Machine learned interatomic potentials

Gábor Csányi

UNIVERSITY OF CAMBRIDGE

A decade old promise. Where are we?

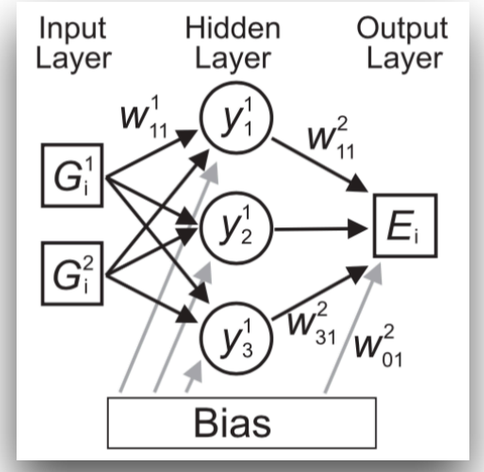
PRL **98,** 146401 (2007) PHYSICAL REVIEW LETTERS week ending 6 APRIL 2007

Generalized Neural-Network Representation of High-Dimensional Potential-Energy Surfaces

Jörg Behler and Michele Parrinello

Department of Chemistry and Applied Biosciences, ETH Zurich, USI-Campus, Via Giuseppe Buffi 13, CH-6900 Lugano, Switzerland

(Received 27 September 2006; published 2 April 2007)



PRL **104,** 136403 (2010) PHYSICAL REVIEW LETTERS week ending 2 APRIL 2010

Gaussian Approximation Potentials: The Accuracy of Quantum Mechanics, without the Electrons

Albert P. Bartók and Mike C. Payne

Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge, CB3 0HE, United Kingdom

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Gábor Csányi

Engineering Laboratory, University of Cambridge, Trumpington Street, Cambridge, CB2 1PZ, United Kingdom

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$$\begin{aligned} \mathbf{Q}_M &= \mathbf{C}_M + \mathbf{C}_{MN}(\mathbf{\Lambda} + \sigma^2\mathbf{I})^{-1}\mathbf{C}_{NM}, \\ \varepsilon_* &= \mathbf{k}_*^T \mathbf{Q}_M^{-1} \mathbf{C}_{MN}(\mathbf{\Lambda} + \sigma^2\mathbf{I})^{-1}\mathbf{y}, \end{aligned}$$

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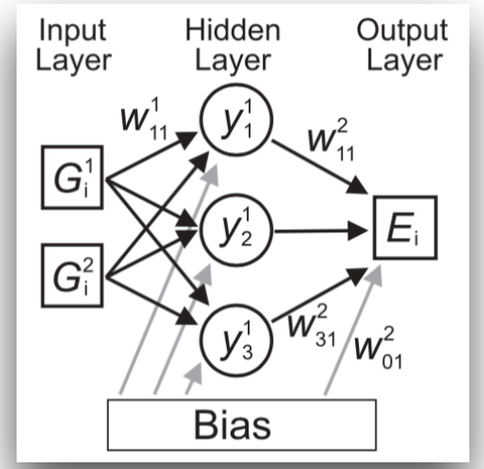

2020 “message”: short range QM regression problem is “solved”.

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Summary of Si material properties

PHYSICAL REVIEW X

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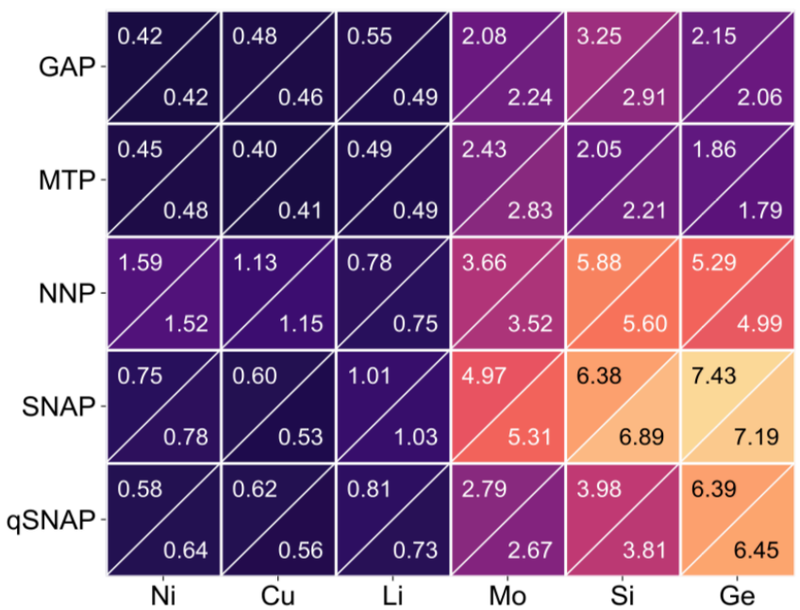
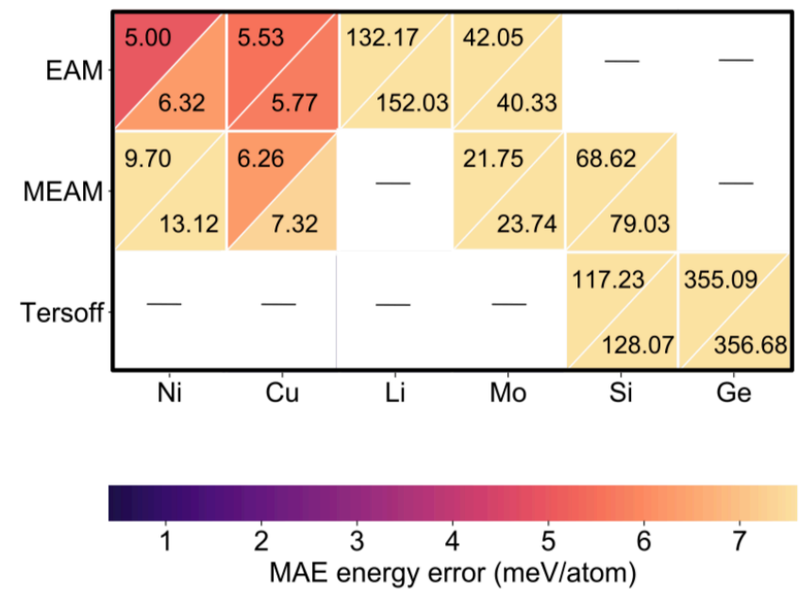
Machine Learning a General-Purpose Interatomic Potential for Silicon

Albert P. Bartók, James Kermode, Noam Bernstein, and Gábor Csányi

Phys. Rev. X **8**, 041048 – Published 14 December 2018

Comparing performance of different ML schemes on elemental metals





(a) Mean absolute errors in predicted energies

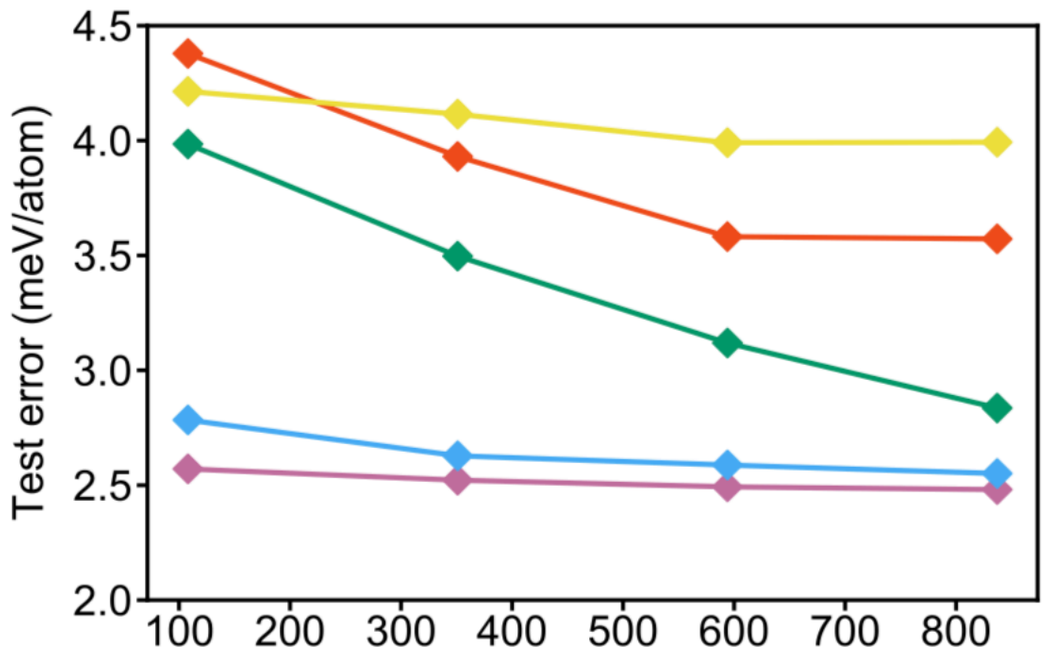


Figure 4: MAE in predicted (a) energies (

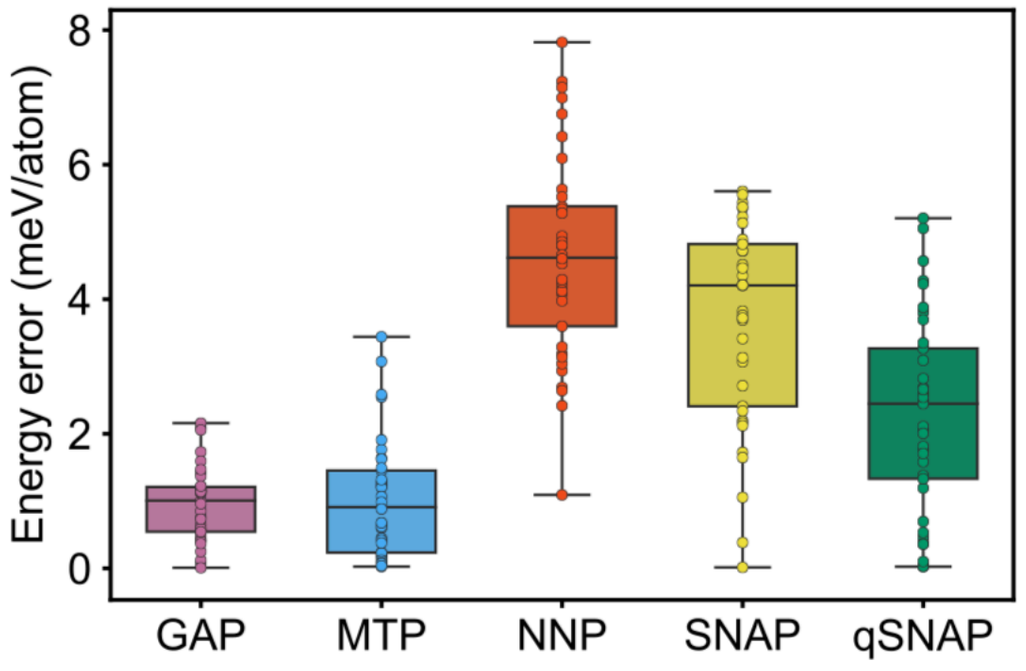


Figure 6: Error distributions in (a) predicted dynamics

Performance and Cost Assessment of Machine Learning Interatomic Potentials

Published as part of The Journal of Physical Chemistry virtual special issue "Machine Learning in Physical Chemistry".

Yunxing Zuo, Chi Chen, Xiangguo Li, Zhi Deng, Yiming Chen, Jörg Behler, Gábor Csányi, Alexander V. Shapeev, Aidan P. Thompson, Mitchell A. Wood, and Shyue Ping Ong*

Cite This: *J. Phys. Chem. A* 2020, 124, 731–745

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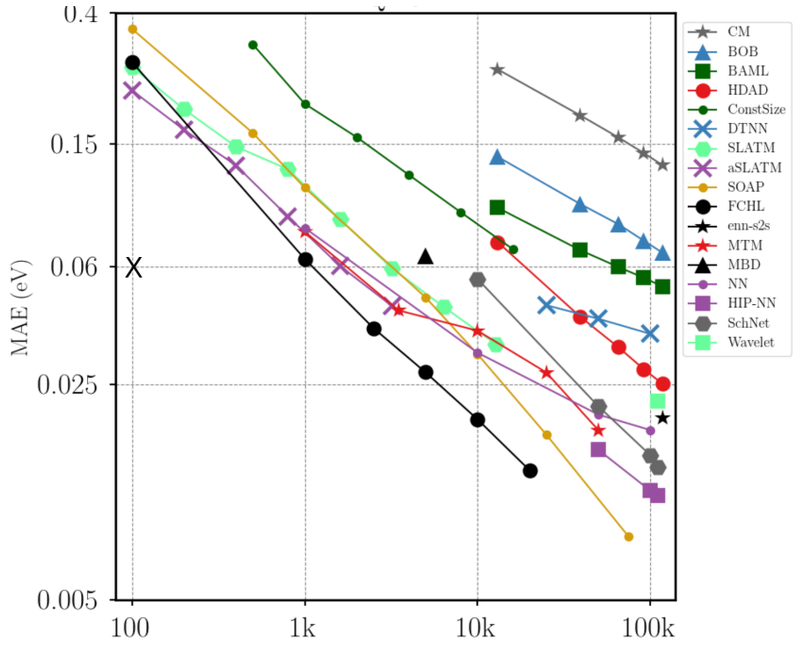
Molecular successes (of others, mostly)

QM9 data set
(small organic molecules)



(Courtesy of O. A von Lilienfeld)

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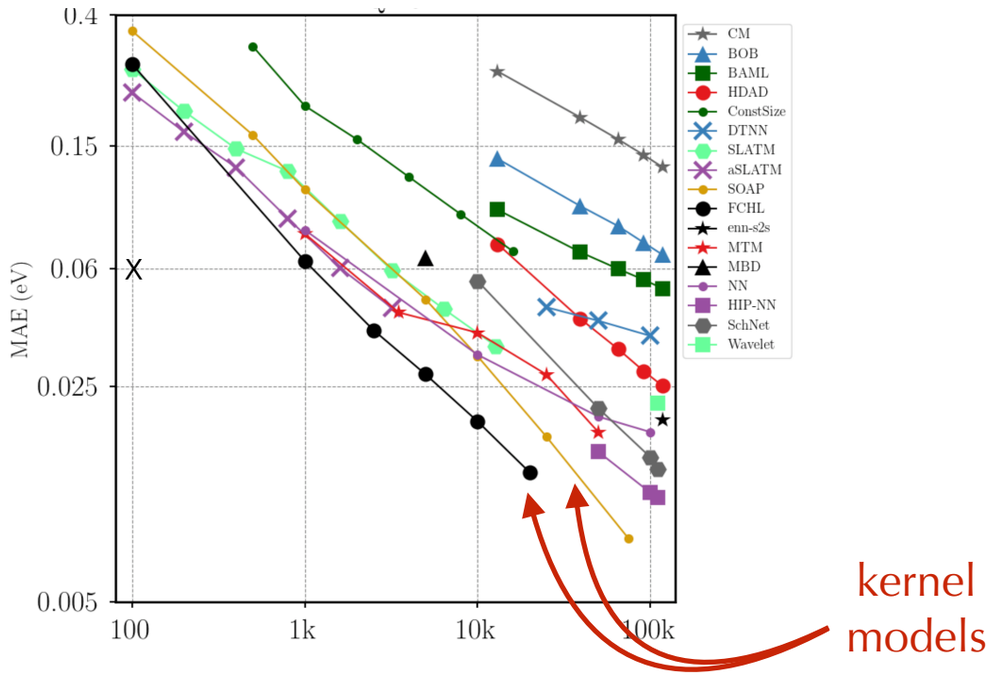

