Recent approaches to machine learning of interatomic potentials seen from a perspective of plasma material interaction and primary radiation damage

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Hydrogen retention in irradiated tungsten

IAEA Coordinated Research Project (CRP) on Plasma-wall interaction with irradiated tungsten and tungsten alloys in fusion devices (2013-2018). See https://www-amdis.iaea.org/CRP/.

Need to understand effect of radiation om microstructure and effect of microstructure on hydrogen retention and migration.

Must use surrogate irradiation; need modelling to interpret experimental data.

Most basic computations: primary radiation damage and hydrogen migration.

Relatively short timescale. (Long timescale: segregation, corrosion.) Molecular dynamics is the main tool.

Problems with potentials for tungsten

Talk by A. Sand (Helsinki, with Kai Nordlund) at IAEA, 2017-11-16: "Energetic cascades in tungsten: sensitivity to interatomic potentials and electronic effects."

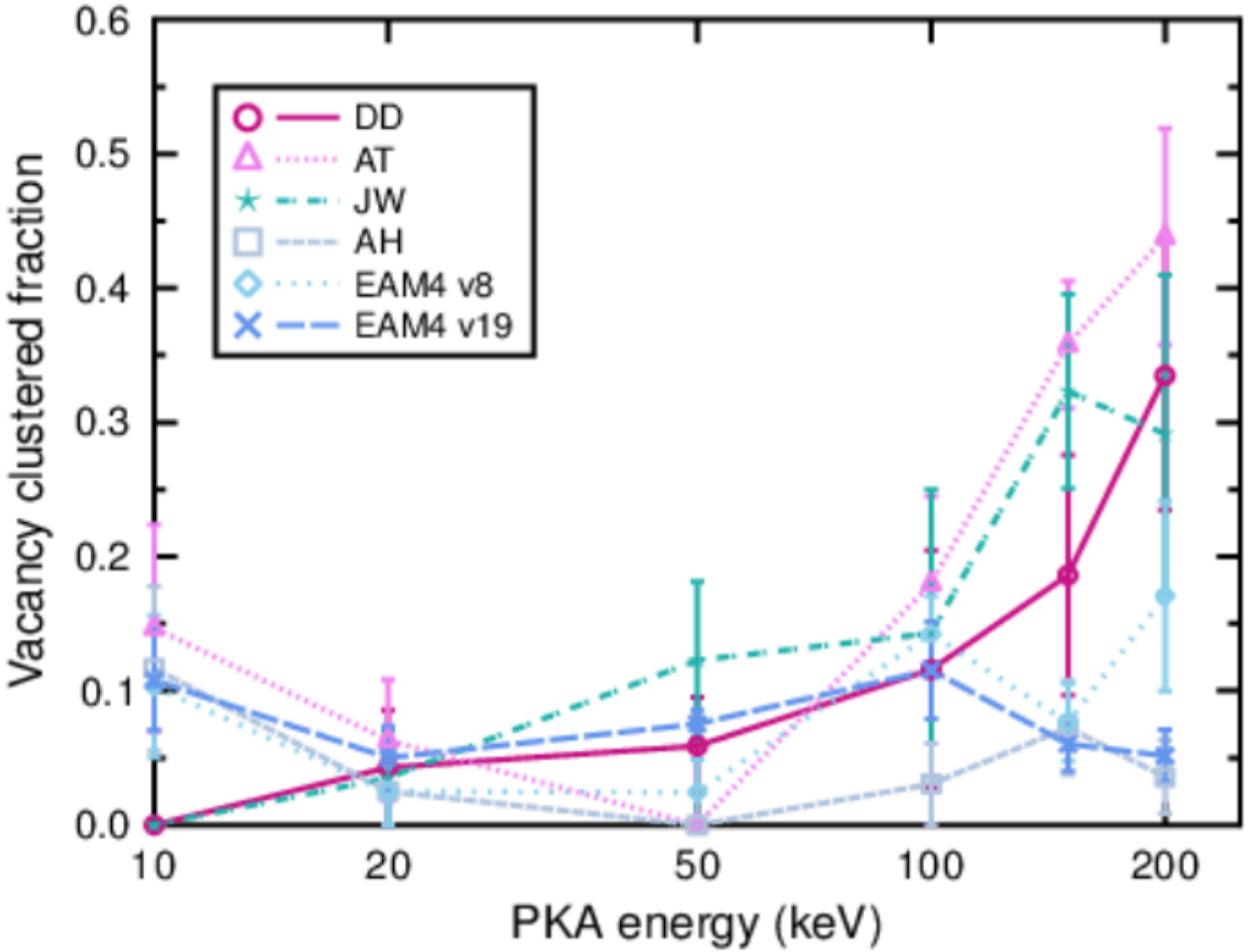
"*Potentials with largely similar point defect formation and migration energies disagree regarding clustered fraction of defects for high PKA energies. Some potentials predict only very small clusters, others show formation of clusters of* > 100 *point defects.*"

