# Machine-learning interatomic potentials for materials science

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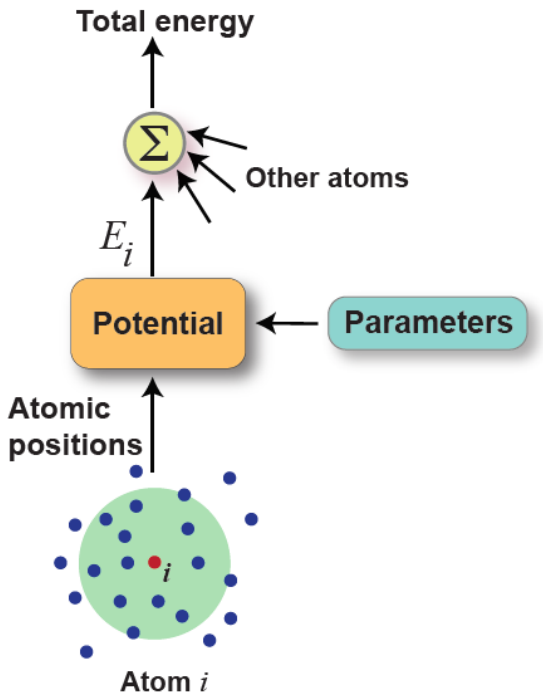
# Potentials

- **Electronic structure method**: based on the direct quantum-mechanical treatment of the electrons (accurate, 100 atoms, 100 picoseconds).
- **Classical interatomic potentials**: parameterize the system's configuration space and express its potential energy *E* as a function of the atomic position (accuracy compromise, exclude any treatment of electric, magnetic or optical properties).
- **Atomic positions**: $\mathbf{r}_i$
- **In the vicinity of the atom**: $\mathbf{R}_i \equiv (\mathbf{r}_{i1}, \mathbf{r}_{i2}, \ldots, \mathbf{r}_{in_i})$
- **The functional form of the potential**: $E_i = \Phi(\mathbf{R}_i, \mathbf{p}_i)$
- **Total energy**: $E = \sum_i E_i$.
- **Force**: $\mathbf{F}_i = -\partial E / \partial \mathbf{r}_i$
- **PES(potential energy surface):** the functional form can be represented by a 3N-dimensional hypersurface.

rotation
translation
permutation



# The physical basis of traditional potentials

- **Traditional potential**: the potential function is based on a physical understanding of interatomic bonding in the material.
- EAM, MEAM, and ADP for metallic systems
- Tersoff and Stillinger-Weber potentials for strongly covalent materials such as silicon and carbon
- COMB, REBO and ReaxFF for chemical reactions

Diverse and incompatible with each other

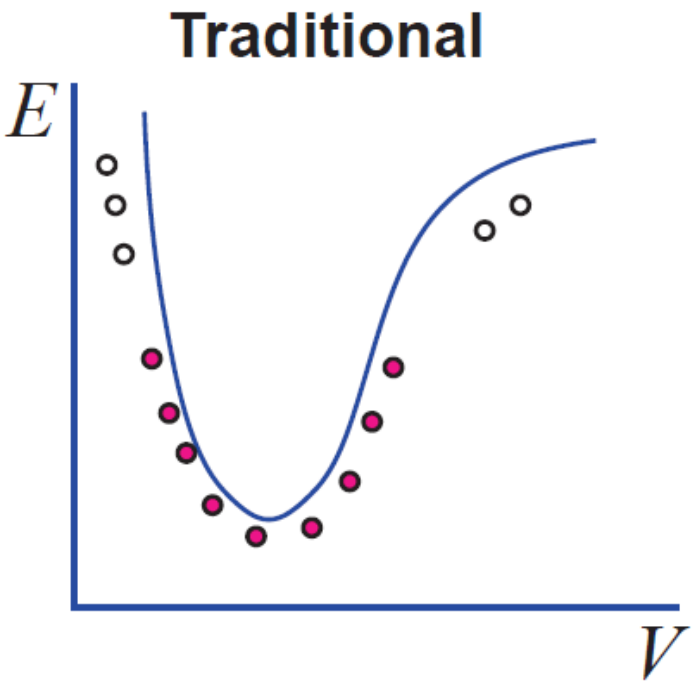
# Training of traditional potentials

- **Parameter**: $E_i = \Phi(\mathbf{R}_i, \mathbf{p}_i)$ $\mathbf{p} = (p_1, ..., p_m)$ (usually 10 to 20, same for all atoms)
- **Training**:
  1. **Database**: experimental data (physical properties) and a relatively small number of DFT energies of forces. (some physical properties require lengthy simulations)
  2. **Fitting**: directly fit the properties, not the PES.
  3. **Loss function**: the mean squared deviation of properties (with weight).
  4. **Human decision**: the hyperparameter. (not be automatized)
- **Multicomponent potentials**: preserve the underlying elemental potentials and only fit the parameters of the cross-interaction function (inheritance).