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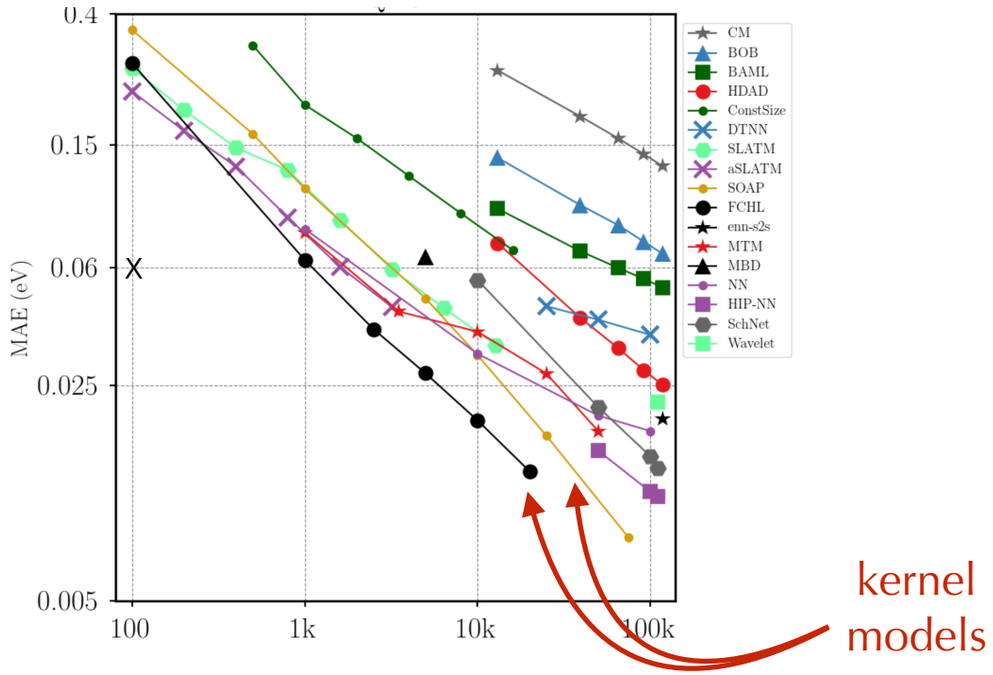
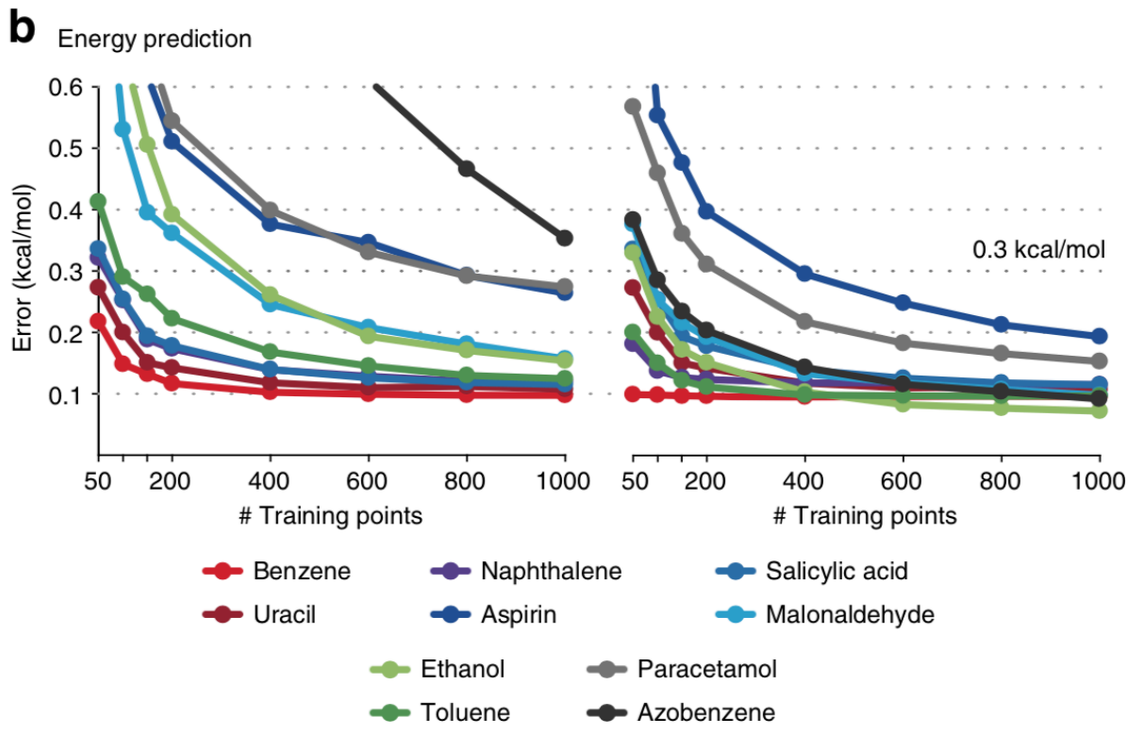


(Courtesy of O. A von Lilienfeld)

KRR models for custom molecular force fields (sGDML of Tkatchenko et al.)



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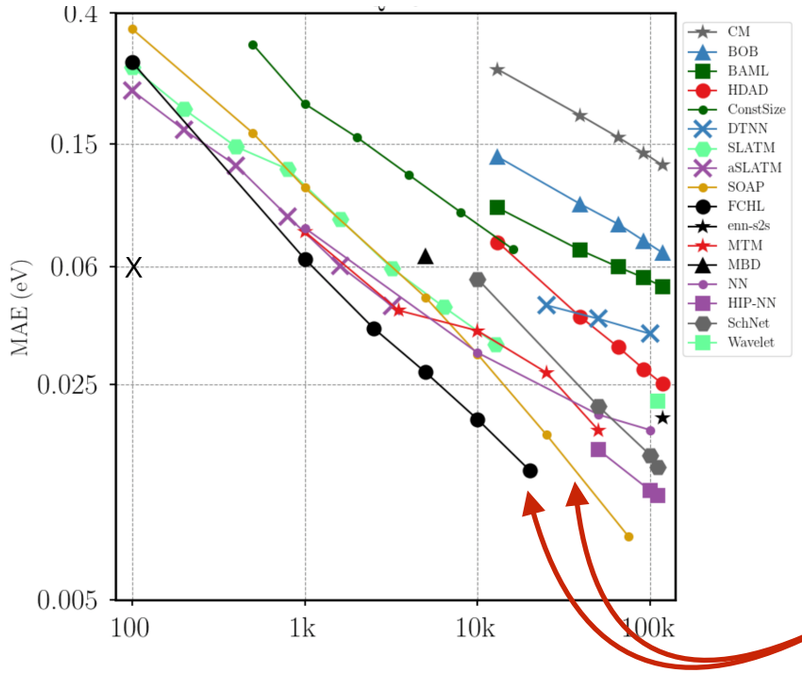

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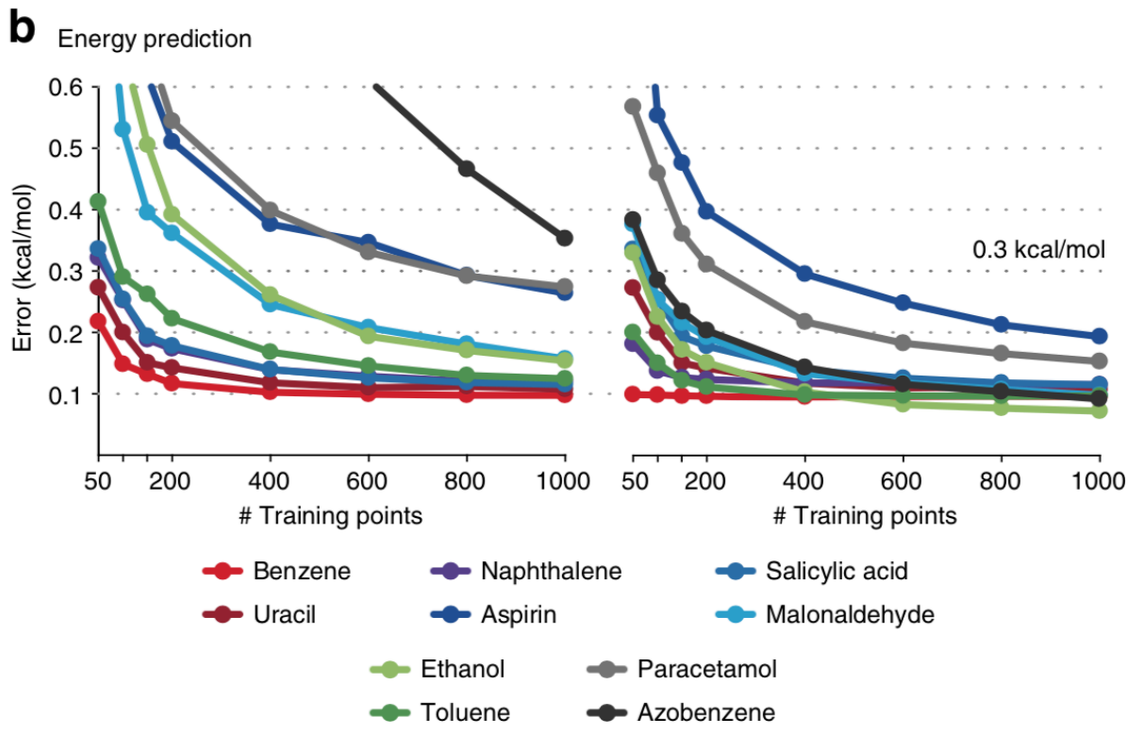
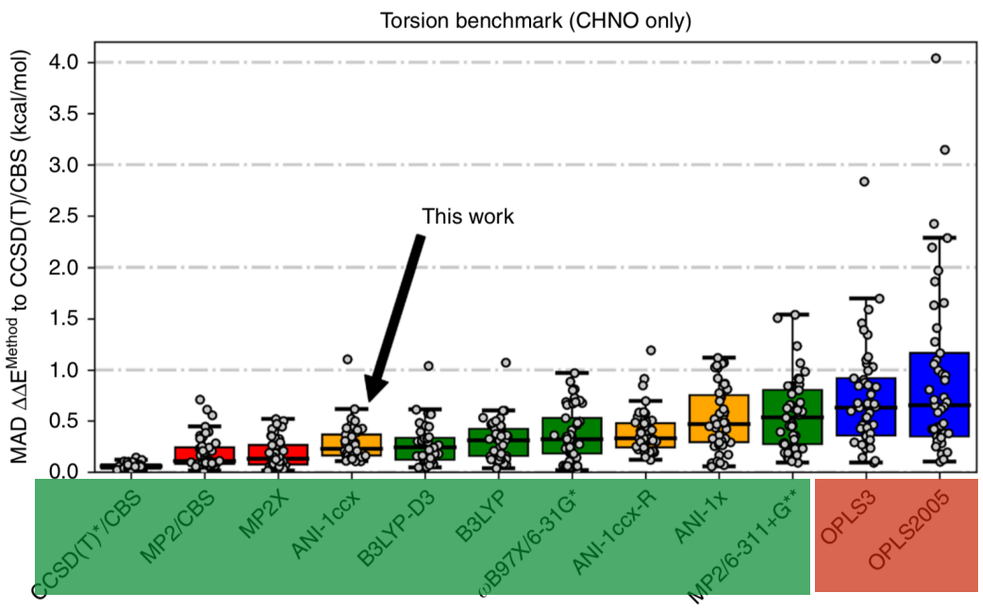
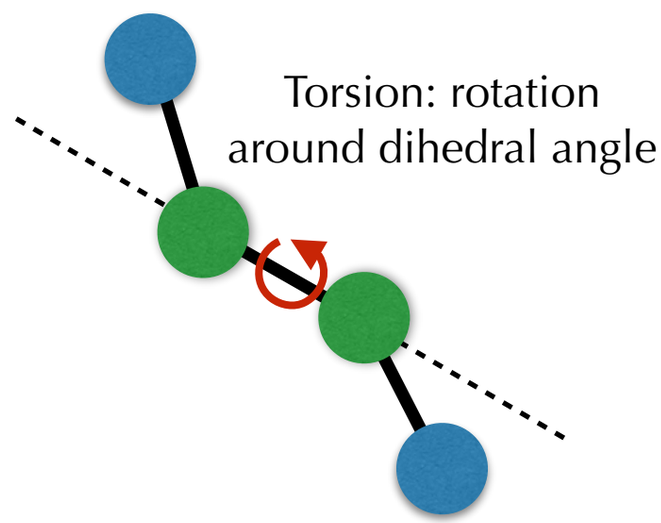


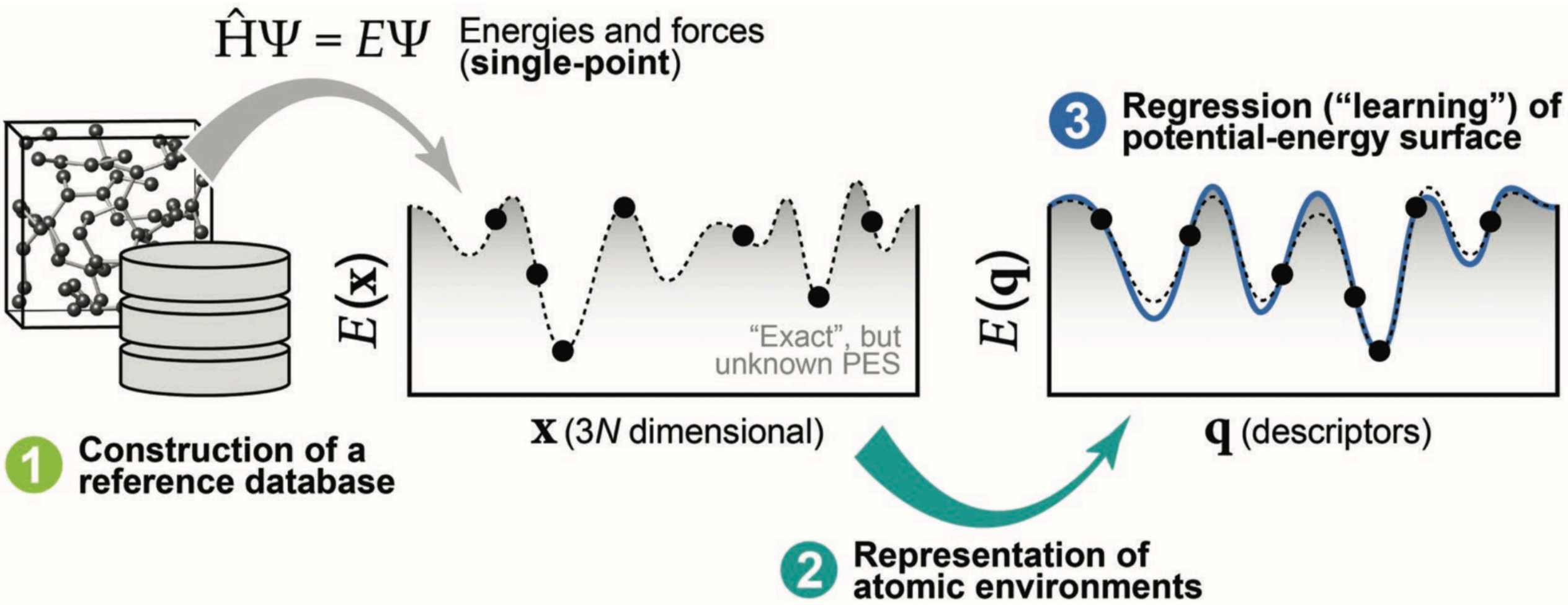
ANI-CC model of Isayev, Roitberg and co. for arbitrary molecules

