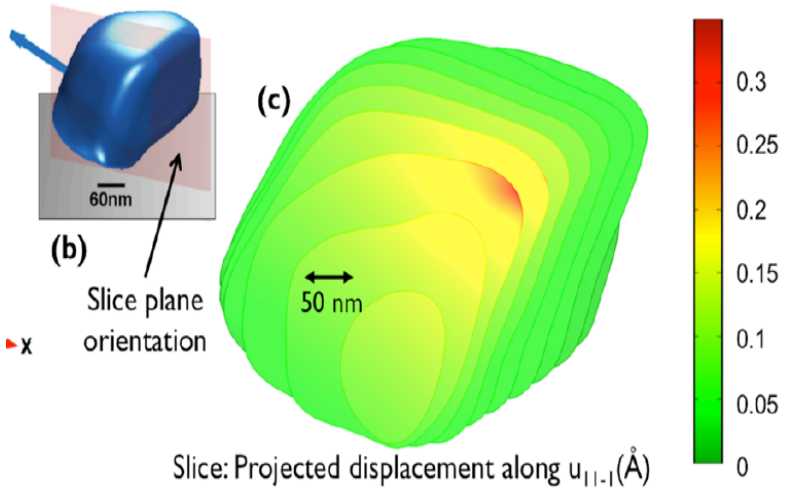
Mechanism of gold lattice strain induced by ascorbic acid