# Accuracy and transferability

| | Potential type | | |
|---|---|---|---|
| | Traditional | ML | Physically-informed ML |
| Physical foundation | Strong | None | Strong |
| Number of fitting parameters | $\sim 10$ | $\gtrsim 10^3$ | $\gtrsim 10^3$ |
| Computational speed | Very high | Slower[a] | Slower[a] |
| Reference database | Small | Large | Large |
| Accuracy (interpolation) | Limited | $\sim 1$ meV/atom | $\sim 1$ meV/atom |
| Transferability (extrapolation) | Reasonable | Poor | Reasonable |
| Reliance on human expertise | Strong | Weaker[b] | Weaker[b] |
| Extension to chemistries | Challenge | Challenge | Challenge |
| Specific to class of materials? | Yes | No | No |
| Systematically improvable? | No | Yes | Yes |
| Can be made artificial? | Yes | Maybe[c] | Maybe[c] |

specific surface reconstructions
complex dislocation core structures
struggle with complex elements Si(exhibit both covalent and metallic types of bonding) and C(bonding complexity and the existence of multiple metastable 3D, 2D, and 1D structures)



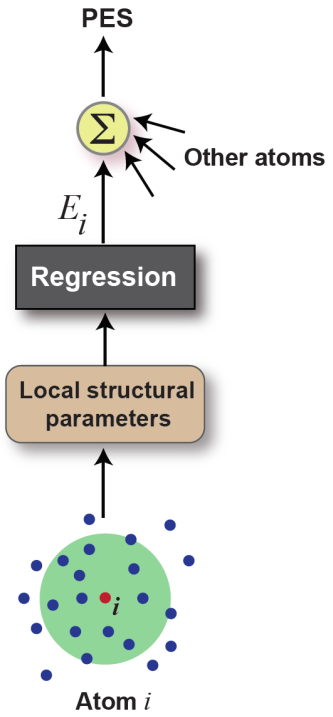
# Classification

- General-purpose potentials
- Special-purpose potentials
- Artificial(synthetic) potentials

# Machine-Learning potentials

| | Traditional potentials | ML potentials |
|---|---|---|
| Targets | A particular physical property or properties | PES |
| Database | Experimental data (a small number DFT) | DFT (without any experiment) |
| Method | Physics-based module | Purely numerical interpolation |

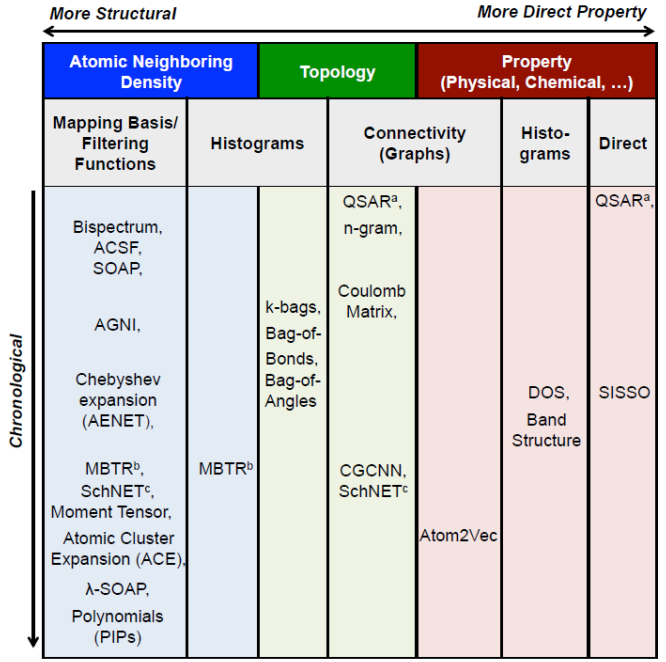
- PES uniquely defines all properties of the system.
  I. Local minima: stable or metastable structures
  II. Saddle points: energy barriers of thermally activated kinetic processes
  III. Curvature: elastic constants and phonon dispersion relations
- **Local structural parameters**: $\mathbf{G}_i = (G_{i1}, G_{i2}, ..., G_{iK})$ (smooth functions of $\boldsymbol{R_i}$ invariant under translations and rotations of the coordinate axes and permutations (relabeling) of atoms).
- **Regression model**: $\mathbf{G}_i \xrightarrow{\mathcal{R}} E_i$