2b-3b environment descriptors
3-layer feed-forward ANN





Potentials as "function interpolators"



PROGRESS REPORT
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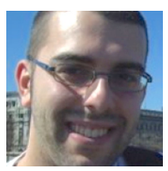
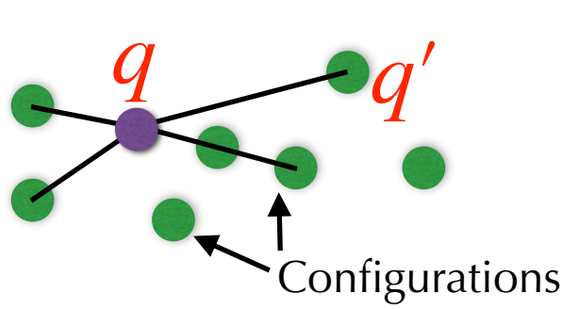
Machine Learning Interatomic Potentials as Emerging Tools for Materials Science

Volker L. Deringer, Miguel A. Caro, and Gábor Csányi*



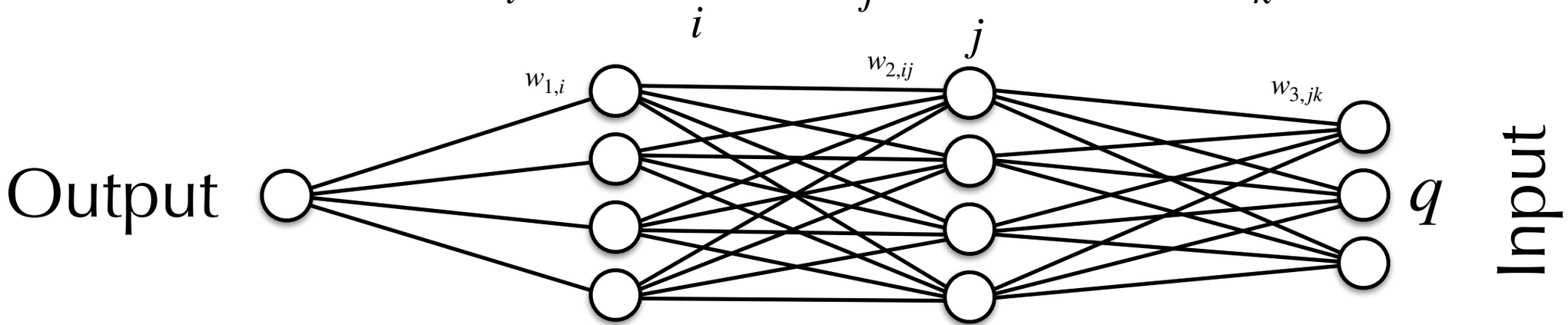
Regression with representation q

- Linear regression $f(q) = \sum_i^N x_i b_i(q)$ basis functions b
- Kernel regression $f(q) = \sum_i^N x_i K(q, q_i)$ similarity kernel $K(q, q')$
  - Also linear but in transformed space
  - Basis set scales with data
  - Equivalent to neural network with 1 hidden layer
  - Also known as "Gaussian process regression"



- Nonlinear regression
  - E.g. >1 hidden layer neural feed-forward networks

$$f(q) = h(b_0 + \sum_i^{N_1} w_{1,i} h(b_{1,i} + \sum_j^{N_2} w_{2,ij} h(b_{2,j} + \sum_k^{\dim(q)} w_{3,jk} q_k)))$$



Regression with representation q

More stringent

Requirements on the properties of the representation q

Less stringent

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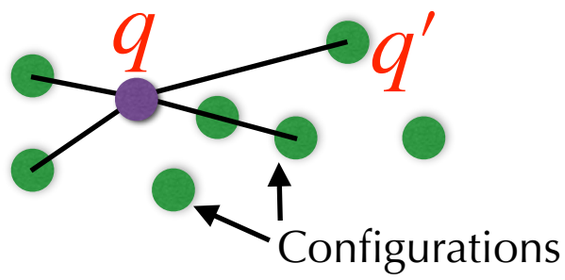

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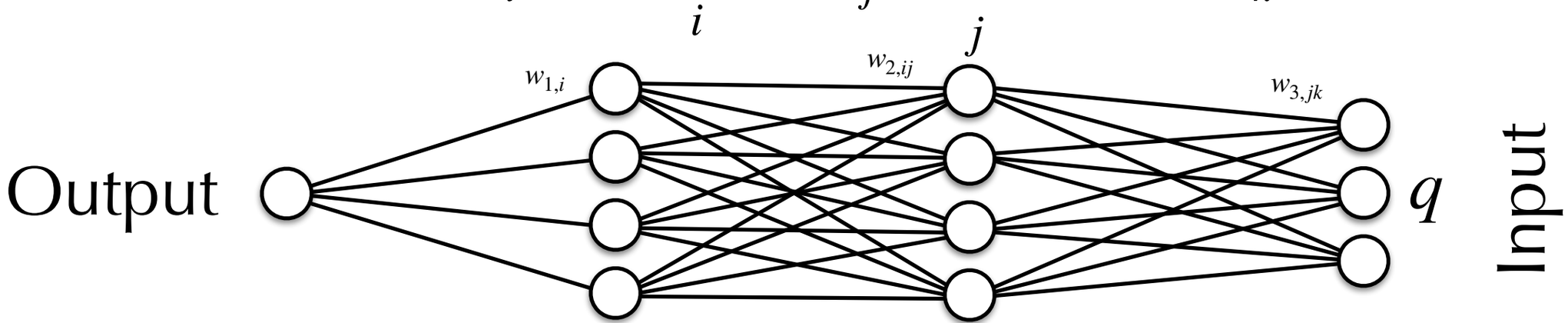
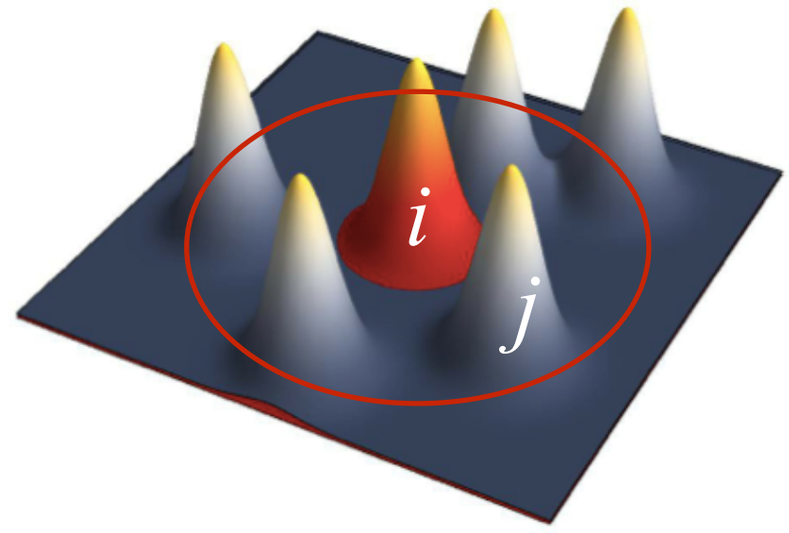


Representation and kernel: atomic neighbour density

$$\rho^{(i)}(\mathbf{r}) = \sum_j G(\mathbf{r} - \mathbf{r}_{ij})\, f_{\text{cut}}(|\mathbf{r}_{ij}|)$$

Convolution kernel $\quad G(\mathbf{r}) = \begin{cases} \delta(\mathbf{r}) \\ e^{-|\mathbf{r}|^2/2\sigma^2} \end{cases}$



short range cutoff: compact support

ρ is permutational invariant, smooth and continuous

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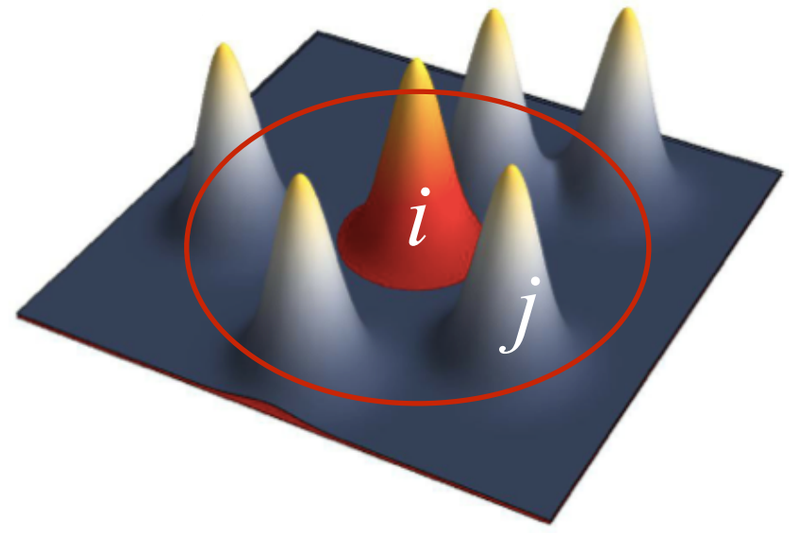

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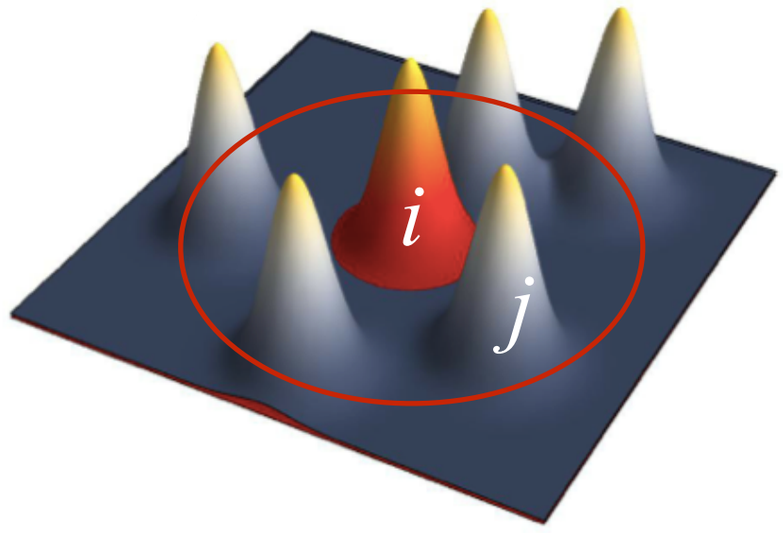

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Smooth overlap of atomic positions (SOAP)

$$\mathbf{p} \cdot \mathbf{p}' = \int_{\hat{R} \in SO_3} d\hat{R} \left| \int d\mathbf{r} \rho(\mathbf{r}) \rho'(\hat{R}\mathbf{r}) \right|^2$$

SOAP kernel: $K(R, R') \equiv K(\rho, \rho') = |\mathbf{p} \cdot \mathbf{p}'|^{\zeta}$

PHYSICAL REVIEW B **87**, 184115 (2013)

On representing chemical environments

Albert P. Bartók,[1,*] Risi Kondor,[2] and Gábor Csányi[1]

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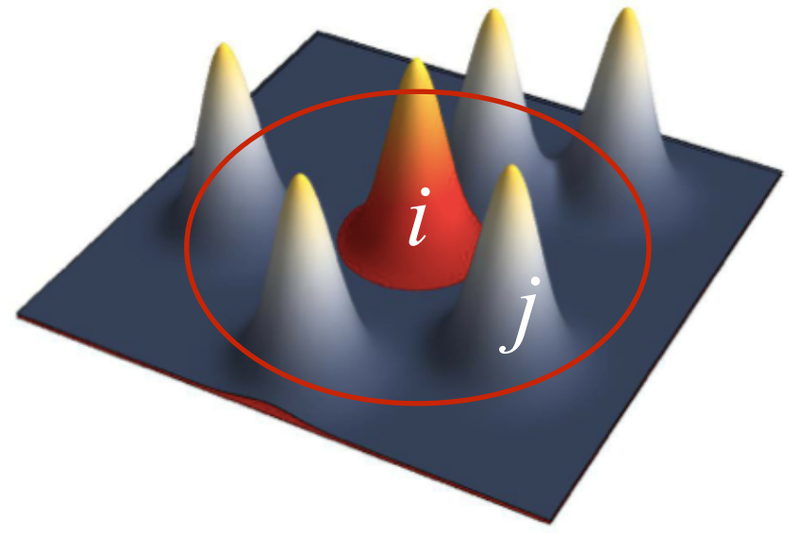

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PHYSICAL REVIEW B **87**, 184115 (2013)

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Many other descriptors (ACSF of Behler and ANI, FCHL, MBTR etc.) are equivalent *in the complete basis limit*

Other nice properties of the Y_{lm} basis

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- Natural extension to basis functions to expand non-scalar properties, vectors and tensors (Grisafi, Wilkins, GC, Ceriotti PRL 2018)

$$k^{\lambda}(\rho,\rho') = \int d\hat{R}\, \mathbf{D}^{\lambda}(\hat{R}) \left| \int d\mathbf{r}\rho(r)\rho'(\hat{R}\mathbf{r}) \right|^2$$

$\mathbf{D}$: Wigner matrix

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D : Wigner matrix

- Further invariants (Ralf Drautz, 2019)

$$\sum \begin{pmatrix} l_1 & l_2 & l_3 \\ m_1 & m_2 & m_3 \end{pmatrix} c_{n_1 l_1 m_1} c_{n_2 l_2 m_2} c_{n_3 l_3 m_3}$$

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$$\vdots$$

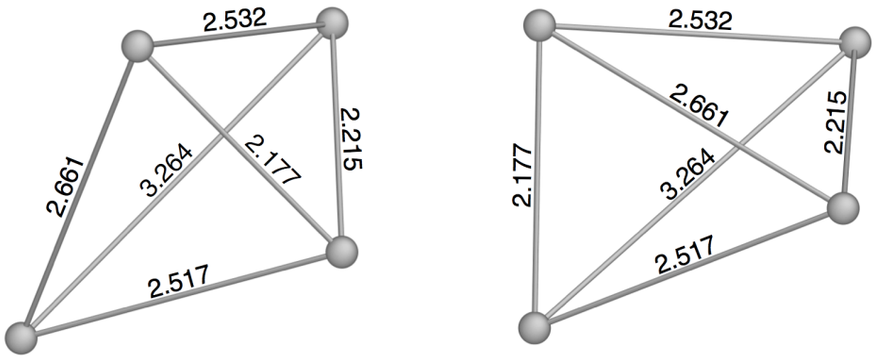
- Variant to describe entire structures, rather than atomic neighbourhoods:

$$\bar{c}_{nlm} = \sum_i c^{(i)}_{nlm} \qquad \text{average over all atoms, no cutoff}$$

$$\bar{p}_{nn'l} = \sum_m \bar{c}^{\dagger}_{nlm} \bar{c}_{nlm} \qquad \text{or cutoff > periodic unit cell}$$

(Other ways to construct kernels for entire structures: De, Bartók, GC, Ceriotti PCCP 2016)

Completeness: can we reconstruct an atomic environment from the representation?