"*Why the different predictions, despite extensively fitted 'good' potentials??*"

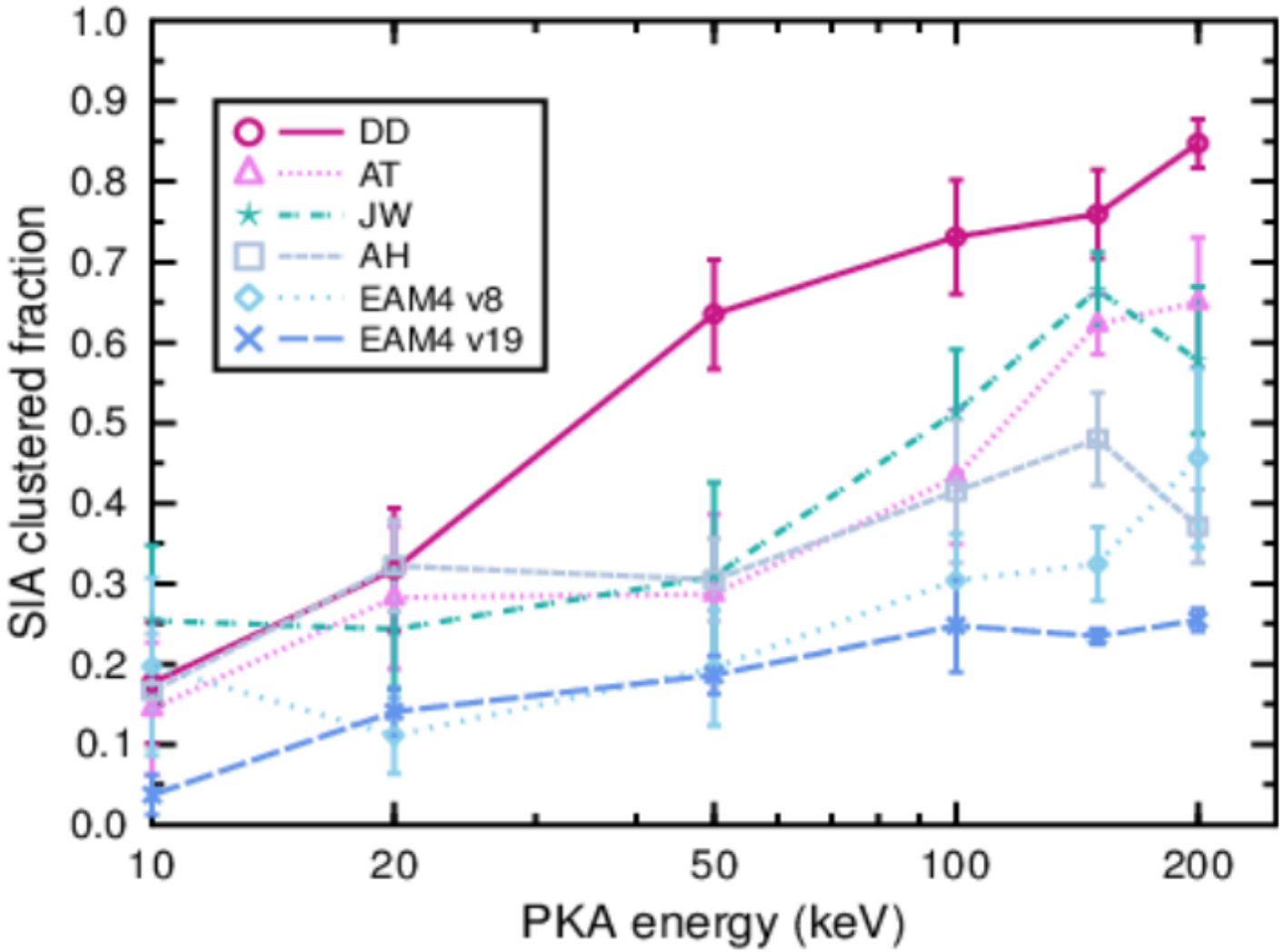
Discuss blending to short-range Ziegler-Biersack-Littmark (ZBL) potential.