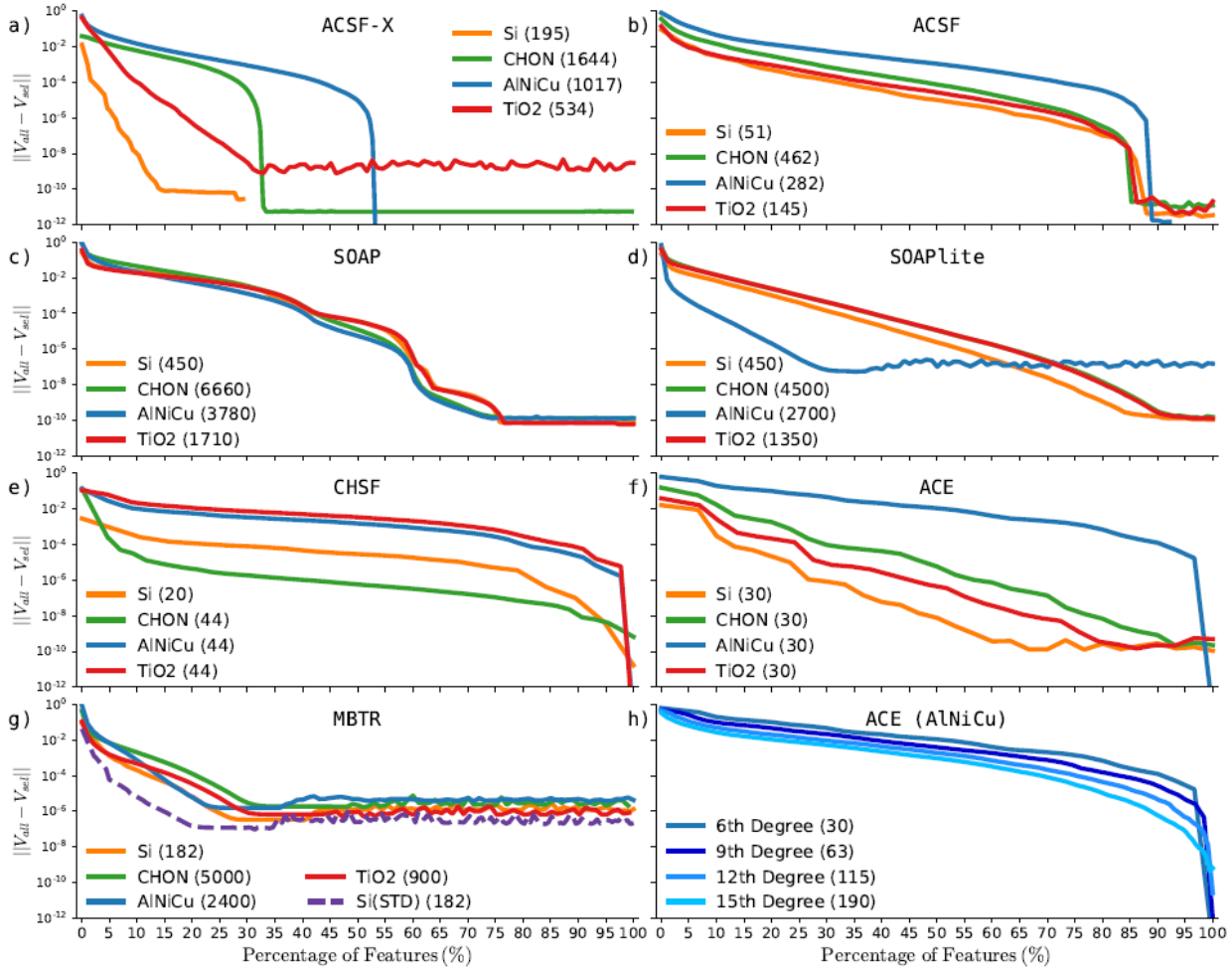

$$\mathbf{R}_i \rightarrow \mathbf{G}_i \xrightarrow{\mathcal{R}} E_i$$

# The local structural descriptors

- Dual effects:
  1. Ensure the mentioned invariance and smoothness of the PES.
  2. Replace the variable size position vectors $\boldsymbol{R}_i$.
- Local structural parameters: $\mathbf{G}_i = (G_{i1}, G_{i2}, ..., G_{iK})$ encode in a fixed number of invariant parameters, often referred to as local fingerprints.
- The common descriptors:
  1. Gaussian descriptors
  2. Zernike descriptors
  3. Moment tensor descriptors
  4. Smooth overlap of atomic positions(SOAP)
  5. Spectral neighbor analysis potential(SNAP)
  6. Atomic cluster expansion(ACE)



# The local structural descriptors

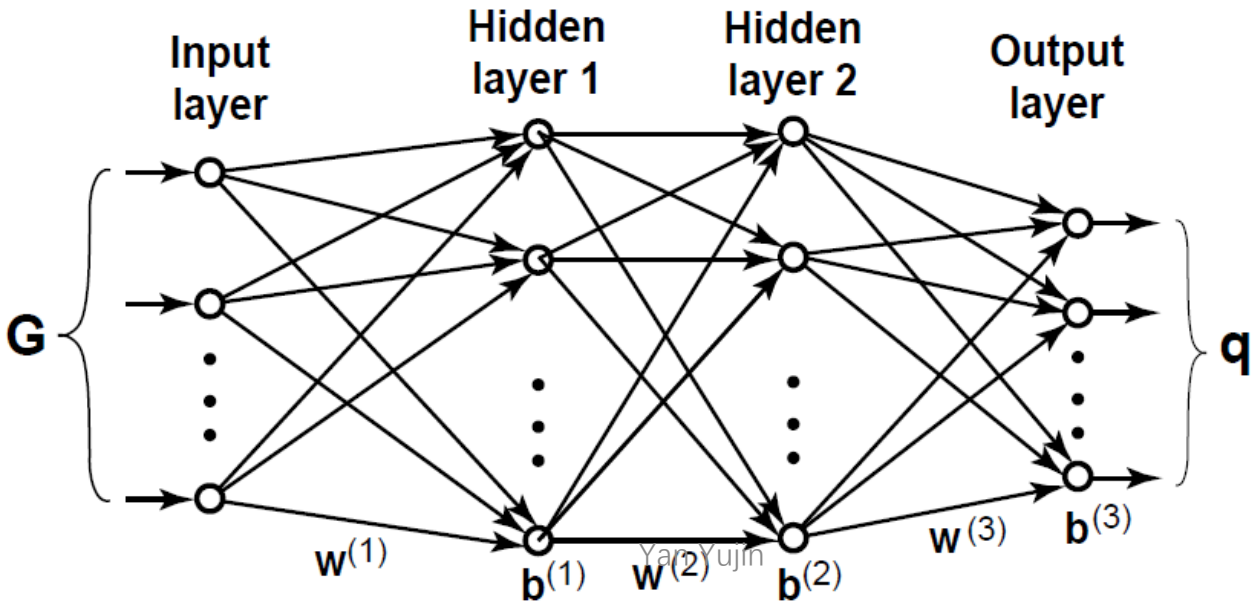


# The regression models

- Several high-dimensional regression models are available for mapping the local environments of atoms onto the PES:
  1. Gaussian process regression
  2. Kernel ridge regression
  3. SNAP module
  4. MTP potentials

  linear
  5. Artificial neural network (NN) regression

flexible, not affected by any constraints specific to the material system, and being universal approximators.



# Training

- Loss function:

$$\varepsilon = \frac{1}{N}\sum_{s=1}^{N}\left(\frac{E^s - E^s_{\mathrm{DFT}}}{N_s}\right)^2 + \tau_1\frac{1}{N}\sum_{s=1}^{N}\sum_{\alpha=1}^{3}\left[F^s_\alpha - (F^s_\alpha)_{\mathrm{DFT}}\right]^2$$
$$+ \tau_2\frac{1}{N}\sum_{s=1}^{N}\sum_{\alpha,\beta=1}^{3}\left[T^s_{\alpha\beta} - (T^s_{\alpha\beta})_{\mathrm{DFT}}\right]^2 + \tau_3\frac{1}{L}\sum_{\kappa=1}^{L}|p_\kappa|^2 .$$