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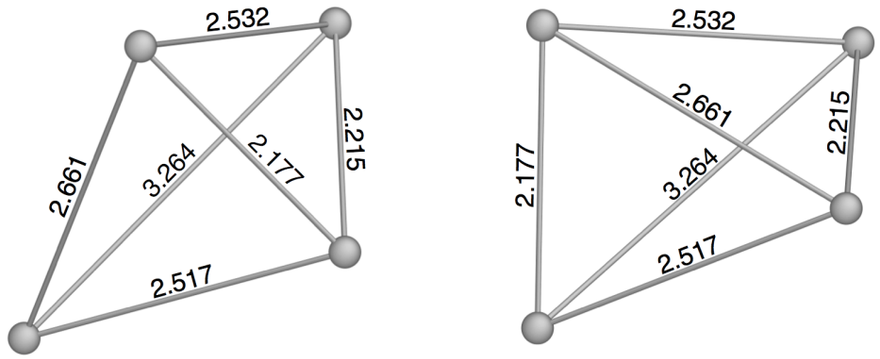
- Well known that unordered distances not enough to reconstruct a point set



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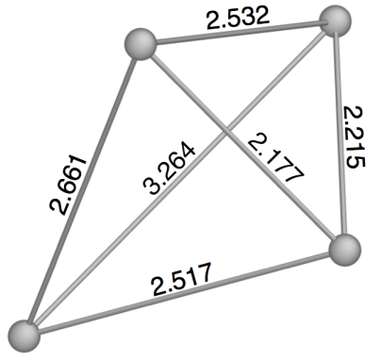
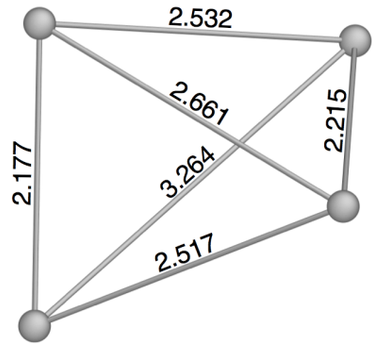
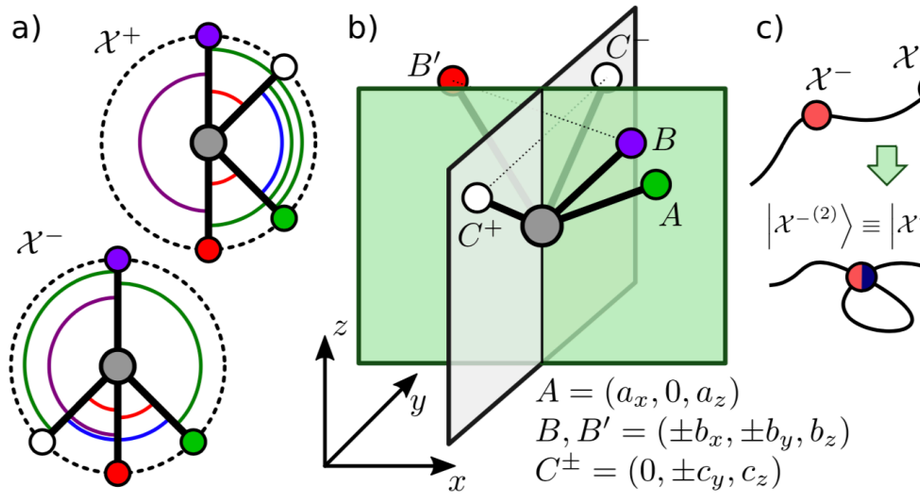


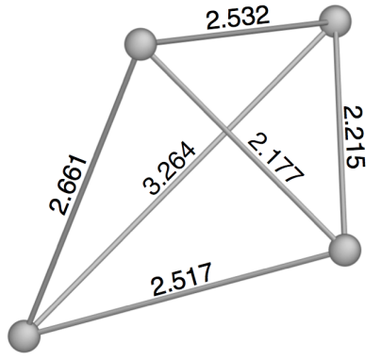


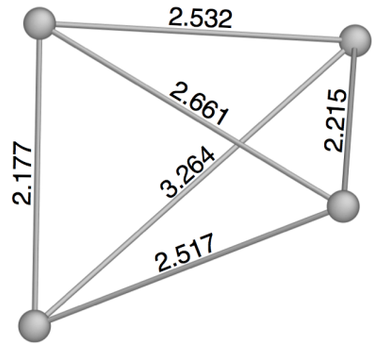


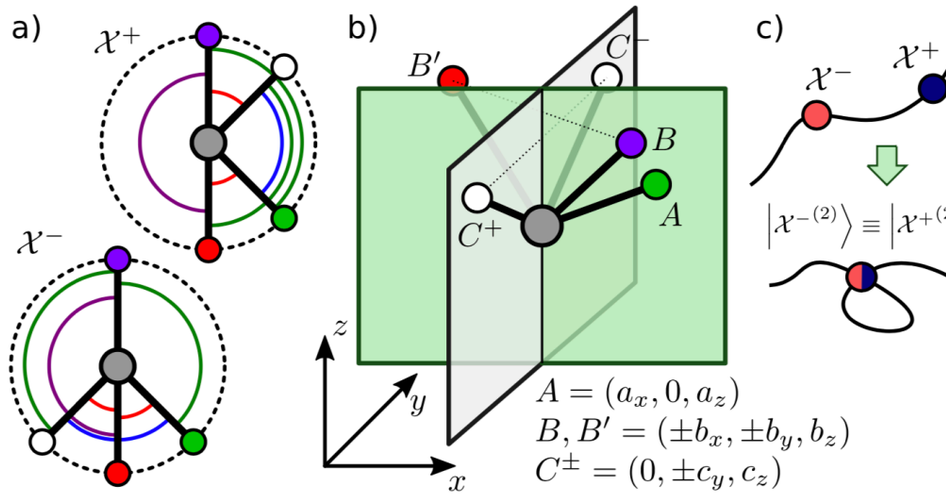
- Unordered lists of distances and angles are not enough either!

Completeness: can we reconstruct an atomic environment from the representation?

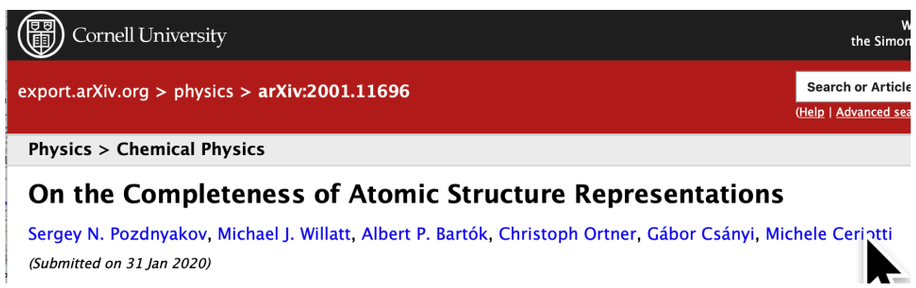
- Well known that unordered distances not enough to reconstruct a point set

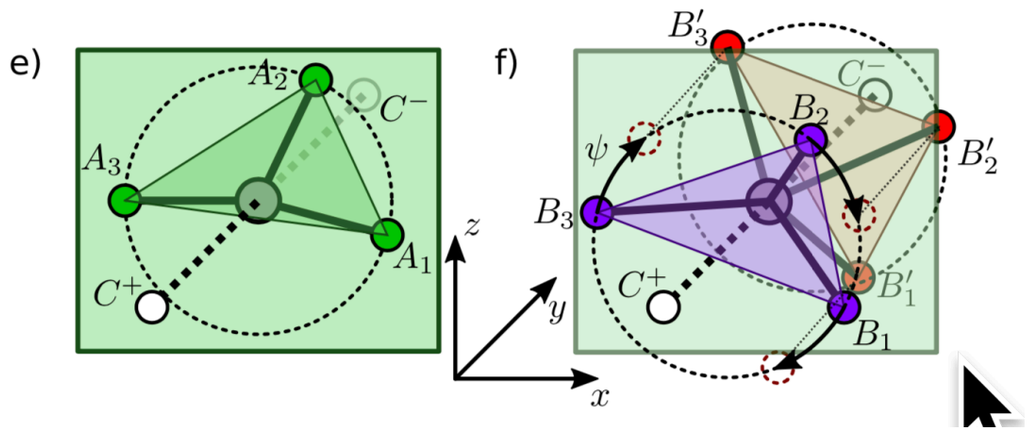




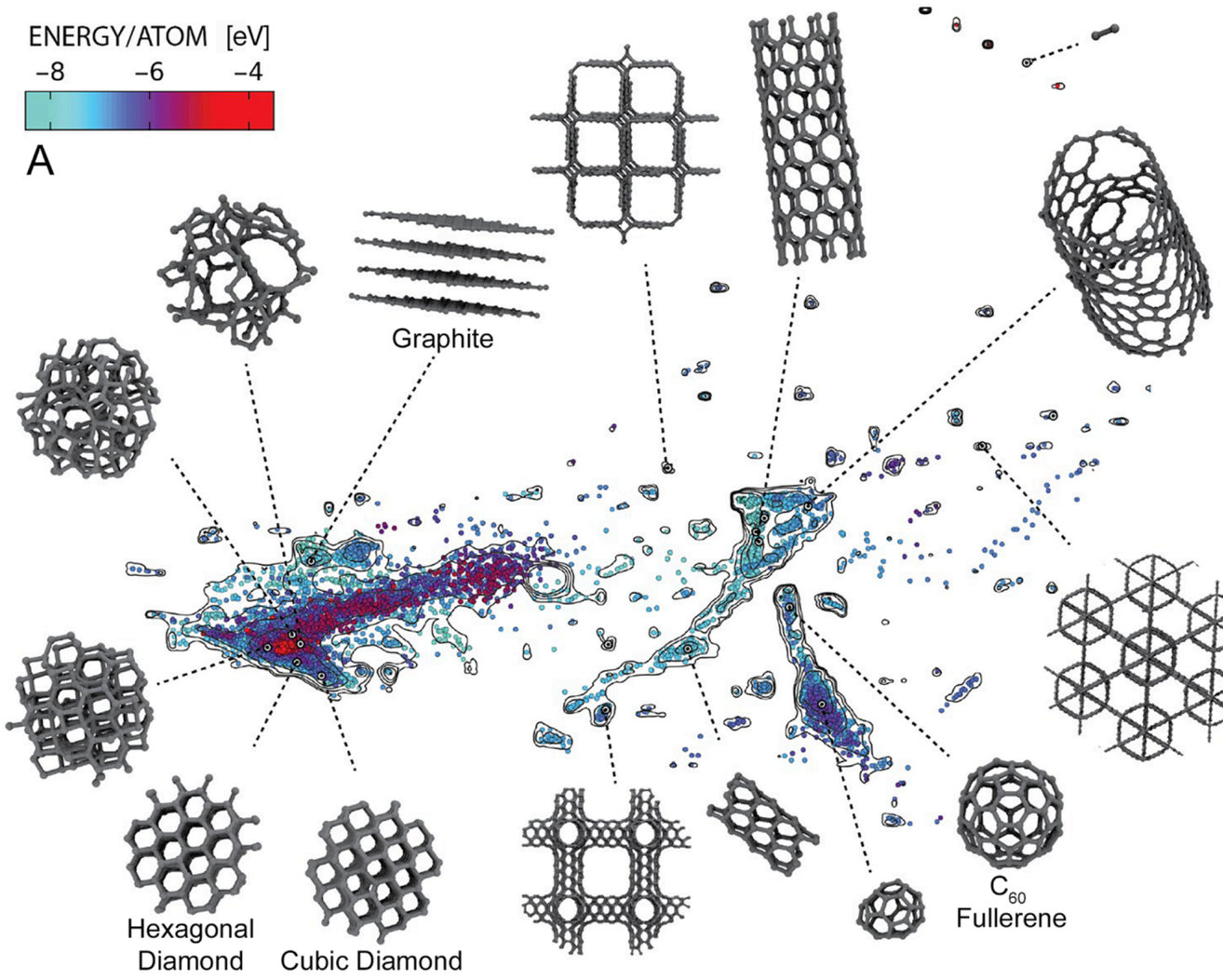


- Unordered lists of distances and angles are not enough either!





General carbon potential (2020)



arXiv.org > physics > arXiv:2006.13655

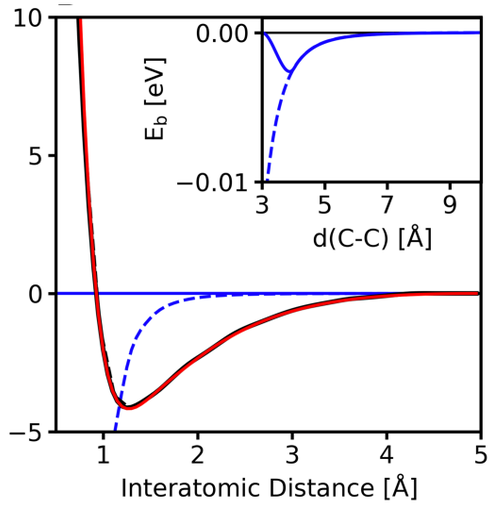
Physics > Computational Physics

[Submitted on 24 Jun 2020]

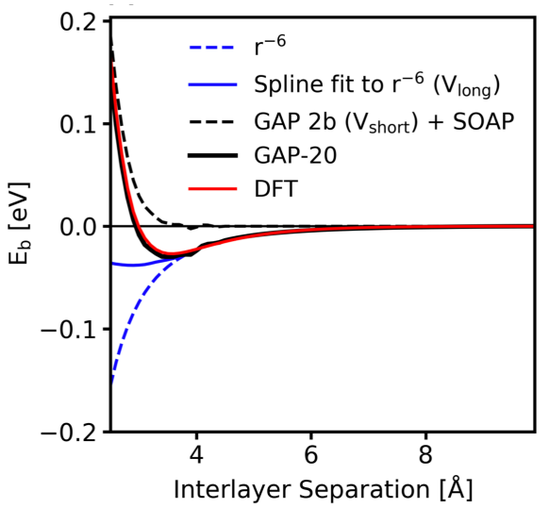
An Accurate and Transferable Machine Learning Potential for Carbon

Patrick Rowe, Volker L Deringer, Piero Gasparotto, Gábor Csányi, Angelos Michaelides