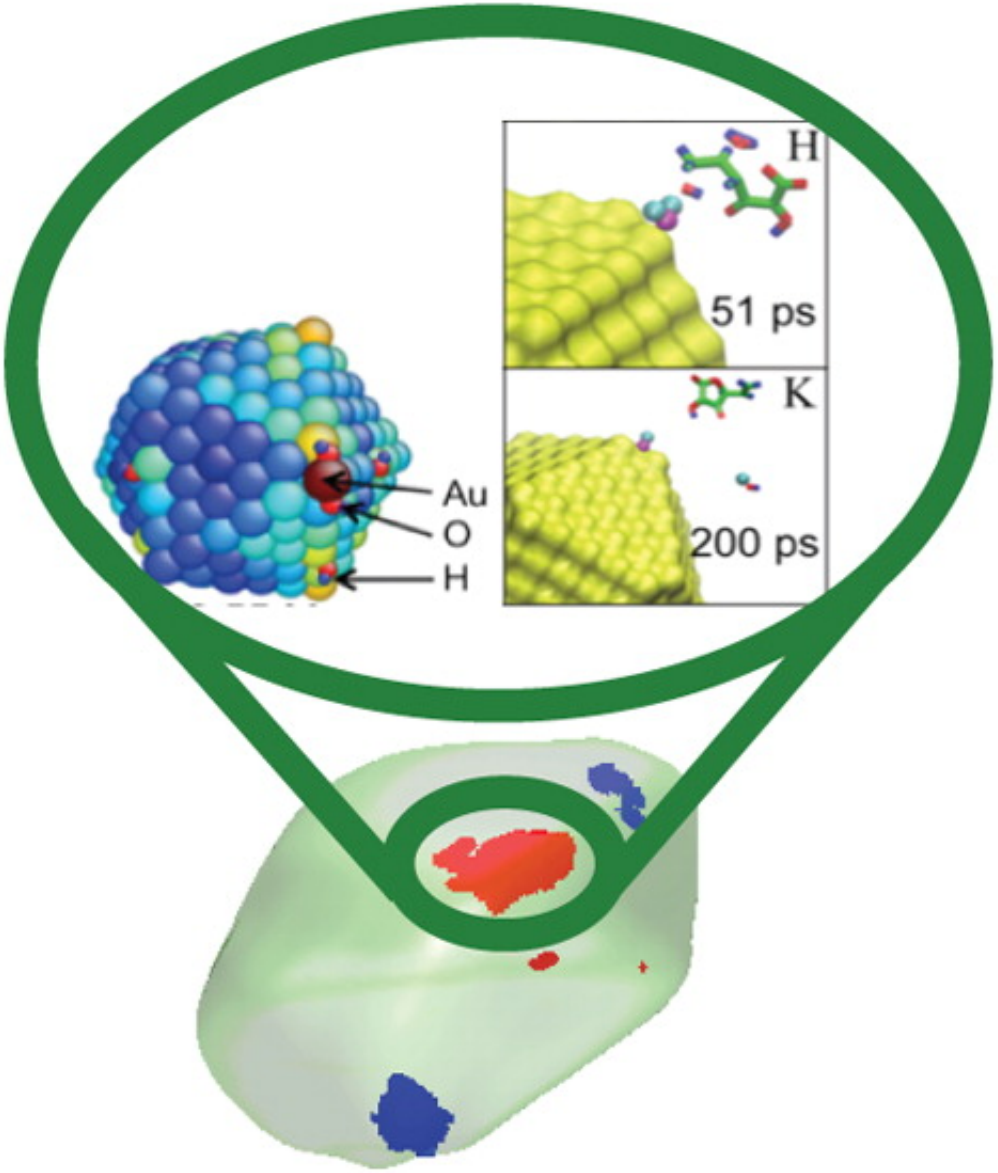


Direct comparison between CDI and Simulations

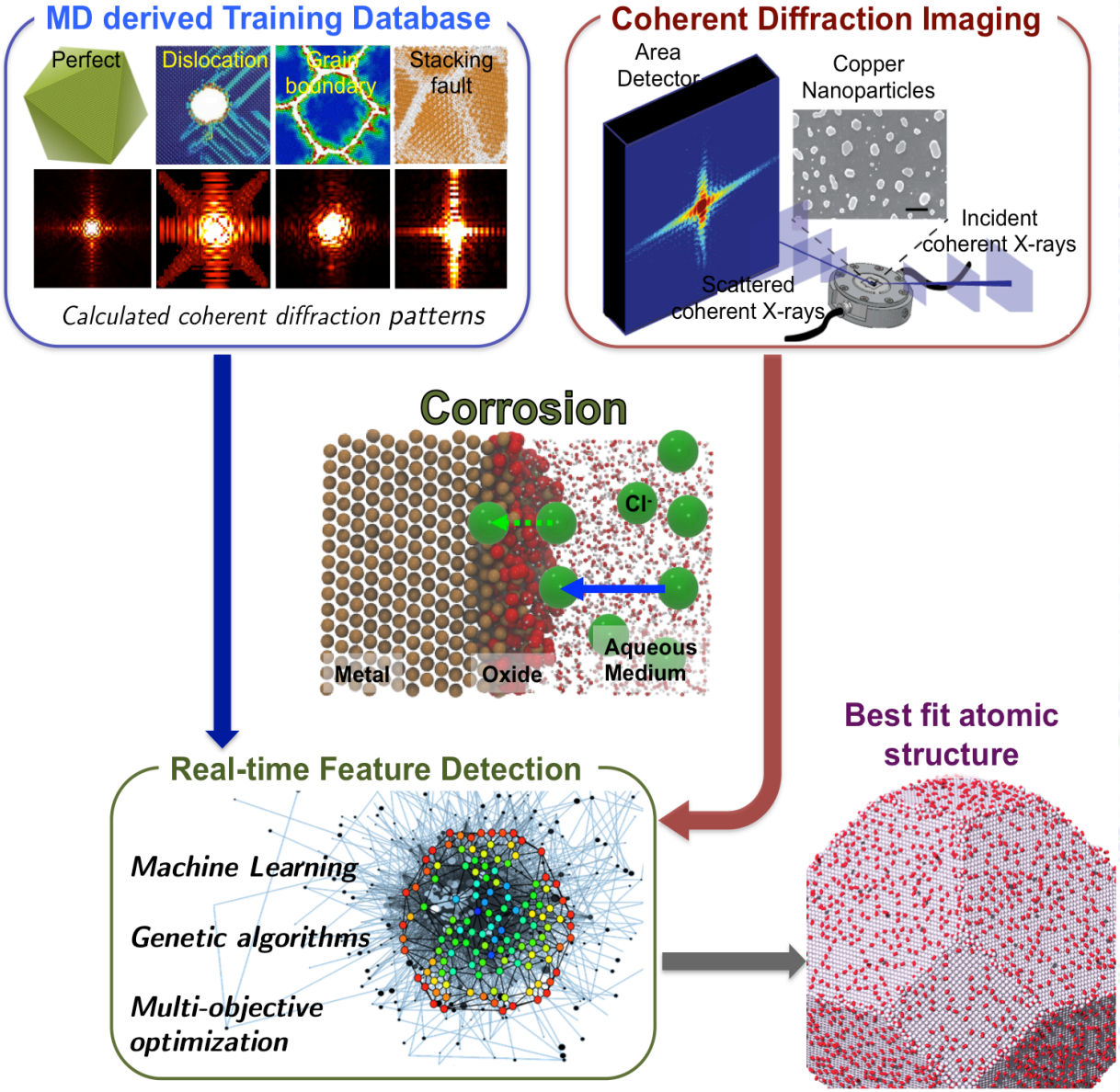


MD + Finite element



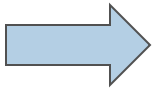


A.I. cdi: Atomistically Informed CDI



Future Directions and Perspectives

- Rapid advances in high performance computing have