Many-body effects beyond embedded atom (EAM) approach.

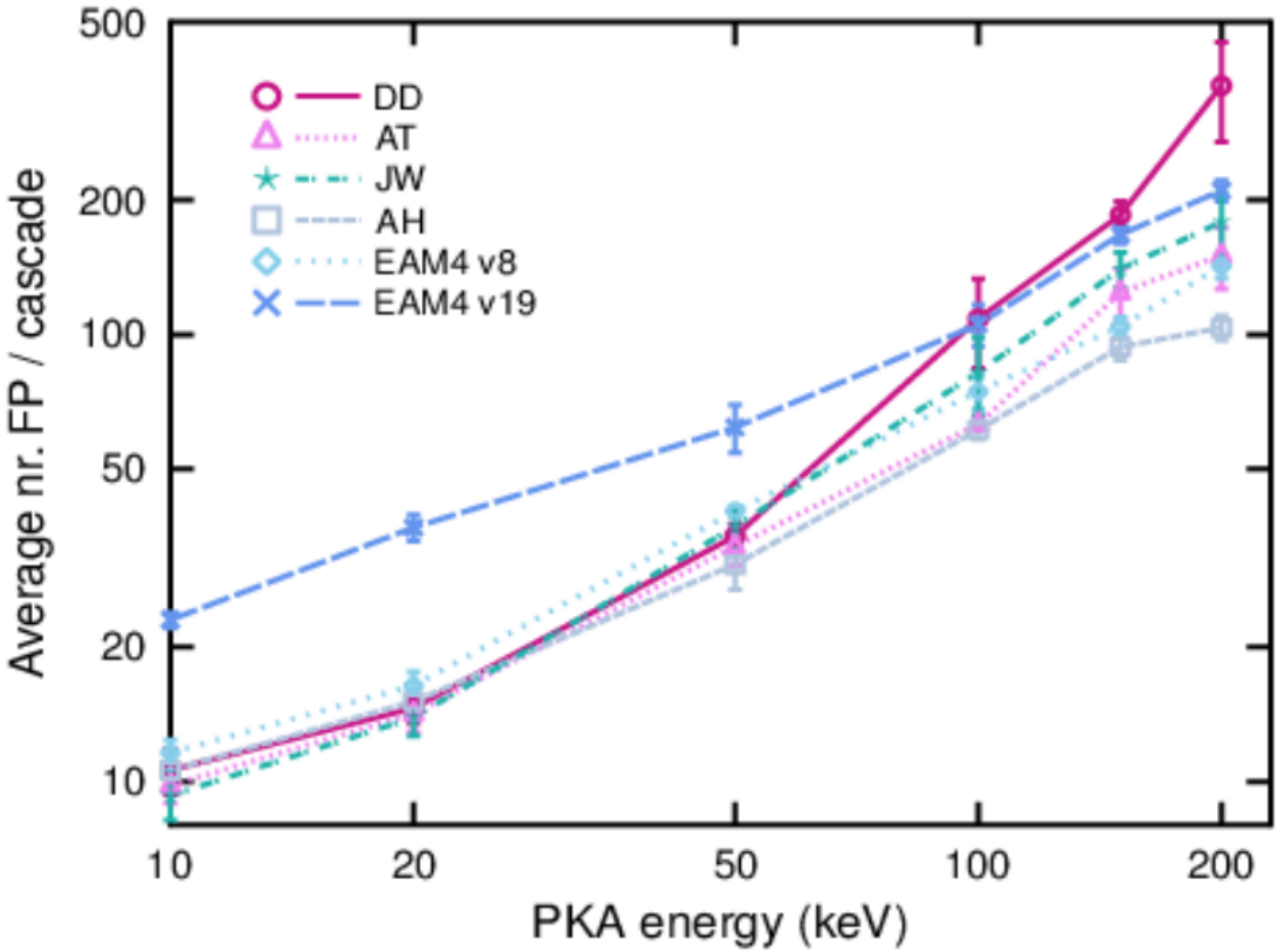
Vacancy Clustered Fraction



Self-Interstitial Atom Clustered Fraction



Average Number Frenkel Pairs per Cascade



Force fields for fusion materials and plasma-material interaction

The potential is (almost) everything; and that needs to be reflected in the effort.

Keep in mind the following target application: Primary radiation damage in W-H-He. PKA event, melt region, resolidification. More difficult than pure W (see above); not as difficult as steel.

Quantum effects on the nuclear motion: barely ever relevant.

Electronic excitation beyond simple stopping: can be important, could be taken into account. (Langevin approach, potential depends on electron temperature.)

Molecular dynamics: "F=ma"

N atoms; classical nuclei, positions $x(i)$, $1 \leq i \leq N$.

Interaction potential $V(X)$ ($X \in \mathbf{R}^{3N}$).

Force $F = -\nabla V$.

$$\frac{d^2x(i)}{dt^2} = -\frac{1}{m_i}\frac{\partial V}{\partial x(i)}$$

Electron dynamics is gone. This is the Born-Oppenheimer (adiabatic) approximation.

For large molecules and condensed phase a local representation may be used: $V = \sum_i V_0(X_i)$ where X_i are the collective nuclear coordinates for a local environment of the i-th atom.

Other uses of potential energy surfaces

Born-Oppenheimer approximation allows quantum nuclei; it is not limited to semiclassical molecular dynamics.

Molecular spectroscopy: Eigenvalue problem $H\Psi = E\Psi$ for the nuclear wavefunction. Tractable for small molecules.