C-C dimer



Graphite layers



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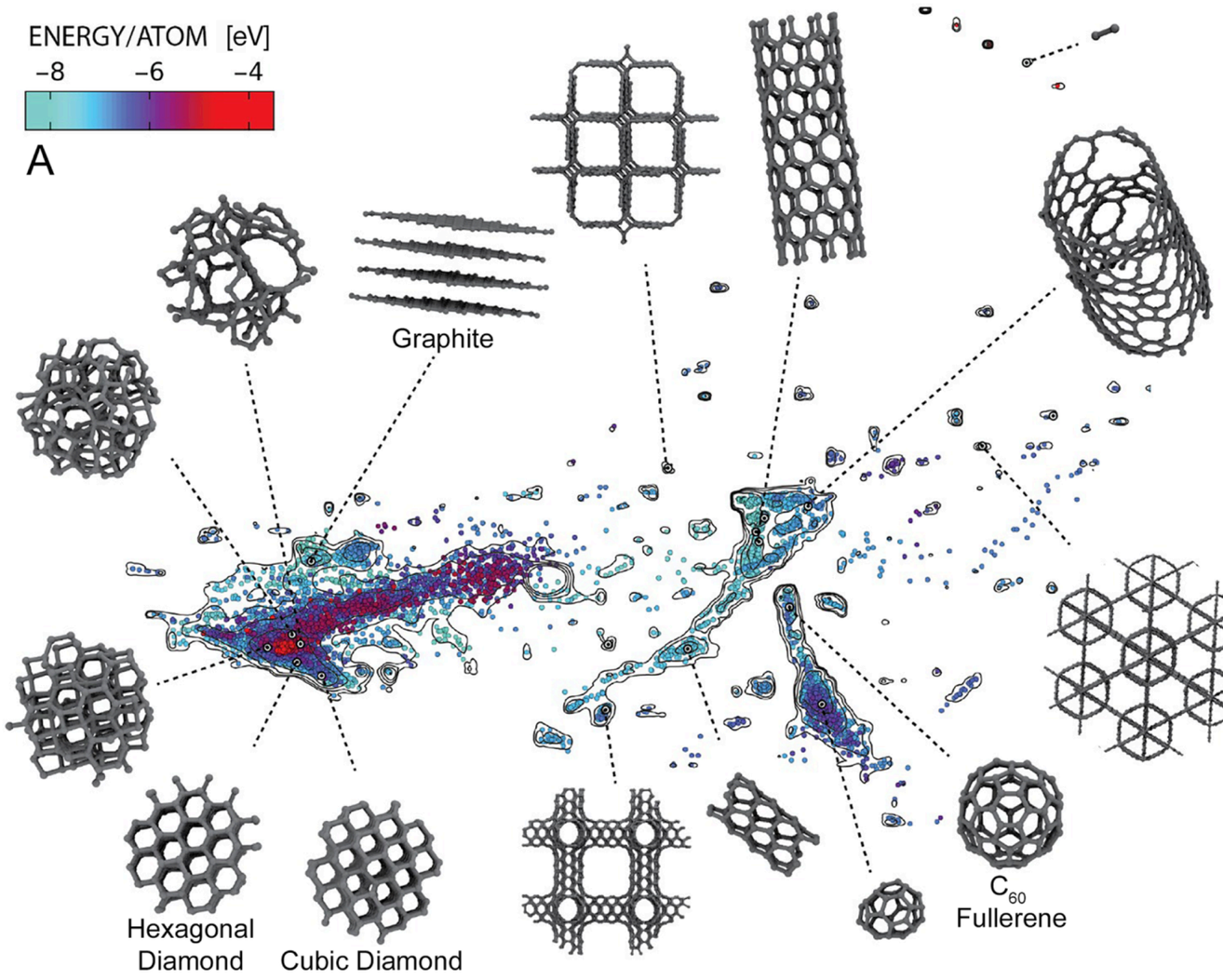

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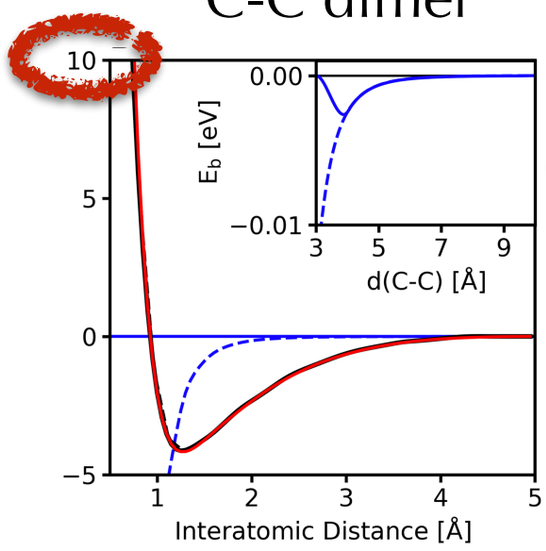

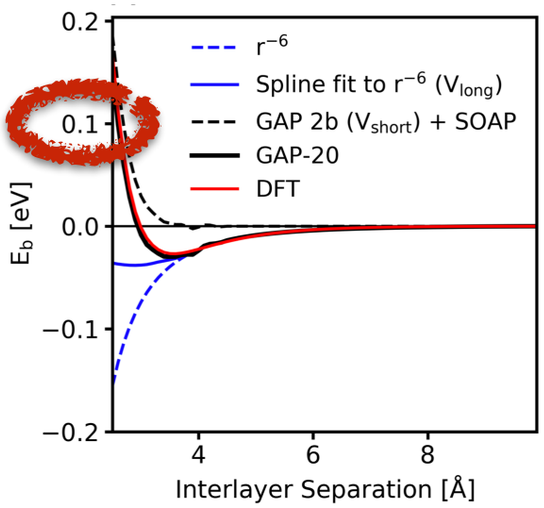

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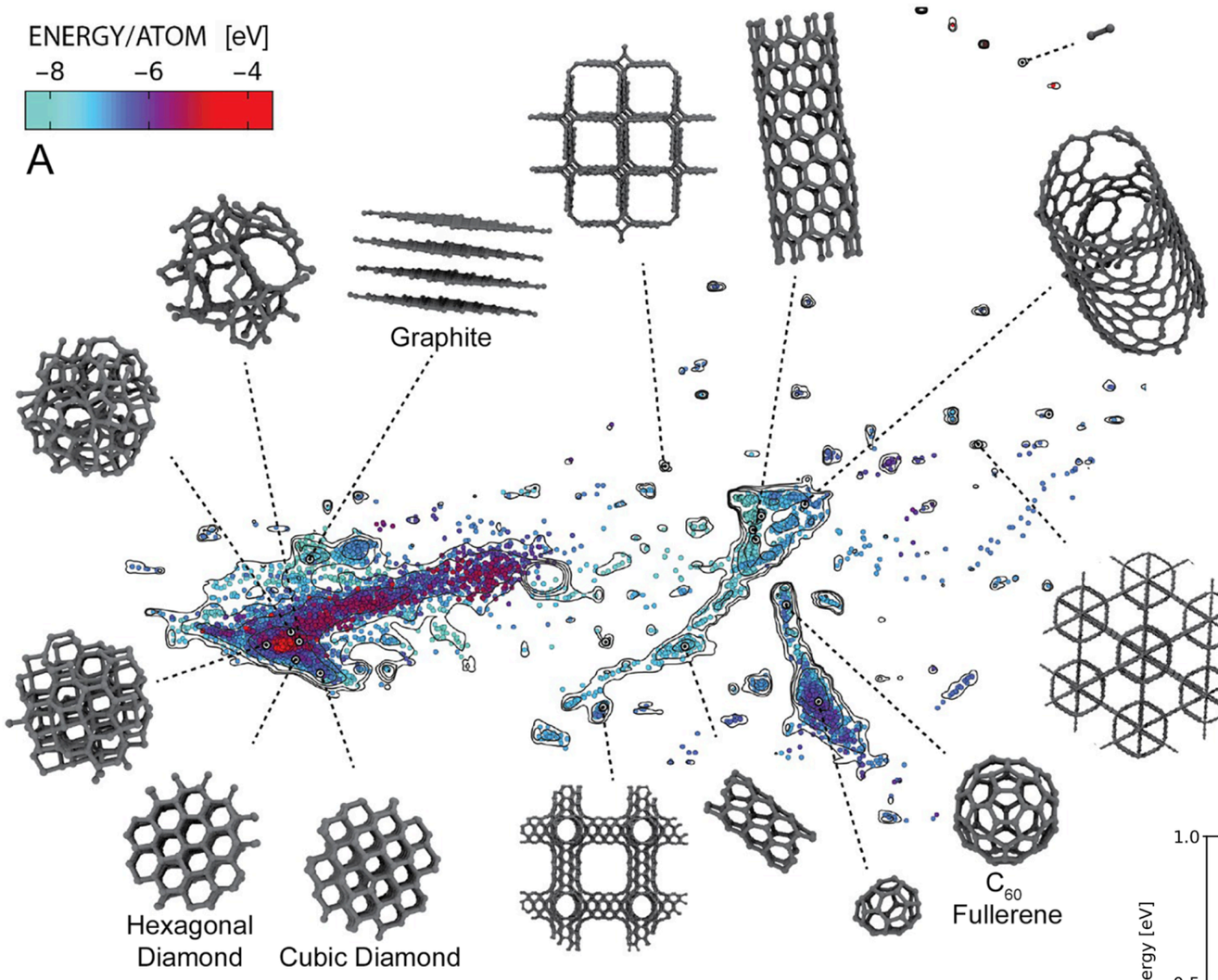

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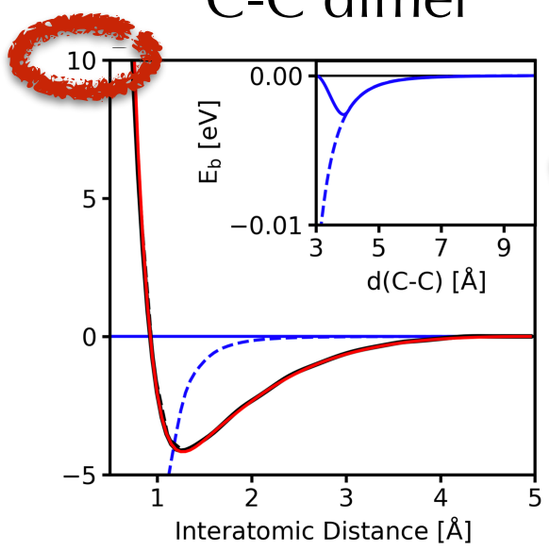
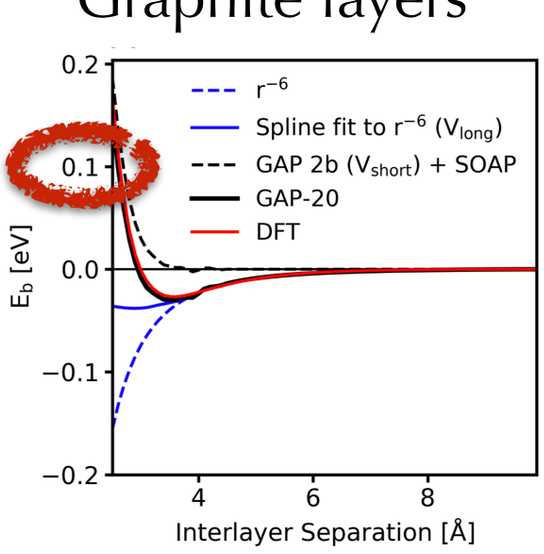
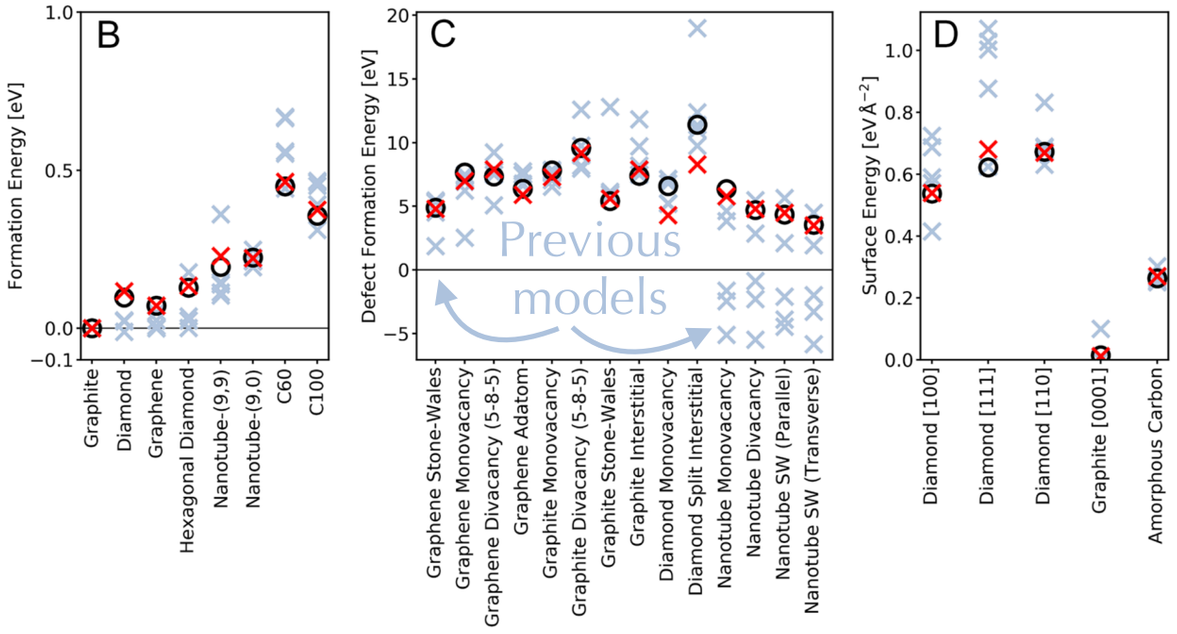






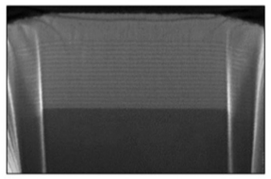
Sparse kernel model (SOAP kernel)
~ 9000 basis functions
~ 6000 oracle evaluations (~400k scalars)
> 10^6 speedup