allowed for mesoscopic simulations with atomistic precision
- Reactive simulations of material interfaces with millions and billions of atoms are now possible
- Accuracy of the force-field remains a key challenge and limitation
- Efficient and elegant ways to bridge the gap between the electronic and atomistic length scales are needed



Our basis expansion technique to achieve a flexible form of force field is a step towards this direction

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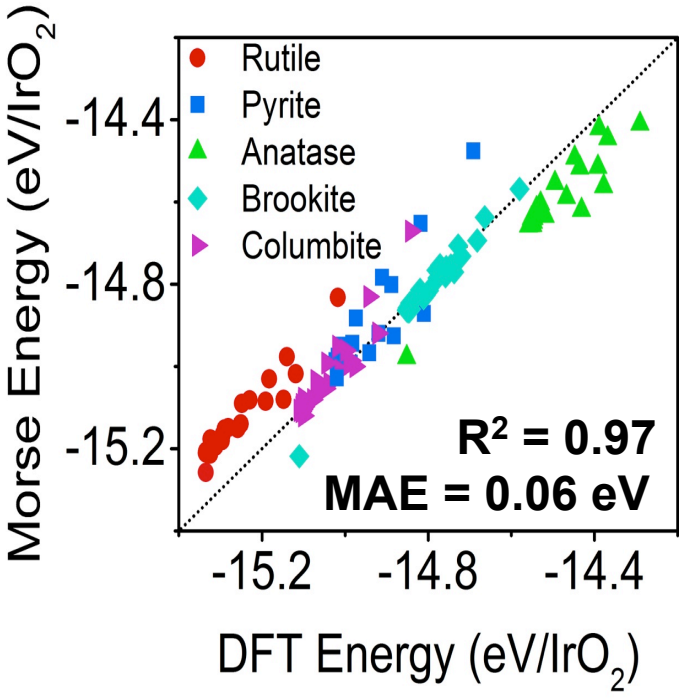
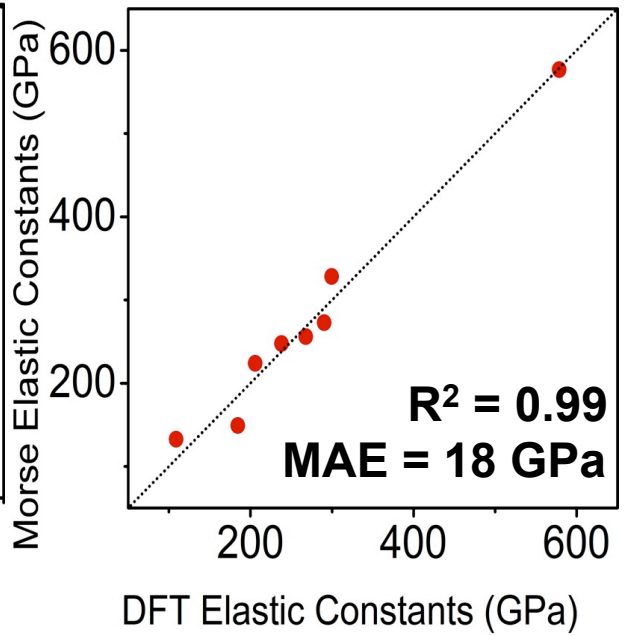
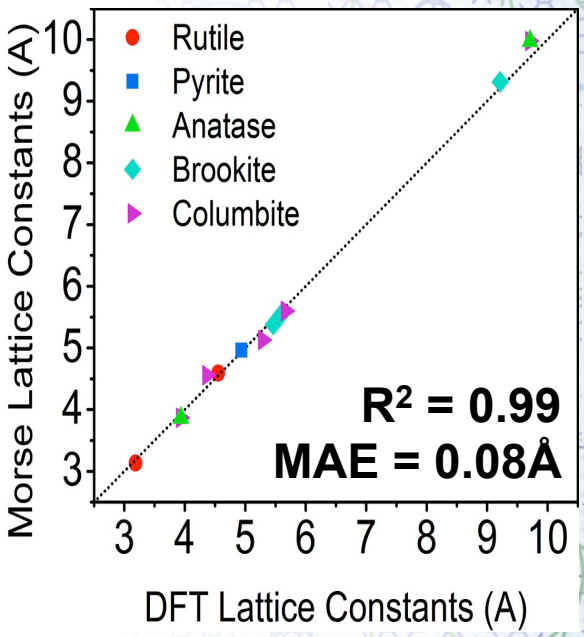
IrO_2: fitted results