- Optimization algorithms:
  1. backpropagation(steepest descent method)
  2. Levenberg-Marquardt
  3. Davidon-Fletcher-Powell(DFP)
  4. Broyden-Fletcher-Goldfarb-Shanno (BFGS)
- Training：
  1. Avoiding local minima: from different initial guesses
  2. Batch processing

# Validation

- Validation: root-mean-square error(RMSE) or mean absolute error(MAE)

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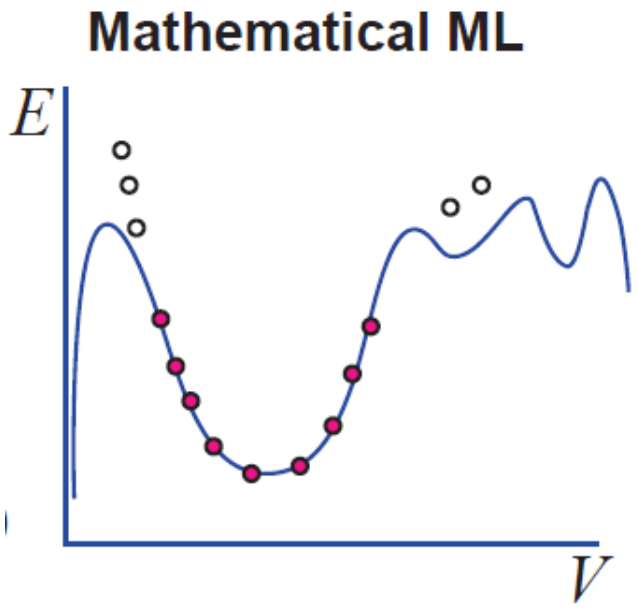

# Software

- Software: some are stand-alone packages, others are interfaced with LAMMPS, VASP, or other popular software.

1. ASE(Atomictic Simulation Environment)
2. Amp(Atomistic Machine-learning Package)
3. N2P2(Nerual Network Potential Package)
4. Aenet(Atomic Energy Network)
5. MLIP(Machine Learning Interatomic Potentials)
6. KLIFF(KIM-based Learning-Integrated Fitting Framework)
7. MAISE(Module for ab initio structure evolution)

# Discussion of ML potentials

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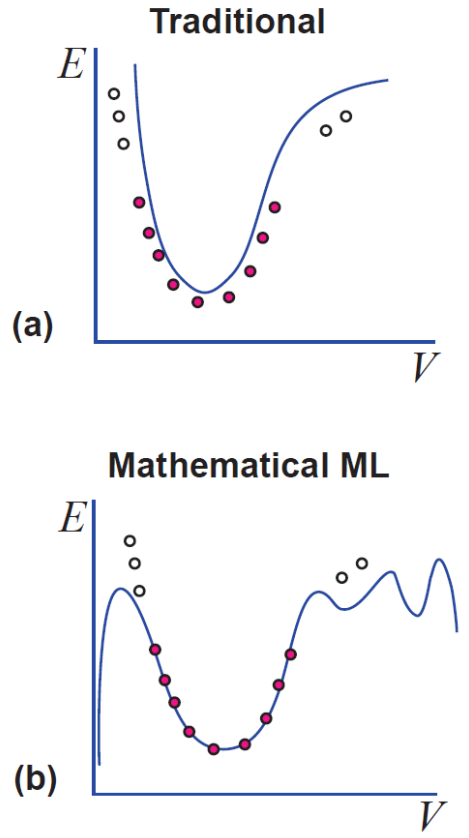
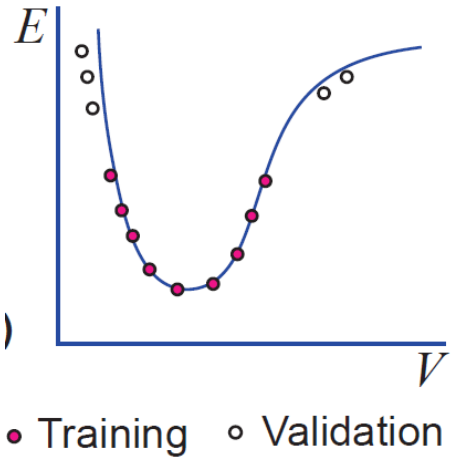
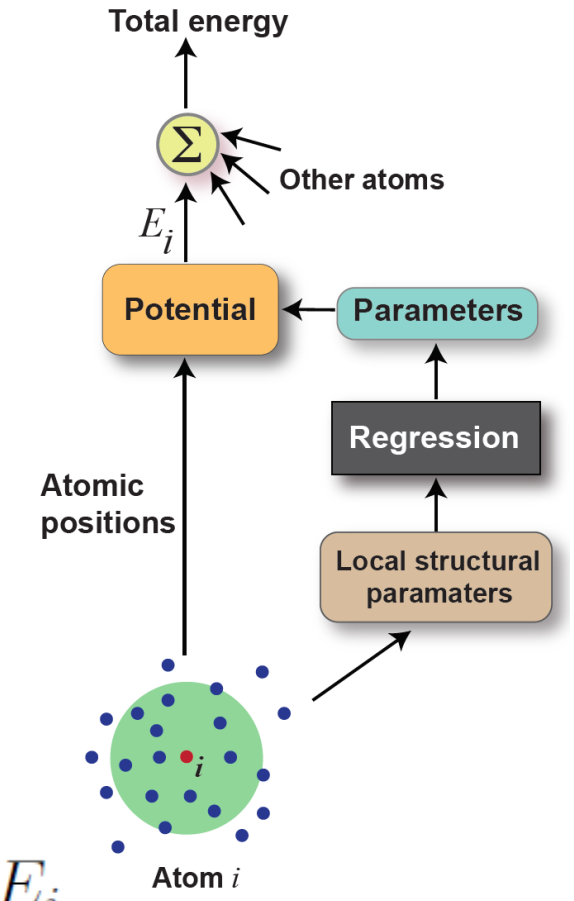