Diffusion Monte Carlo for the ground state nuclear wavefunction: Random walk with birth and death processes.

Quantum statistics: $< A >_\beta = \frac{1}{Z(\beta)} \mathrm{tr}(Ae^{-\beta H})$. Averages calculated using Path Integral Monte Carlo.

Ring Polymer Molecular Dynamics and variants; PIMC plus time evolution. Model for nuclear quantum effects.

Quantum scattering: $i\hbar \frac{\partial}{\partial t} \Psi(X,t) = H\Psi(X,t)$. Application to reaction dynamics is pretty much limited to 4-atom systems.

New Approaches from Big Data and Machine Learning

Machine Learning has brought specific methods, e.g. deep convolutional neural networks (Vision, Go).

Machine Learning is also bringing a change of attitude...

Nothing wrong with optimizing over very many variables (Stochastic Gradient Descent).

Nothing wrong with lots of local minima, even inequivalent ones. Don't ask for a guaranteed global optimum.

(NN with 20 layers and 256 nodes per layer and a ReLU nonlinearity has multiplicity of about 10^{10139}, being $(256!)^{20}$; with tanh nonlinearity about 10^{11680}, being $(2^{256} \times 256!)^{20}$.)

Fitting or learning in the presence of symmetries

Example: Want to fit or learn $f : \mathbf{R}^N \to \mathbf{R}$, $f(x) = z$, using data $f(X_\alpha) = z_\alpha$. Typical point $x = (x_1, ..., x_N)$. Say that the underlying true function is totally symmetric in the $(x_i)_i$. Options:

(a) Ignore the symmetry, use any plausible model. (Maybe replicate the data using symmetry.) Obtain symmetry via accuracy.