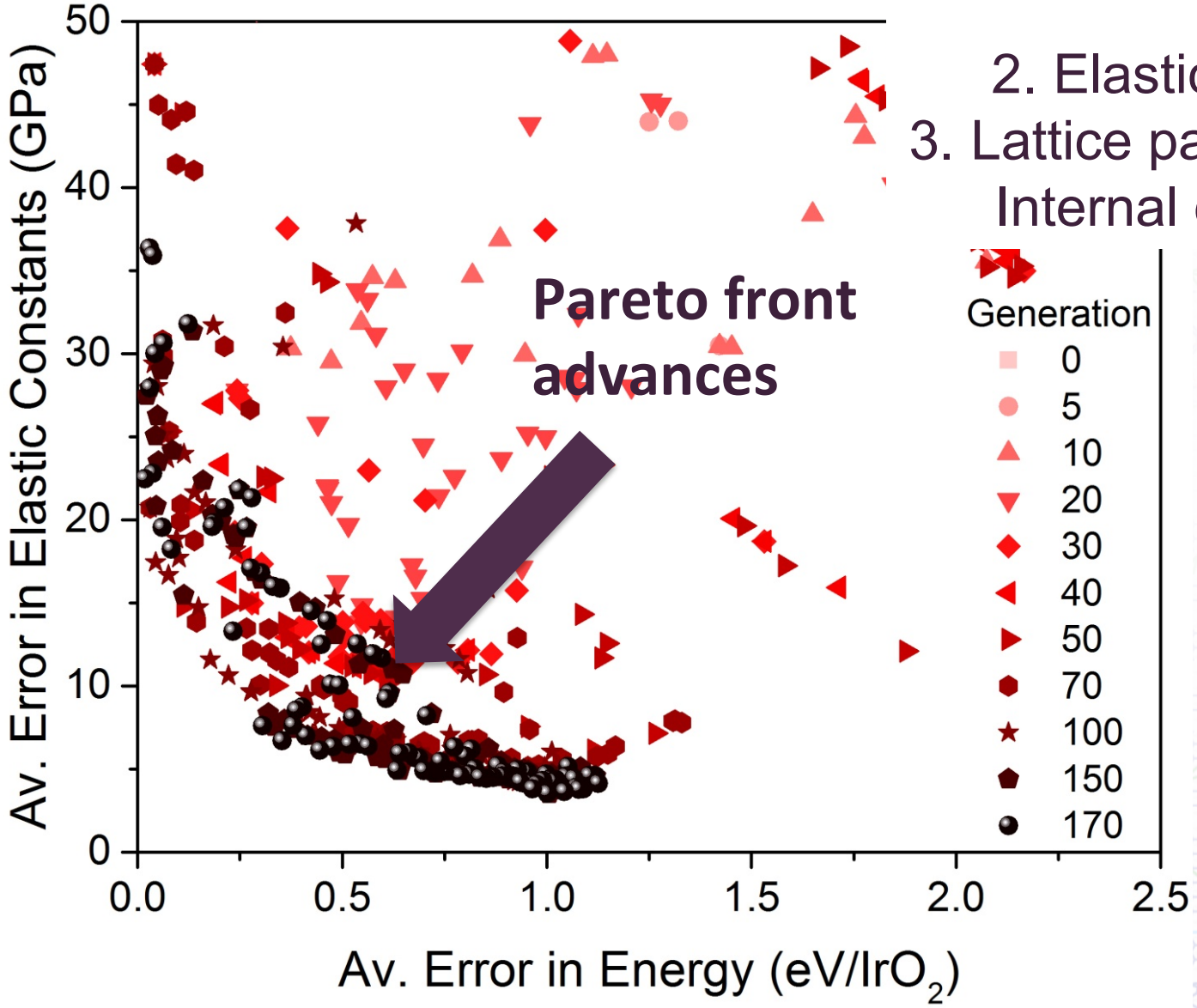


Accuracies represent a trade-off between objectives

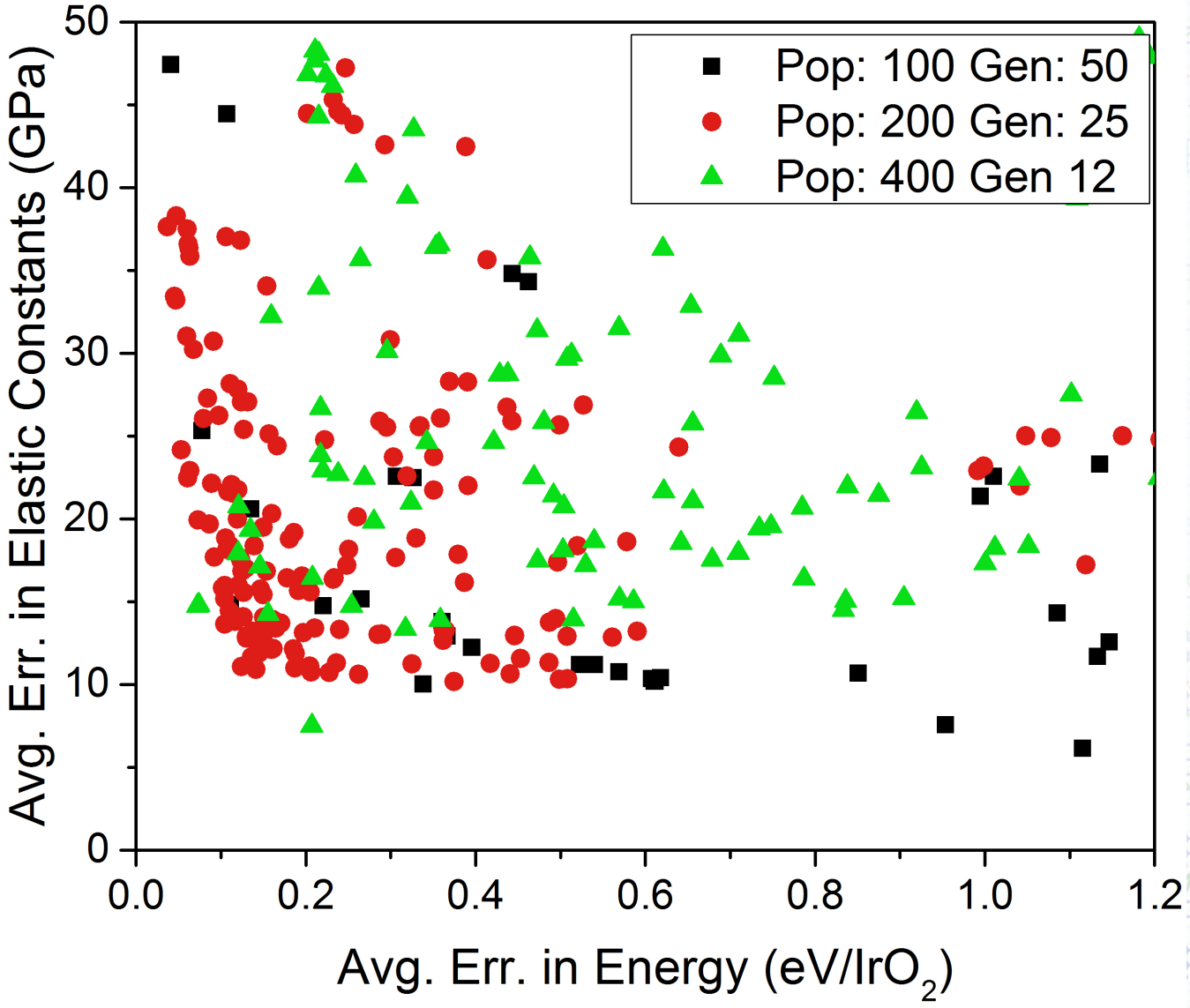
Energy-only fit gives MAE of 5 meV/atom