Target ○

Model ×

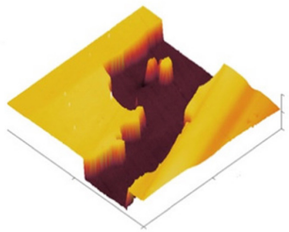
Diamond-like Carbon (DLC) coatings

Hard wearing, biocompatible

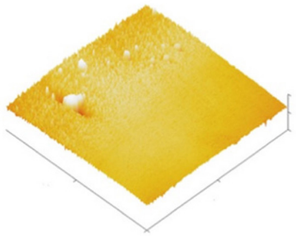


Oblique angle view of cross-sectioned DLC

Ti-6Al-4V stem and head as a single part without having a taper joint

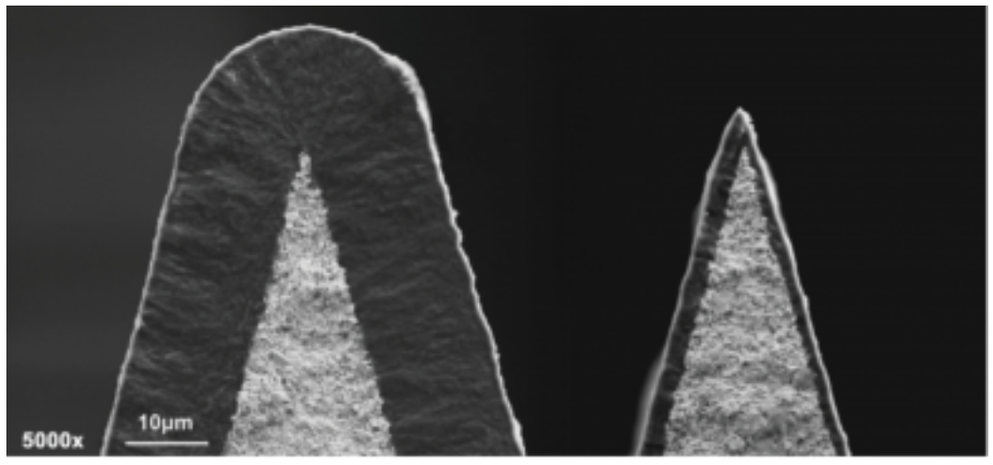


Delaminated wear track for low-DLC sample



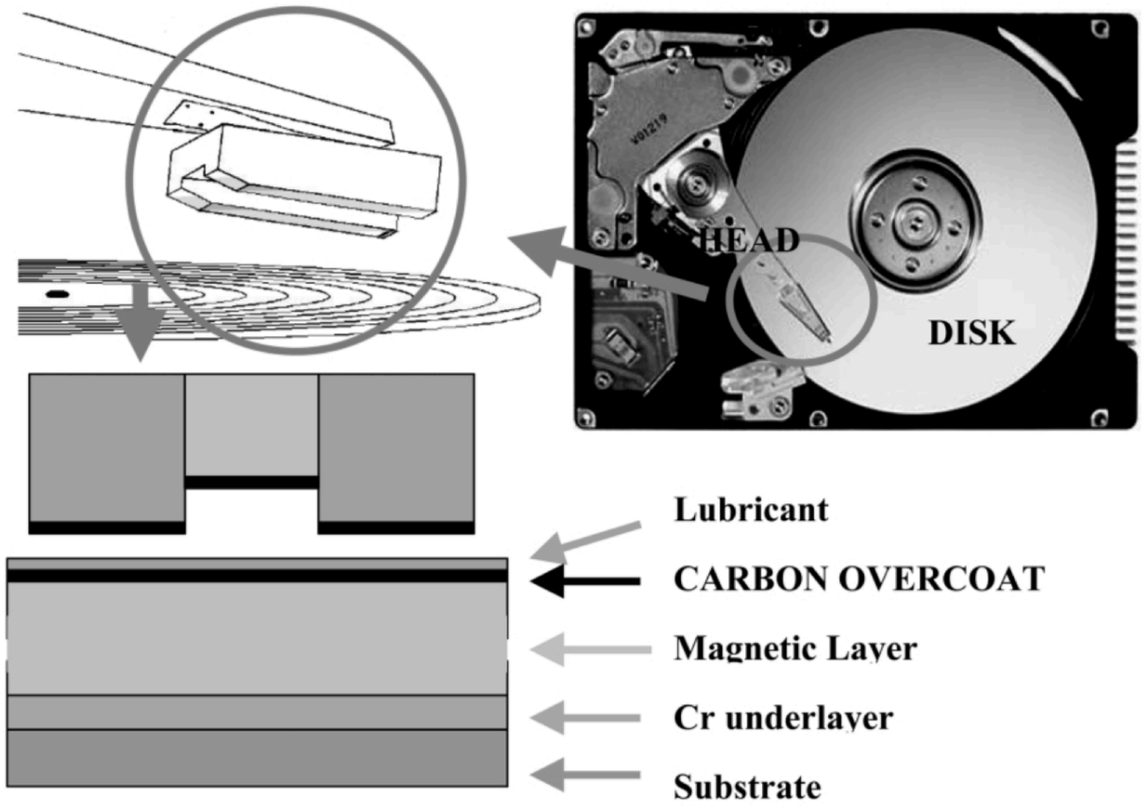
Un-delaminated wear track for High-DLC sample

Replacement hip joint



Blade with diamond coating Blade after plasma sharpening

Razor blade



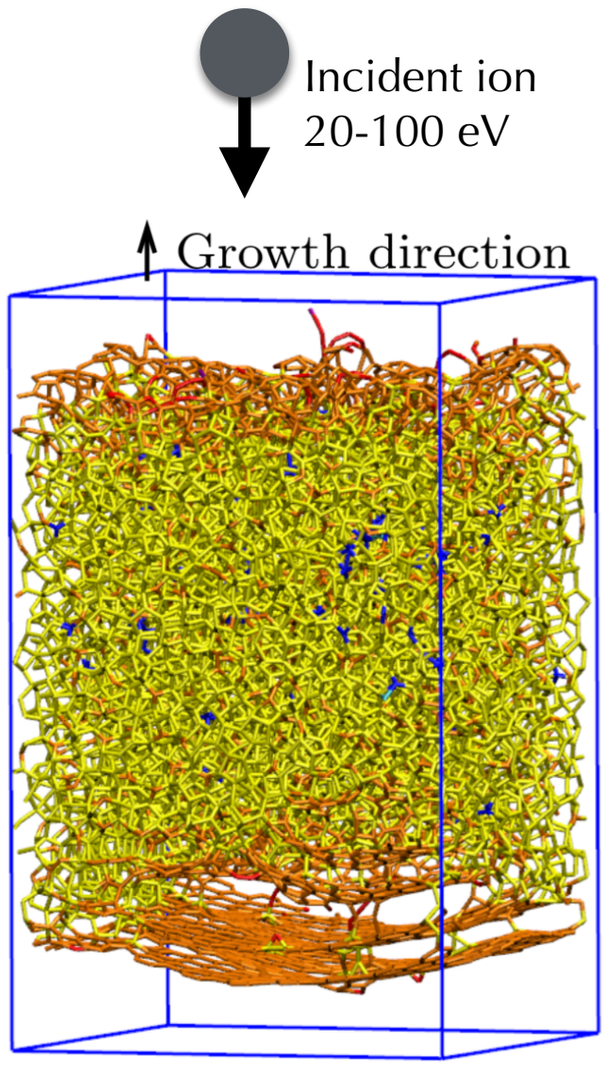
Hard disk drive

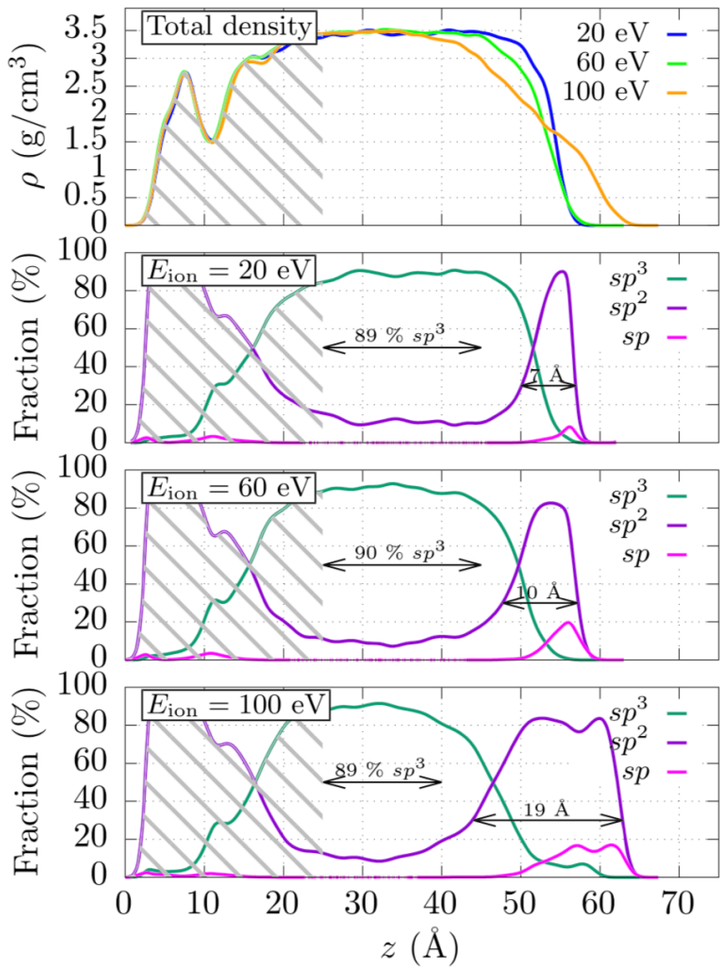
PHYSICAL REVIEW LETTERS **120,** 166101 (2018)

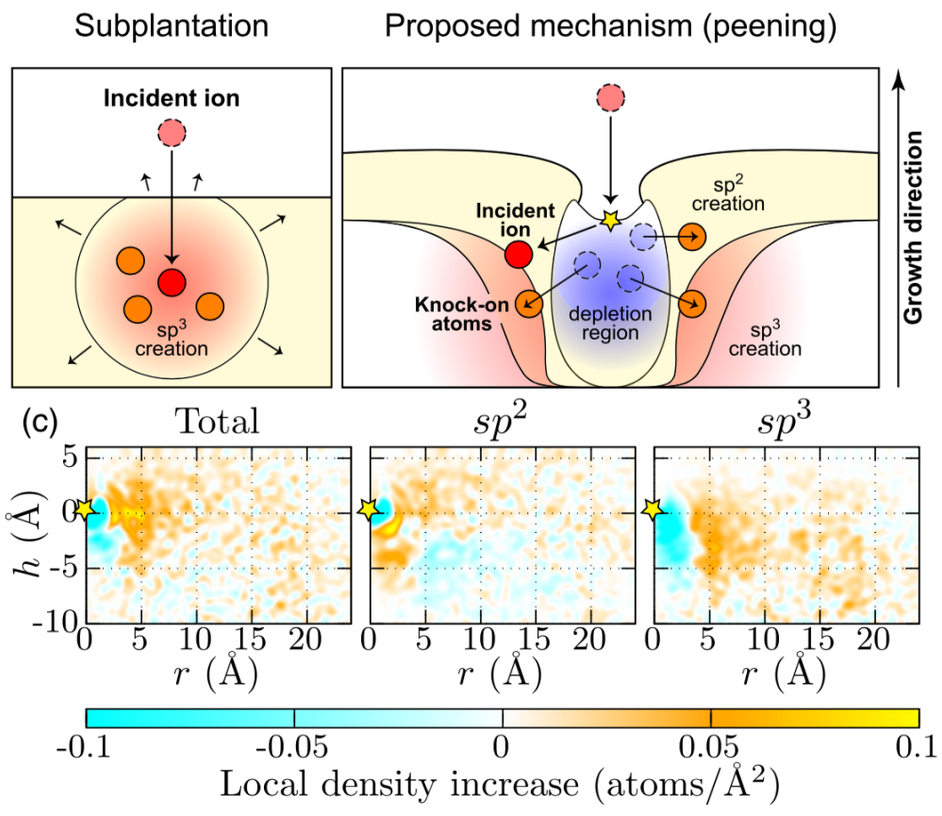
Editors' Suggestion | Featured in Physics

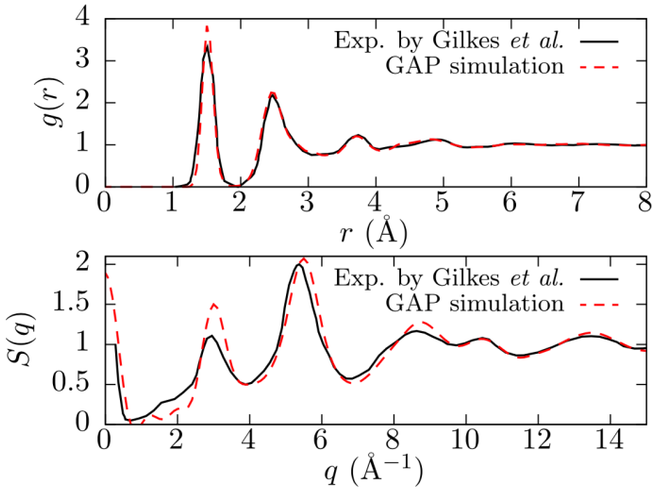
Growth Mechanism and Origin of High sp^3 Content in Tetrahedral Amorphous Carbon

Miguel A. Caro,[1,2,*] Volker L. Deringer,[3,4] Jari Koskinen,[5] Tomi Laurila,[1] and Gábor Csányi[3]