(b) Use explicit invariants of a good functional form. Example: $y_k = p_k(x)$ (where the p_k are elementary symmetric polynomials for $1 \leq k \leq N$), then $f(x) = g(y(x))$ with some plausible model for g. Efficient; technically difficult for more complicated symmetries. (Braams+Bowman at Emory University.)

(c) Use explicit invariants of an easily generalizable form. Example: $y = \mathrm{Sort}(x)$, then $f(x) = g(y(x))$. Introduces nonsmoothness, often discontinuities. Obtain smoothness via accuracy.

GAP-SOAP approach of G. Csányi, B. Bartók et al.

Key reference: Bartók, Kondor, Csányi (2013) Phys Rev B 87.

Gaussian Approximation Potential (GAP), also referred to as Kernel Ridge Regression, high-dimensional version of Radial Basis Functions. Authors use both language of Machine Learning and language of function fitting, regression analysis.
$f(X) = \sum_\alpha w_\alpha K(X, X_\alpha)$.

Smooth Overlap of Atomic Potentials (SOAP) kernel $K(X, X')$.

$X \mapsto \rho$, $S(\rho, \rho') = \int \rho(r)\rho'(r)dr$.

$K(X, X') = \int |S(\rho, R.\rho')|^n \, dR$, $R \in O(3)$.

Integrals evaluated via spherical harmonic expansion.

SchNet, Deep Tensor network from TU Berlin

Key reference: Schütt, Sauceda, Kindermans, Tkatchenko, Müller (2018) J Chem Phys 148. Also Nature (2017).

Say N atoms. Each NN layer contains atomic feature vectors x_i for each atom ($1 \leq i \leq N$); positions are global parameters.

Transitions between layers, $l \rightarrow l+1$ (before the nonlinearity):

Dense atom-wise, $x_i^{l+1} = W^l.x_i^l + b^l$;

Convolution feature-wise: $x_i^{l+1} = (X^l \star W''^l)_i$. Convolutions depend on relative distances.

Smooth shifted SoftPlus instead of ReLU.

Weights to be fitted as functions of relative positions.

Spherical Wavelet Expansion approach led by S. Mallat

Key reference: Eickenberg, Exarchakis, Him, Mallat, Thiry (2018) J Chem Phys 148.

General with respect to chemical elements.

Global transform, no explicit reference to local environments.

Smoothed densities $\rho(r) = \sum_i n_i g(r - r_i)$, smooth kernel g; separate densities for core and valence electrons. (Optional bond densities; ignore those.)

Solid harmonic wavelet basis functions $\psi^m_{j,l}$; convolutions with densities ρ.

Now symmetrize with respect to rotations and translations ...

Finally multilinear regression.

Atomic cluster expansion by R. Drautz, Bochum

Key reference: Drautz (2019) PRB 99.

Local environment of atom i, descriptors $A_{iv} = \sum_j \phi_v(\vec{r}_{ji})$, where the ϕ_v are a family of basis functions: $v = (nlm)$ and then

$$\phi_v(\vec{r}) = \sqrt{4\pi} R_{nl}(\|\vec{r}\|) Y_l^m(\vec{r}/\|\vec{r}\|).$$

Cluster products involving Clebsch-Gordon coefficients

$$B^{(K)}_{i,\vec{n},\vec{l}} = \sum_{\vec{m}} \mathrm{CG}(\vec{l},\vec{m}) \times A_{i,n_1,l_1,m_1} \cdots A_{i,n_K,l_K,m_K}.$$

Finally $E_i = \sum_{K,\vec{n},\vec{l}} c^{(K)}_{\vec{n},\vec{l}} B^{(K)}_{i,\vec{n},\vec{l}}$

Drautz (2019) also describes a nonlinear version to overcome slow convergence of the cluster expansion.

DeepMD approach led by E and Car, Princeton Univ.

Key reference: Zhang, Han, Wang, Car, E (2018) PRL 120.

Recall $V = \sum_i V_0(X_i)$ where X_i are the collective nuclear Coordinates for a local environment of the i-th atom.