Multi-objective GA optimization

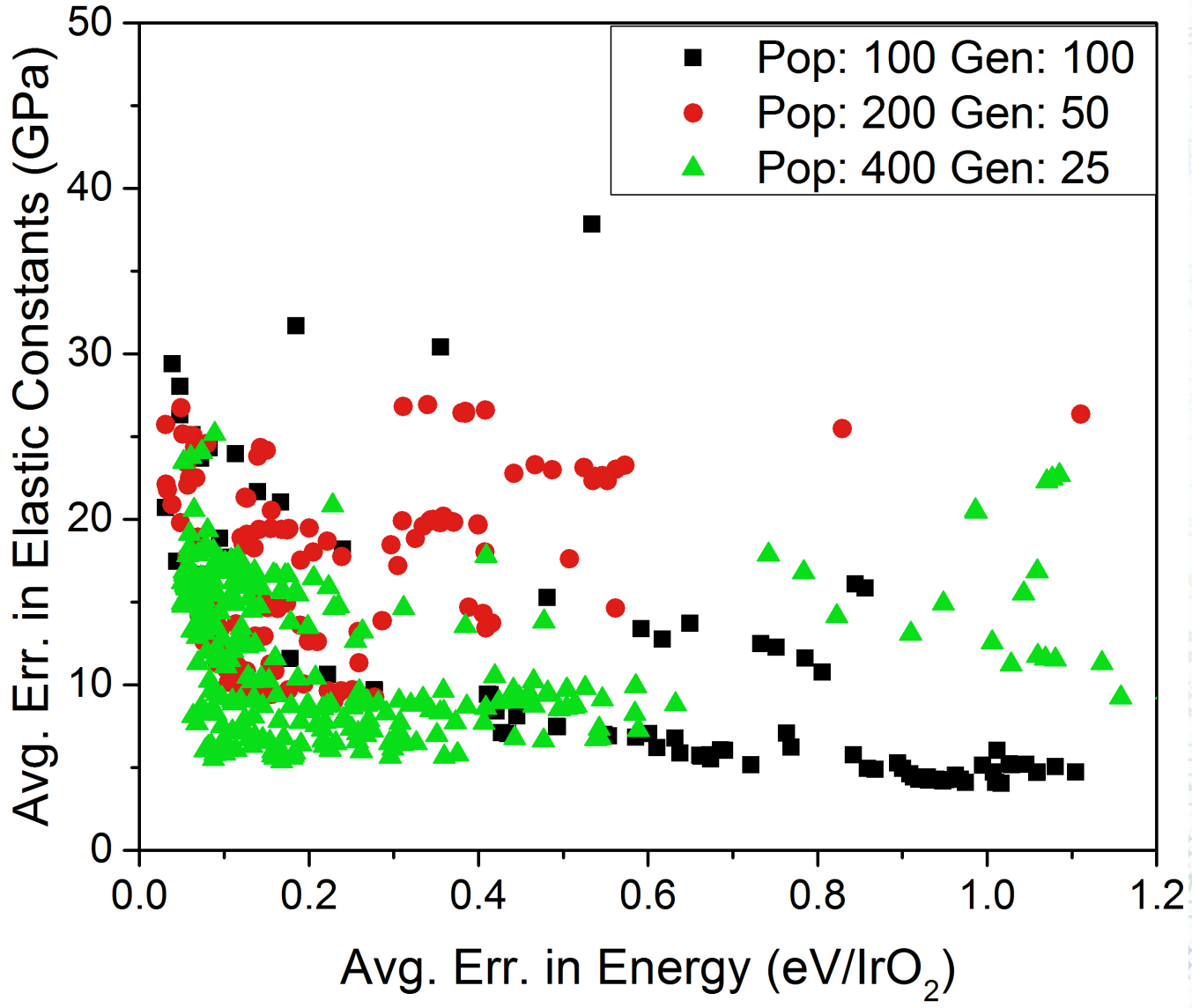
Objectives:
1. Energies
2. Elastic Constants
3. Lattice parameters & Internal coordinates