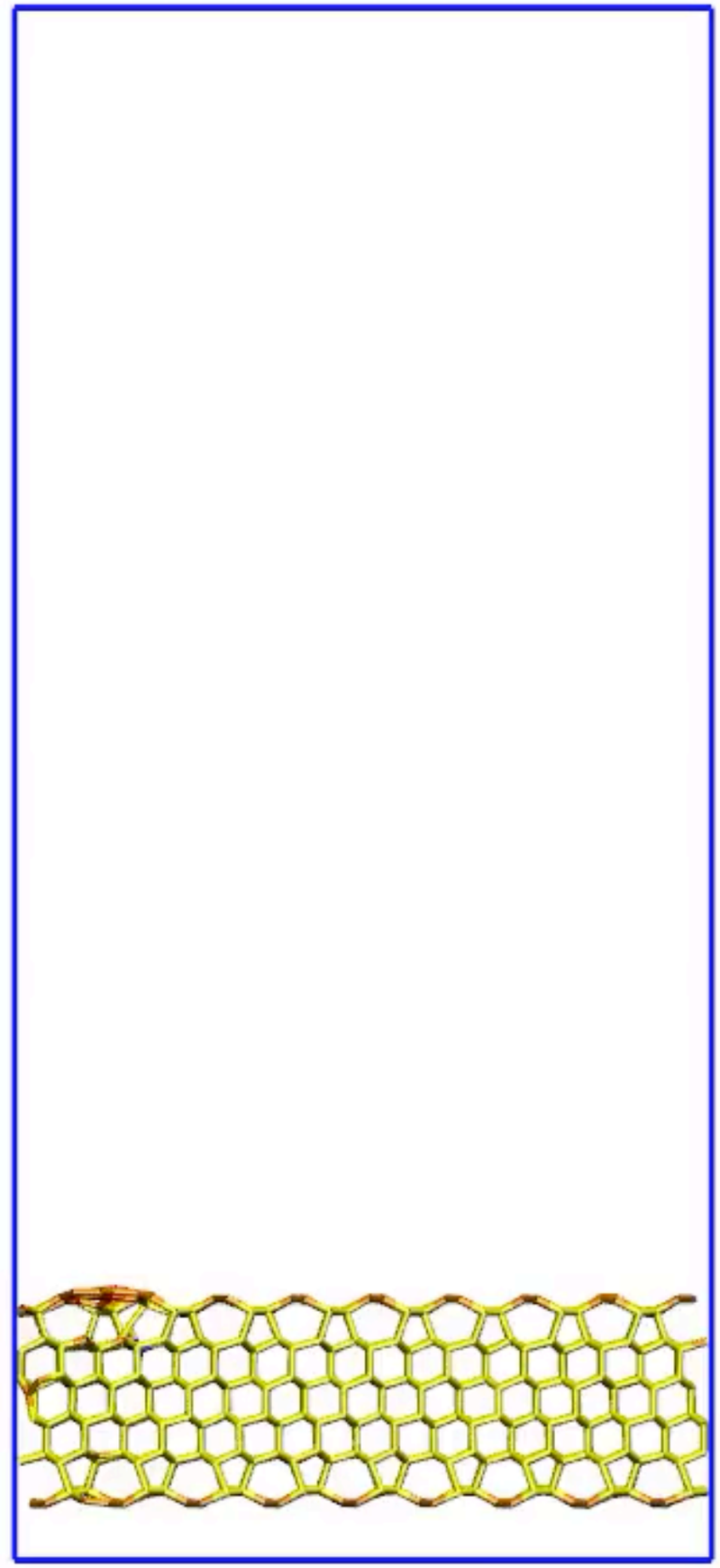


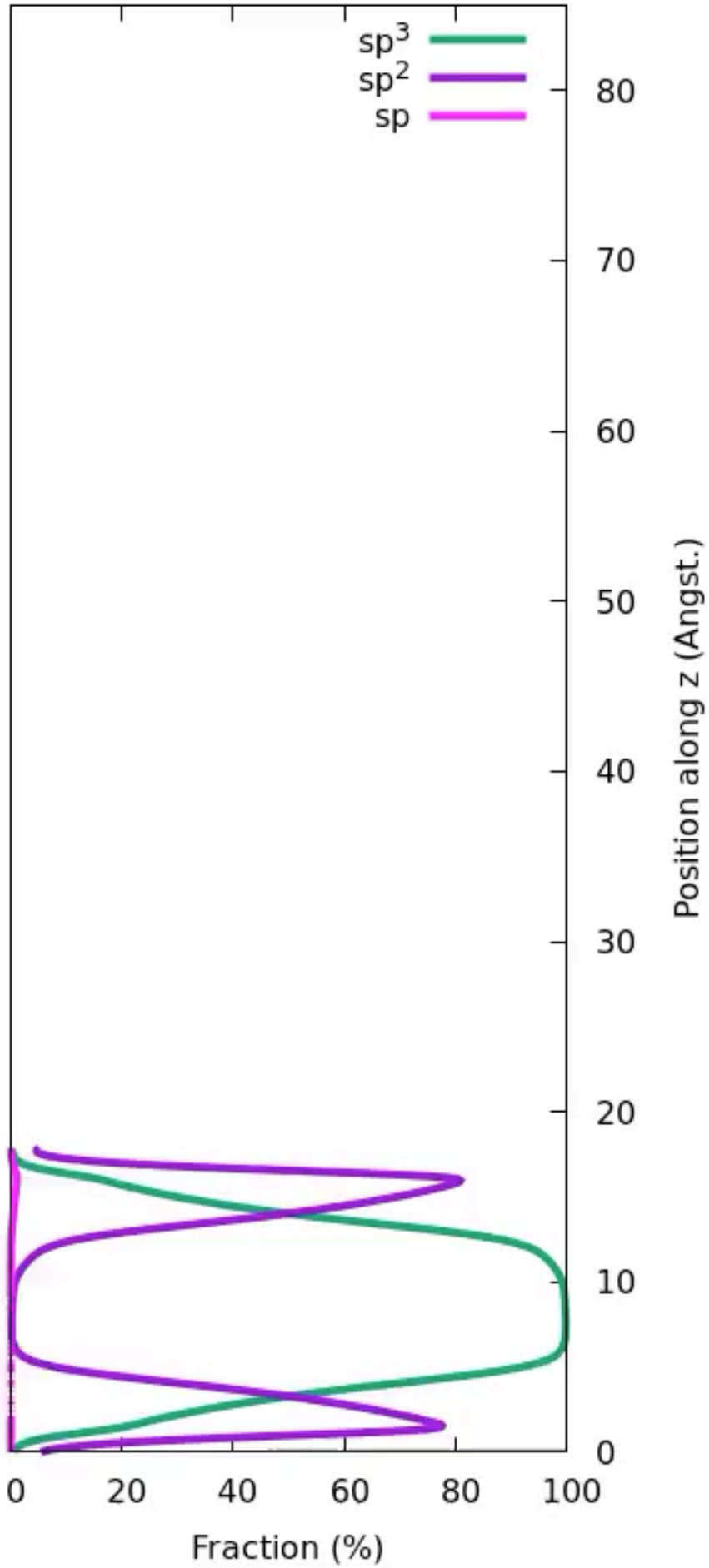


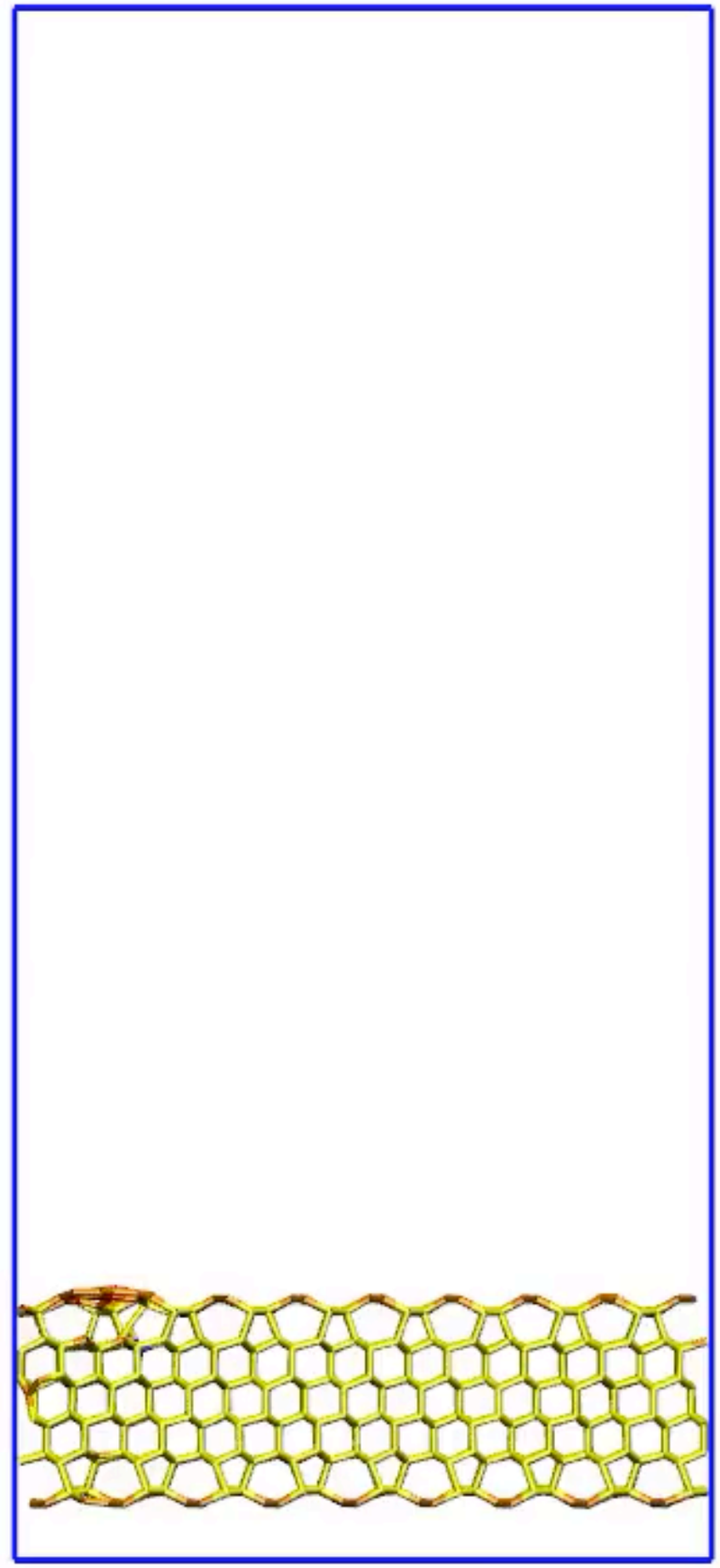




Speedup wrt. explicit electronic simulation: ~ 10^5
would have taken 30,000 years


60 eV 00001 impacts


sp3
sp2
sp
0
10
20
30
40
50
60
70
80
Position along z (Angst.)
0
20
40
60
80
100
Fraction (%)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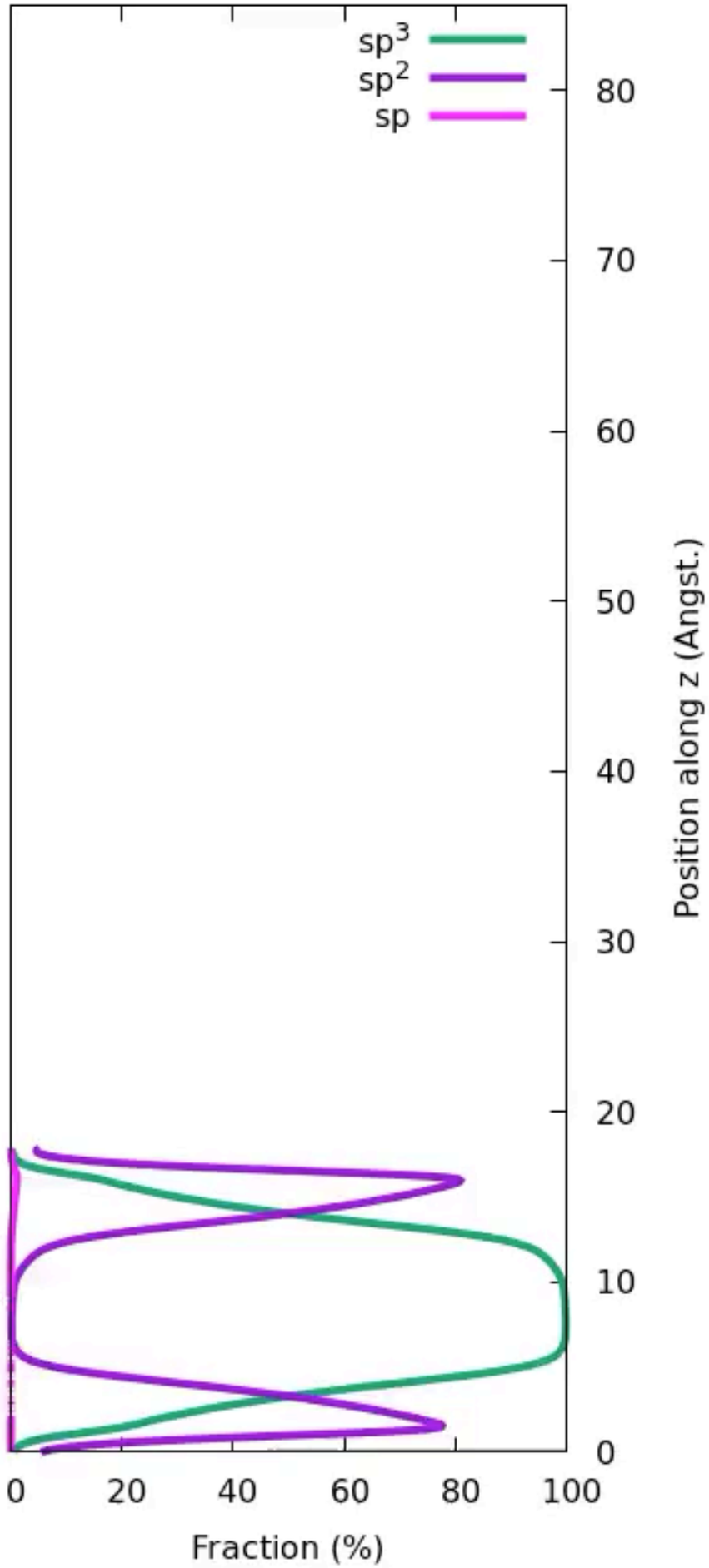
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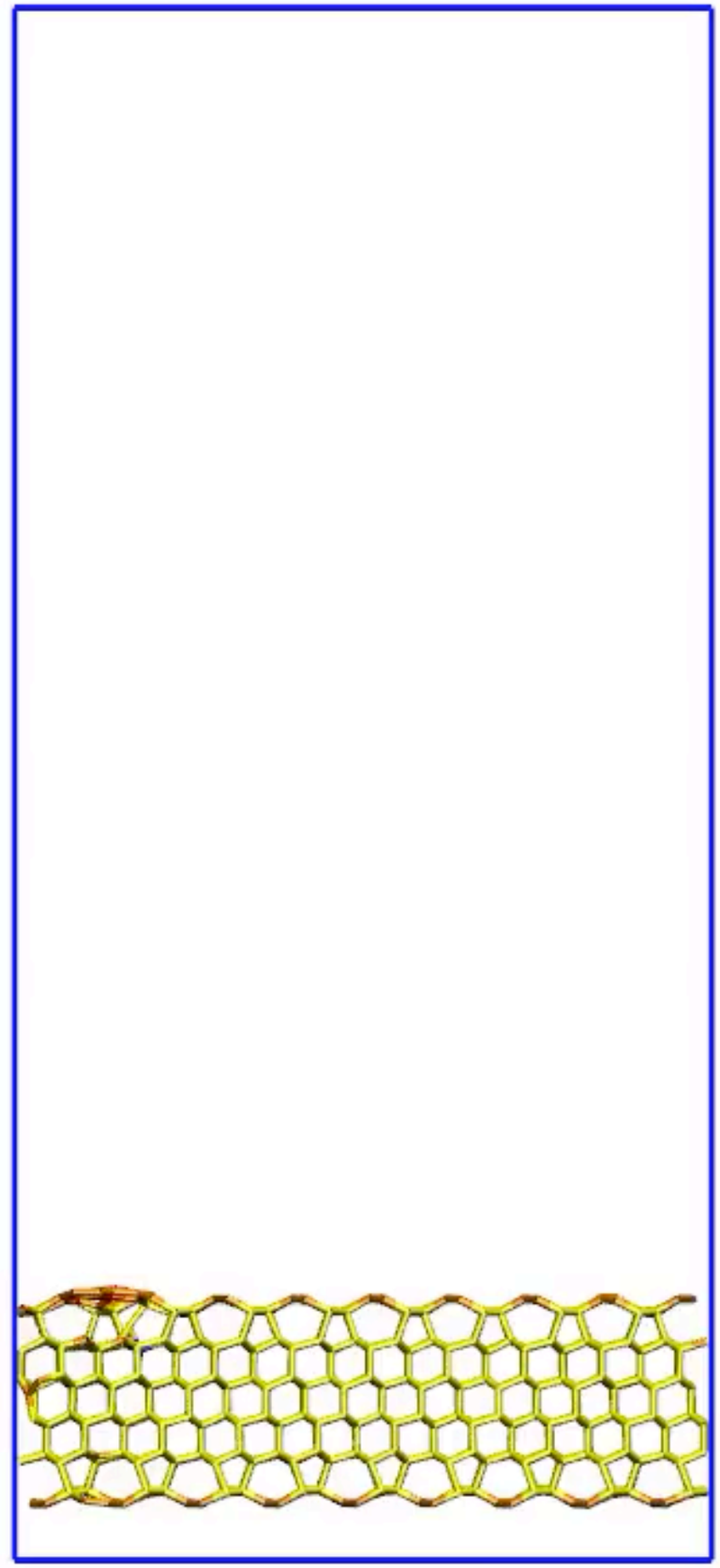
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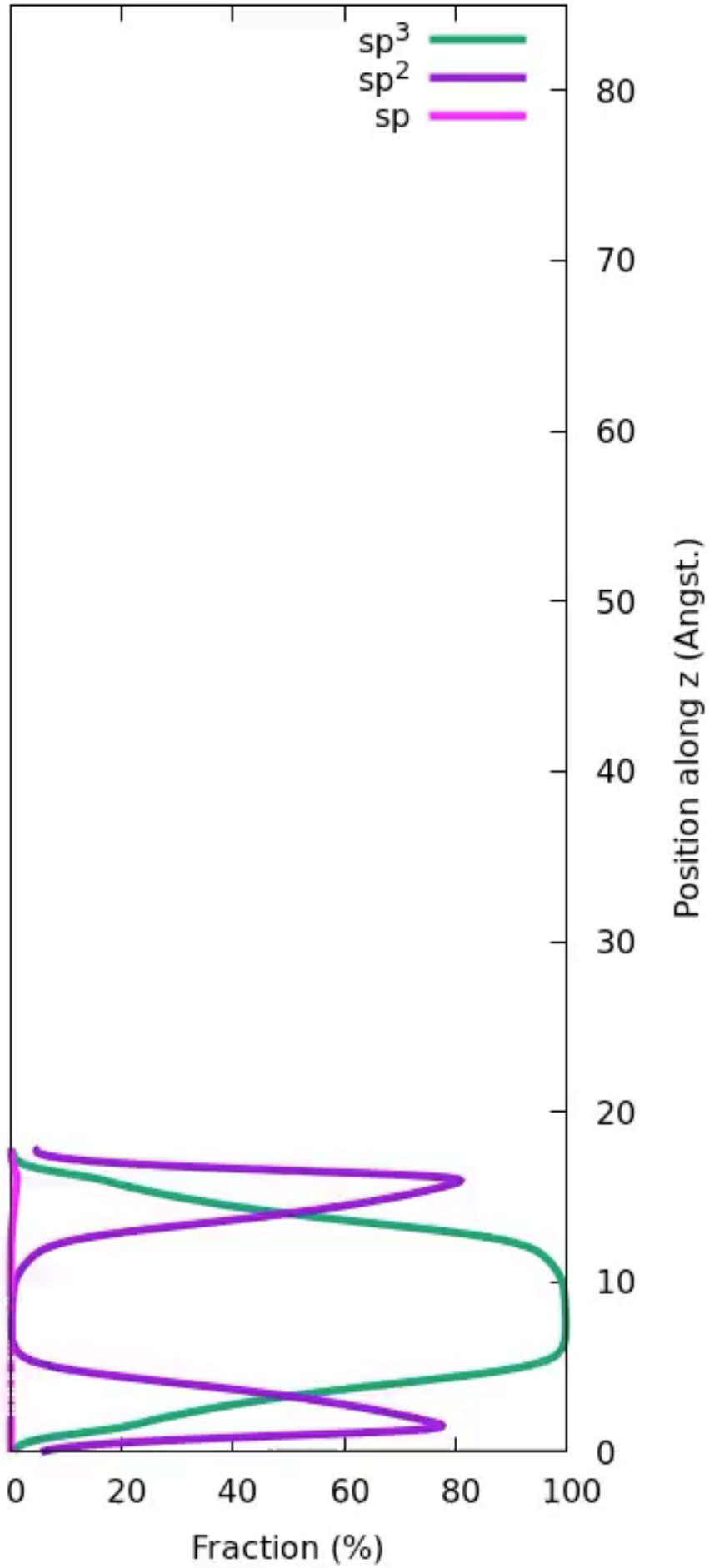
sp3
sp2
sp
0
10
20
30
40
50
60
70
80
Position along z (Angst.)
0
20
40
60
80
100
Fraction (%)