# Physically-informed machine-learning potentials

- Instead of directly predicting the atomic energy $E_i$, the regression $\mathcal{R}$ outputs a set of potential parameters $\boldsymbol{p}_i$ most appropriate for the environment of the particular atom $i$.

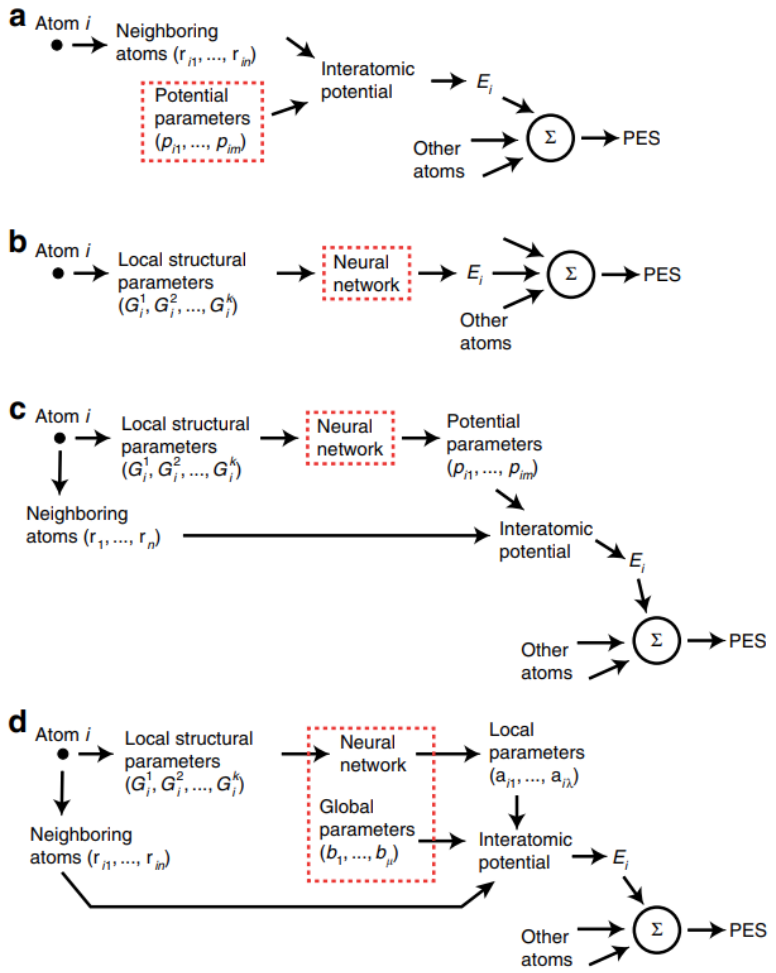
$$\Phi(\mathbf{R}_i, \mathbf{p}_i).$$




$$\mathbf{R}_i \rightarrow \mathbf{G}_i \overset{\mathcal{R}}{\rightarrow} \mathbf{p}_i \overset{\Phi}{\rightarrow} E_i$$