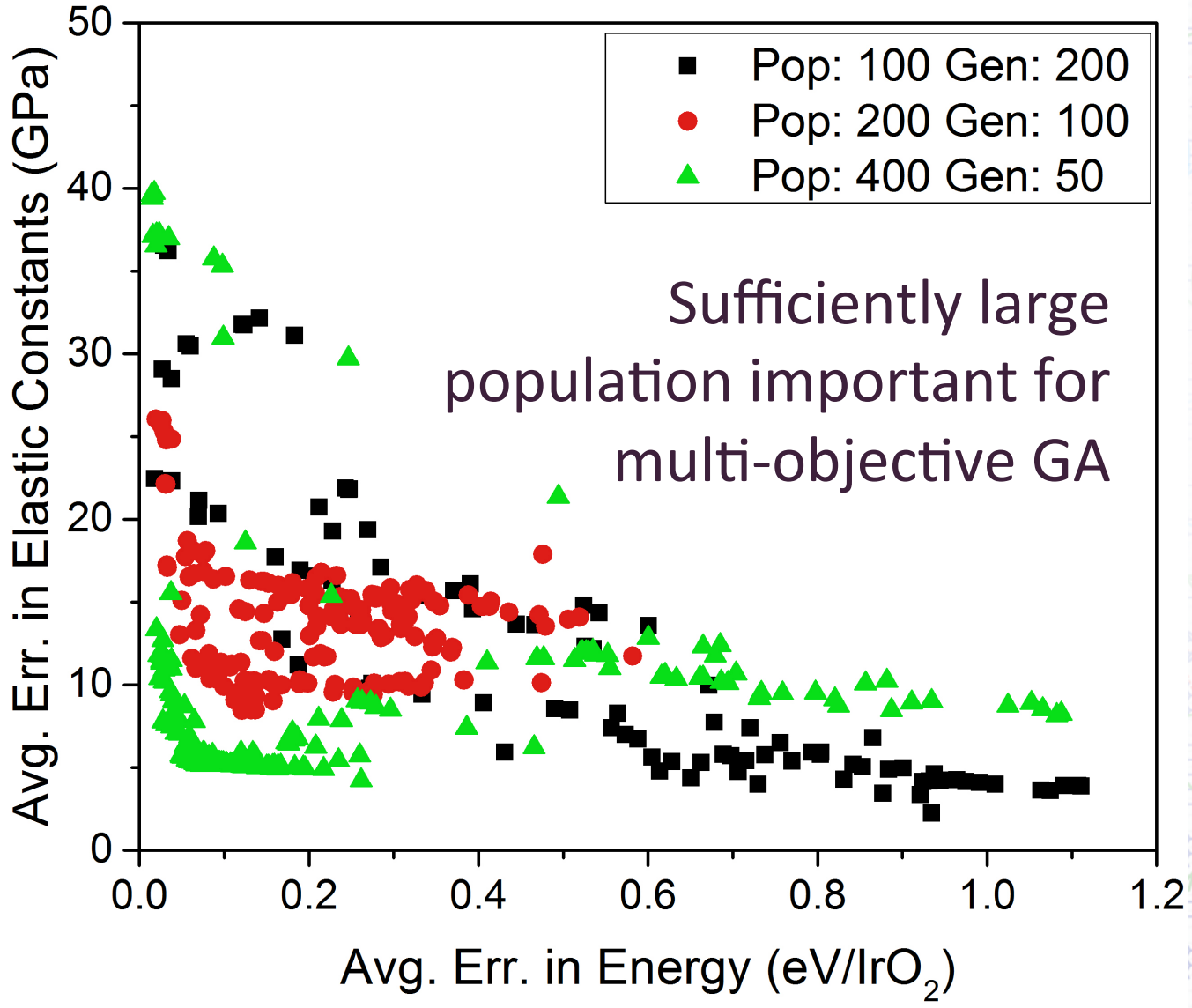
Population size effects: 5000 evaluations



Population size effects: 10000 evaluations



Population size effects: 20000 evaluations



Population size effects: 100th generation