Local environment is shifted to i-th atom, rotated in a problem-dependent manner. Let j enumerate neighbouring atoms within a cut-off distance.

Environment descriptor $D(X_i) = \mathrm{Sort}\{D_{ij}\}$ sorted by chemical species and by distance R_{ij}.

$D_{ij} = (1/R_{ij}, x_{ij}/R_{ij}^2, y_{ij}/R_{ij}^2, z_{ij}/R_{ij}^2)$

Obtain V_0 as output of a deep neural network with inputs the ordered $(D_{ij})_j$.

DeepPot-SE, deep potential smooth edition, from Princeton University and IAPCM

Key reference: Zhang, Han, Wang, Saidi, Car, E, NIPS 2018.

Follow-on to the DeepMD work, but now with due respect for continuity, energy conservation and vector covariance.

Environment descriptors without rotation or sort:
$D_{ij} = s_{ij}(r_{ij}) \times (1, x_{ij}, y_{ij}, z_{ij})$; $s_{ij} \to 0$ for large r_{ij}.

Two-stage NN: an encoding network and a fitting network.

Encoding network maps local environment to a feature space preserving point group and permutation symmetry.

Fitting network is fully connected feed-forward neural network with skip connections.

Perspective from Machine Learning .. Intro

Basic feed-forward neural network: $v^{(l)} = \phi(W^l_{l-1}.v^{(l-1)})$.

$v \in R^b$; ϕ e.g. pointwise ReLU; $W \in \mathrm{Mat}(b, b)$ to be learned.

Structured network: $v \in X^b$, $X \sim \mathbf{R}^N$ with some structure. W has a corresponding structure.

Example (Convolutional Neural Network, CNN): $X \sim \mathbf{R}^{m \times n}$, pixelized greyscale image. Elements of W are convolutions with compact kernel.

Parameterized network: Data depend on parameters p, weights to be learned as a function of p. (Linear regression formulation.)

Invariants and Covariants

Vector spaces U and V, group G with representations on U and V. For $g \in G$ we write $g.u$ or $g.v$ for the action of g on a typical element $u \in U$ or $v \in V$.

Function $f : U \to V$ is covariant (equivariant) if for all $g \in G$ and $u \in U$, $f(g.u) = g.f(u)$.

(Invariants are the special case $g.v \equiv v$.)

Example, the dipole moment $\mathbf{d}(X)$, $X \in R^{3N}$. Invariant under $\mathrm{Sym}(N)$, covariant under $O(3)$.

Represent it by effective charge model: $\mathbf{d}(X) = \sum_i w_i(X)\mathbf{r}_i$; then the weight vector $w \in R^N$ is covariant under $\mathrm{Sym}(N)$ and invariant under $O(3)$.

Point Cloud Convolutional Networks

Permutation equivariant neural networks ("Deep Sets"): Zaheer, Kottur, Ravanbhakhsh *et al.*, NIPS 2017.

Structured and parameterized: $X \sim \mathbf{R}^N$, $v \in \mathbf{R}^{N \times b}$; interpret as feature vector of size b for each of N points in $\mathbf{R}^3$. Parameters p: positions $\mathbf{r}_i$ $(1 \leq i \leq N)$. Weights W represent local convolutions; depend on local distances.

Point cloud convolutional networks are covariant (equivariant) under $\mathrm{Sym}(N)$.

See also: [SpiderCNN convolutional filters on point sets: Xu, Fan, Xu, Zeng, Qiao, Proc ECCV 2018], [PointConv deep convolutions on point sets: Wu, Qi, Fuxin, Arxiv 2018], other work on permutation equivariant NN.

Point Clouds with Additional Structure

Basic point cloud network has $v \in \mathbf{R}^{N \times b}$; feature vector of size b for each of N points. Group is $\mathrm{Sym}(N)$; feature vectors are unstructured. Imagine additional structure in the feature vector.

Additional group G of transformations on v, especially permutation group or $\mathrm{SO}(3)$; parameters and feature vector have definite transformation properties under G.