True extrapolation is still problem in high dimensional fits

MACHINE LEARNING
Science and Technology

PAPER

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OPEN ACCESS

RECEIVED
15 July 2019

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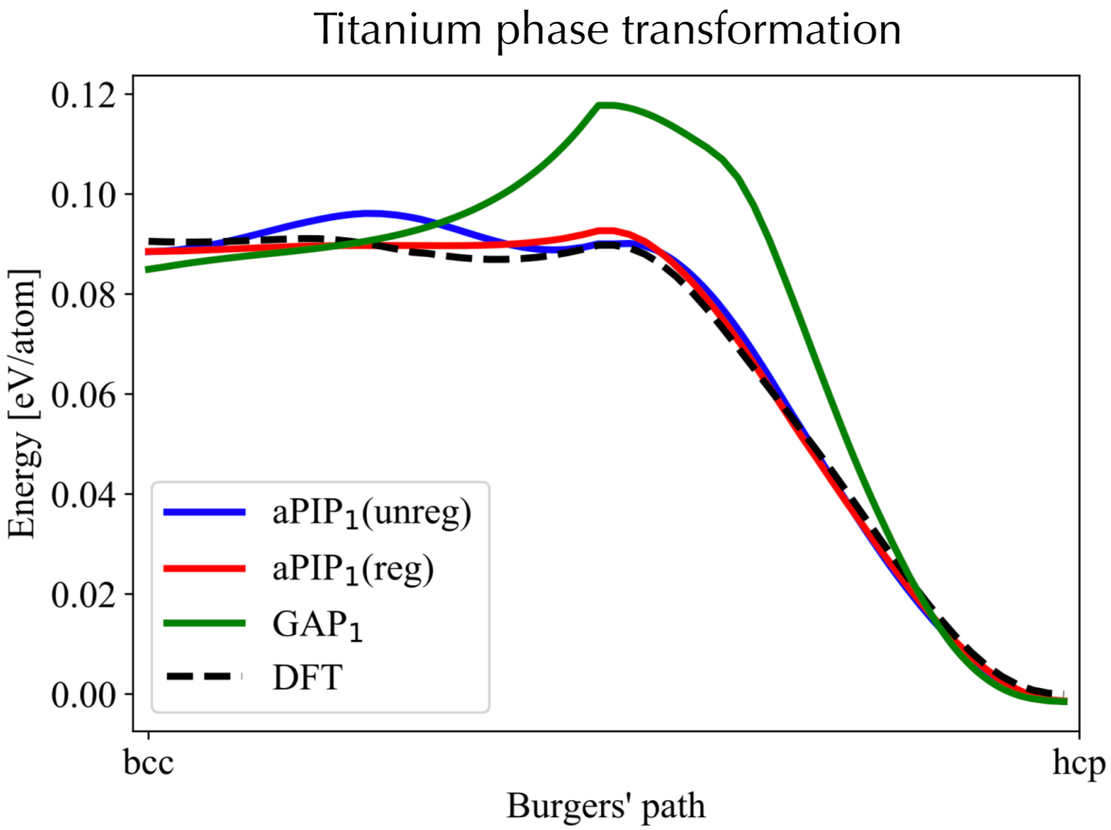
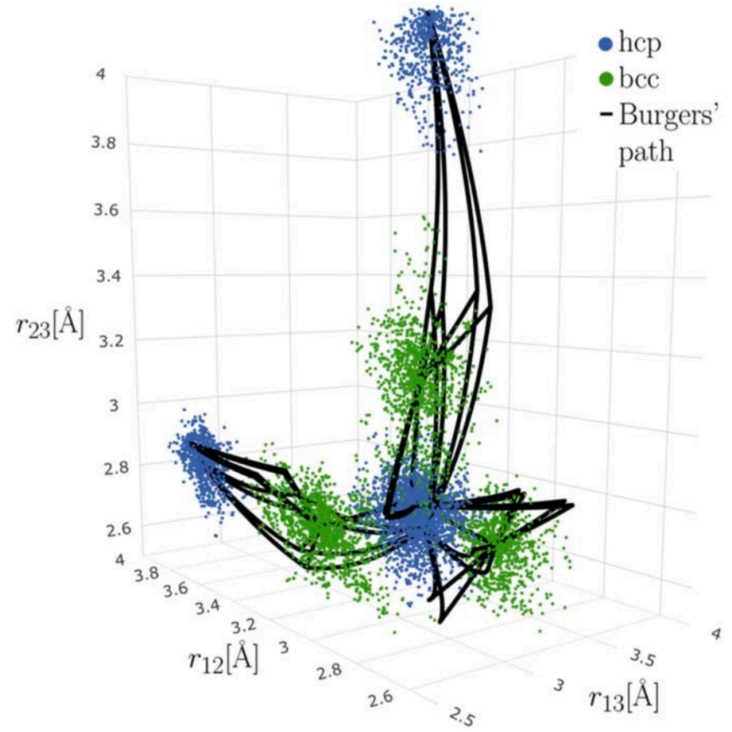
ACCEPTED FOR PUBLICATION
16 October 2019

Regularised atomic body-ordered permutation-invariant polynomials for the construction of interatomic potentials

Cas van der Oord[1,3], Geneviève Dusson[2,3], Gábor Csányi[1] and Christoph Ortner[2]

[1] Engineering Laboratory, University of Cambridge, Trumpington Street, Cambridge, CB2 1PZ, United Kingdom
[2] Mathematics Institute, University of Warwick, Coventry CV47AL, United Kingdom
[3] These authors contributed equally to the work.





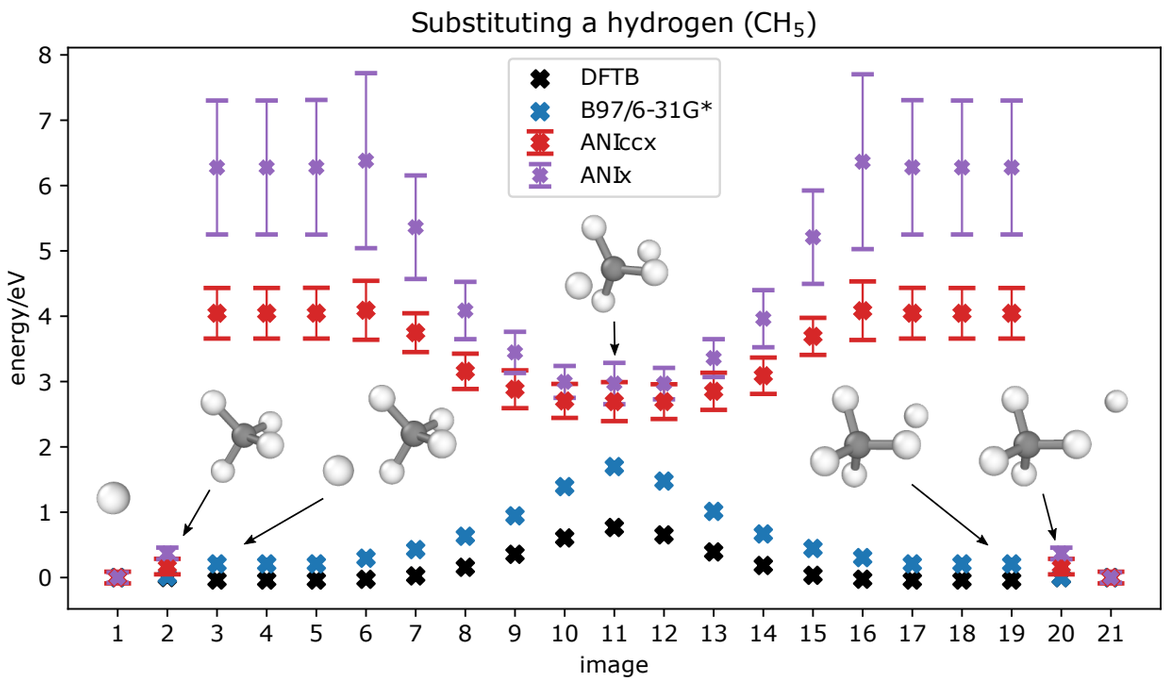


Figure: Elena Gelzinyte

Short range terms in force fields

Short range terms in force fields

Empirical FFs

all (?) classical force fields

- Atomic body-ordered expansions (bond, angle, dihedral)
- Atom types
- 1D functions (parametrised)
- *Extensive, transferable*

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ML potentials/force fields

SOAP/GAP, NNs: Jorg Behler, ANI (Isayev), DeepMD

- Many body description of atomic neighbourhood
- Descriptors are 2 and 3-body
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PIPs

permutationally invariant polynomials
(Bowman & Braams, Paesani etc)

- 3N-D functions, systematic
- Linear regression
- Fixed system (not extensive, not transferable)
- used to fit molecular body-ordered functions (monomer, dimer, etc)

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higher body order descriptors allow accurate linear fitting

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aPIPs

close to "fragment PIPs" (Qu et al)

- Atomic body-ordered expansions 1B, 2B, 3B, 4B
- 0D, 1D, 3D, 6D
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- Spatial cutoffs
- Introduce atomic body-order in polynomials

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- Free up functional forms (use polynomials)
- *Make "atom types" continuous variables*

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- Basis sets equivalent
- Different scaling with
  #neighbours
  #atomic species

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Ideas for long range terms

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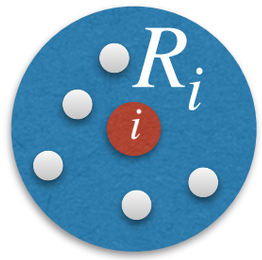
1. Do nothing *(always best if it works)*

$$E = \sum_i E_i(R_i)$$



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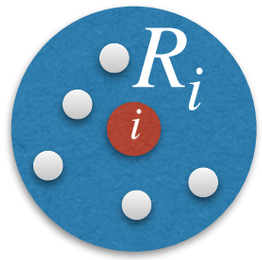
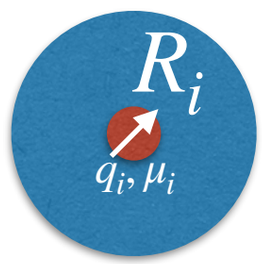


2. *Fixed* long range baseline model
TTM4 electrostatics (e.g. Paesani et al 2012+), 1/r^6 dispersion (Veit, 2019), many others

$$E = E_{\text{long-range}}(q_i, \mu_i) + \sum_i E_i(R_i) \qquad q_i, \mu_i \text{ fixed}$$



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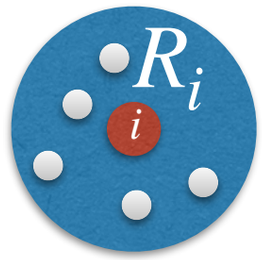

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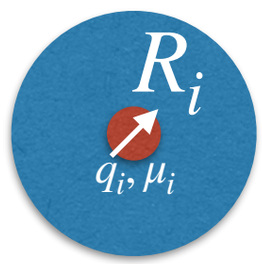

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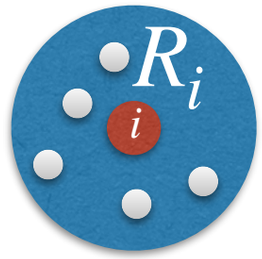

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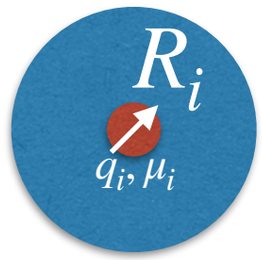

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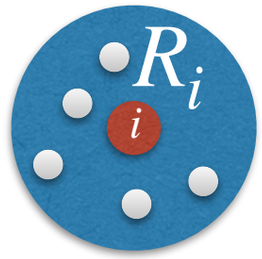

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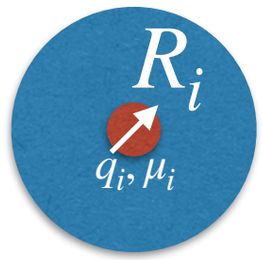

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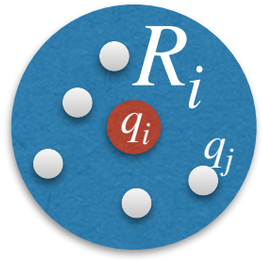

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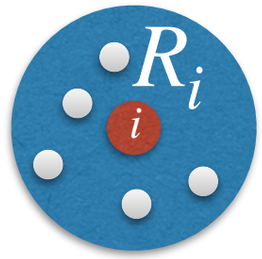

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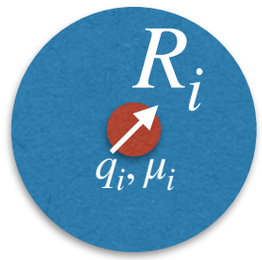

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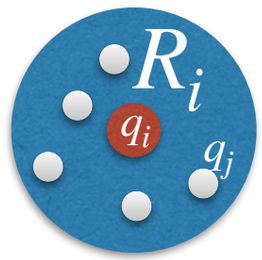
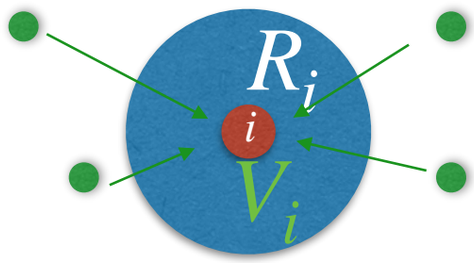


6. Multiscale descriptors: LODE (Grisafi, Ceriotti)

$$E = \sum_i E(R_i, V_i) \qquad V_i : \text{long-range field descriptor}$$



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- Synergy between force field ideas and other molecular problems: ??
  - Generative models, unsupervised learning problems, large scale classifications
  - Effective Hamiltonians and parametrisation of other operators
  - *Eigenfunctions* (wave functions) rather than just eigenvalues