# PINN


a
Atom i
Neighboring atoms (r_i1, ..., r_in)
Potential parameters (p_i1, ..., p_im)
Interatomic potential
E_i
Other atoms
Σ
PES
b
Atom i
Local structural parameters (G_i^1, G_i^2, ..., G_i^k)
Neural network
E_i
Other atoms
Σ
PES
c
Atom i
Local structural parameters (G_i^1, G_i^2, ..., G_i^k)
Neural network
Potential parameters (p_i1, ..., p_im)
Neighboring atoms (r_1, ..., r_n)
Interatomic potential
E_i
Other atoms
Σ
PES
d
Atom i
Local structural parameters (G_i^1, G_i^2, ..., G_i^k)
Neural network
Local parameters (a_i1, ..., a_iλ)
Global parameters (b_1, ..., b_μ)
Neighboring atoms (r_i1, ..., r_in)
Interatomic potential
E_i
Other atoms
Σ
PES

# Physically-informed machine-learning potentials

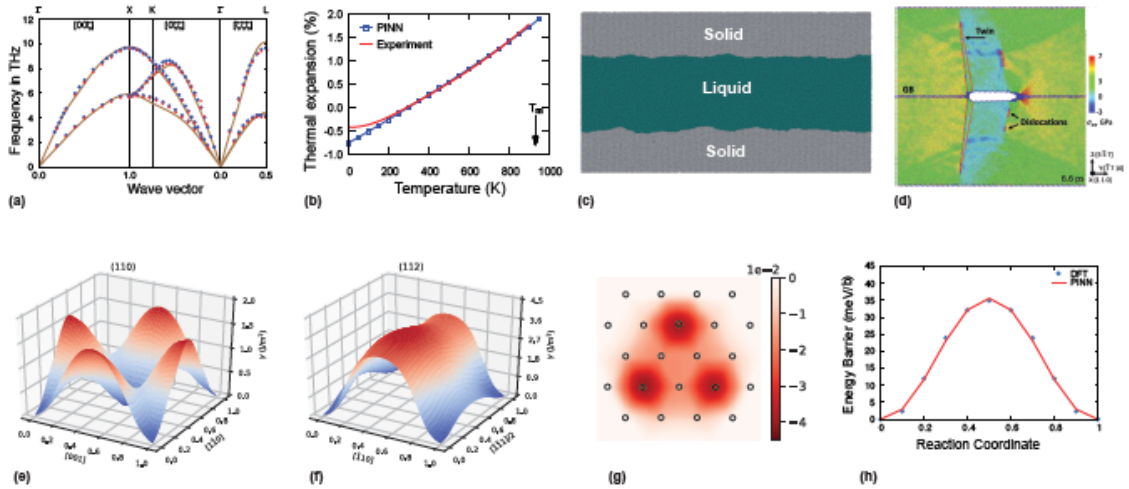



Figure 6: Examples of properties calculated with the PINN potentials for (a-d) Al [92] and (e-h) Ta [93]. (a) Phonon dispersion relations computed with the PINN potential (curves) compared with experimental data (points). (b) Linear thermal expansion relative to room temperature predicted by the PINN potential (points) compared with experiment (curve). (c) Simulation block used for computing the solid-liquid interface tension by the capillary fluctuation method. (d) MD simulation of crack nucleation and growth on a grain boundary. (e,f) $\gamma$-surfaces in body-centered cubic Ta on (110) and (112) planes, respectively. (g) Nye tensor plot of the core structure of the $\frac{1}{2}\langle 111\rangle$ screw dislocation in Ta predicted by the PINN potential. (h) Peierls barrier of the $\frac{1}{2}\langle 111\rangle$ screw dislocation predicted by the PINN potential (lines) in comparison with DFT calculations (points).

# Summary and outlook

- **Research direction**: demonstration of the new capabilities, the search for more effective descriptors and regression models, design of new algorithms to optimize and automatize the DFT database construction, and computational benchmarking.
- **Future**: discovering new and/or explaining known materials phenomena or predicting properties that cannot be computed otherwise.