Group-Equivariant CNN: Cohen and Welling, ICML 2016.

Spherical Convolutional Neural Networks: Cohen, Geiger, Köhler, Welling, ICLR 2018.

Tensor Field Networks: Thomas, Schmidt, Kearnes *et al.*, Arxiv, 2018.

Gauge Equivariant CNN: Cohen, Weiler, Kicanaoglu, Welling, Arxiv, 2019.

Conclusions

There are promising new approaches to force fields with inspiration from big data and machine learning. (No assessment here of relative merits.)

There are valuable related developments from machine learning community inspired at least in part by application to atomistic force fields.

BJB wish list for developments ...

Simultaneous learning of energy, dipole, quadrupole moment through local charges with long-range interactions.

Fit or learn bands in solids invariant under $\mathrm{SL}(3, \mathbf{Z})$.

Learn local effective Hamiltonians for excited states.

Other uses of potential energy surfaces

Supplementary slides.

Molecular spectroscopy

Eigenvalue problem $H\Psi = E\Psi$:

$$-\sum_i \frac{\hbar^2}{2m_i}\Delta_i\Psi(X) + V(X)\Psi(X) = E\Psi(X)$$

Configuration interaction approach (Hartree products):

$$\Psi(X) = \sum_\alpha c_\alpha \Psi_\alpha(X)$$

$$\Psi_\alpha(X) = \Pi_i \psi^{(i)}_{\alpha(i)}(x(i))$$

This provides the ro-vibrational spectrum. Tractable for small molecules: e.g. $2(H_2O)$, CH_3OH; up to 9 atoms in our work.

Diffusion Monte Carlo

Ground state wavefunction: $H\Psi = E_0\Psi$.

$$-\sum_i \frac{\hbar^2}{2m_i}\Delta_i\Psi(X) + V(X)\Psi(X) = E_0\Psi(X)$$

Steady state for reaction-diffusion equation:

$$\frac{\partial\Psi}{\partial t} - \sum_i \frac{\hbar^2}{2m_i}\Delta_i\Psi(X) + V(X)\Psi(X) = E_0\Psi(X)$$

Can be solved in many dimensions using random walk with birth and death processes.

Result is ground state energy E_0; plus sample from the ground state wavefunction. (Sample $|\Psi^2|$ via descendant weighting.)

Quantum statistics

Partition function $Z(\beta) = \mathrm{tr}(e^{-\beta H})$. Thermal averages:

$$< A >_\beta = \frac{1}{Z(\beta)} \mathrm{tr}(A e^{-\beta H})$$

Use $e^{-\beta H} = (e^{-(\beta/n)H})^n$; $H = T + V$; $n \to \infty$. Let $\beta_n = \beta/n$, $\omega_n = 1/\beta_n \hbar$;

$$V_n(\mathbf{X}) = \sum_i (V(X_i) + \frac{1}{2} m \omega_n^2 (X_{i+1} - X_i)^2)$$

$$Z_n(\beta_n) = \int e^{-\beta_n V_n(\mathbf{X})} dx$$

Path Integral Monte Carlo.

Ring Polymer Molecular Dynamics

Due to David Manolopoulos (Oxford). PIMC plus time evolution. Classical hamiltonian:

$$H_n(x,p) = V_n(x) + \sum_i \frac{p_i^2}{2m}$$

$$\frac{dx}{dt} = \frac{\partial H_n}{\partial p}\,, \quad \frac{dp}{dt} = -\frac{\partial H_n}{\partial x}$$

Seen as a model for calculating the quantum Kubo correlation function.

$$\tilde{c}_{A,B}(t) = \frac{1}{\beta Z(\beta)} \int_0^\beta \mathrm{tr}(e^{-(\beta-\lambda)H} A(0) e^{-\lambda H} B(t)) d\lambda$$

Quantum scattering

Time-dependent Schrödinger equation:

$$i\hbar\frac{\partial}{\partial t}\Psi(X,t) = H\Psi(X,t)$$

Wavepacket propagation in an unbounded domain. Application to reaction dynamics is pretty much limited to 4-atom systems.