Prediction of the Surface Energy from the Robust Extraction of Local Energy via Atomic Neural Network

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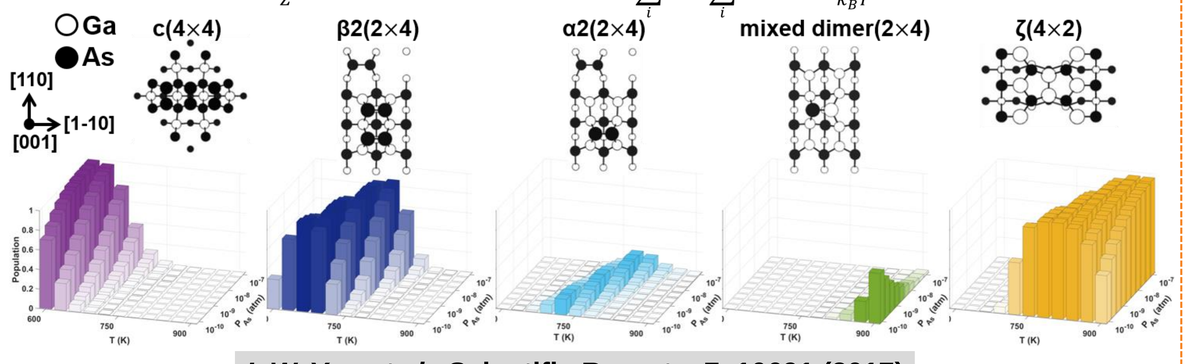
Computational Materials Design

Introduction & Motivation

Surface population (T,P) of GaAs (100) reconstructions

Population of reconstruction i:

$$c_i = \frac{Z_i}{Z} \; where \; i \in \{reconstructions\}, \qquad Z = \sum_i Z_i = \sum_i g_i \exp\left(-\frac{\gamma_i(T,P)A}{k_B T}\right)$$

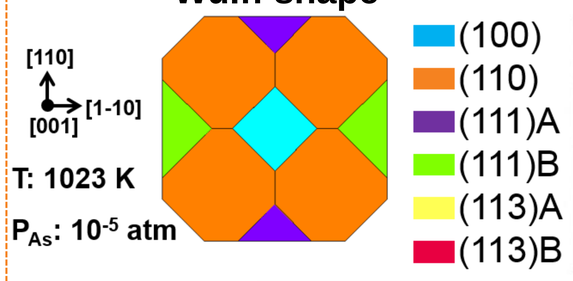


***I. W. Yeu et al.*, Scientific Reports, 7, 10691 (2017)**

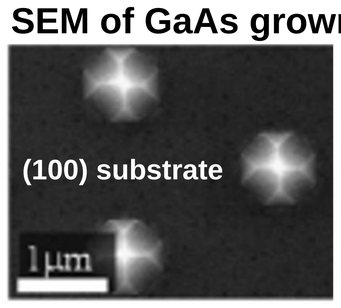
- The DFT calculation of surface energy along with ab-initio thermodynamics enables us to predict stable surface structures and population as a function of temperature and pressure.

Equilibrium crystal shape (T,P) of GaAs with new recontruction

Wulff shape



SEM of GaAs grown by SA-MOVPE



T: 1023 K

[TMGa]: 2.7×10^{-6} atm

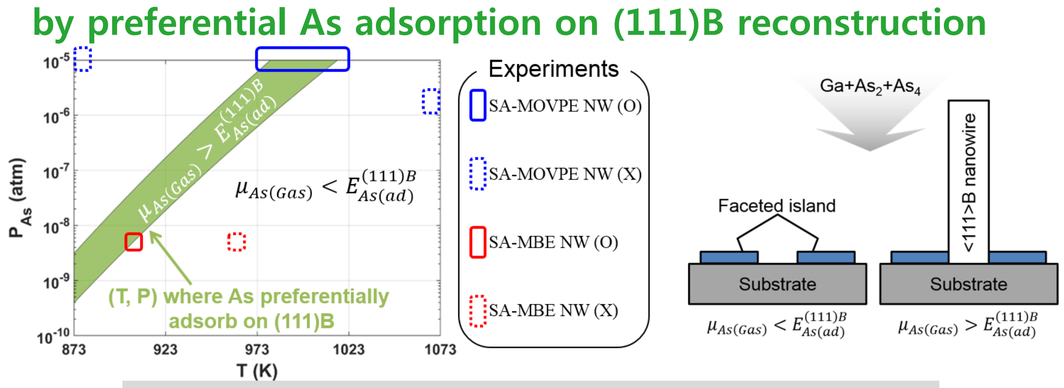
[AsH_3]: 5.0×10^{-4} atm

[J. Cryst. Growth. 298, 616 (2007)]

***I. W. Yeu et al.*, Scientific Reports, 9, 1127 (2019)**

- In particular, the absolute surface energy is crucial to compare the relative stability of each facet.

Facet-driven unidirectional <111>B growth under specific (T,P) by preferential As adsorption on (111)B reconstruction

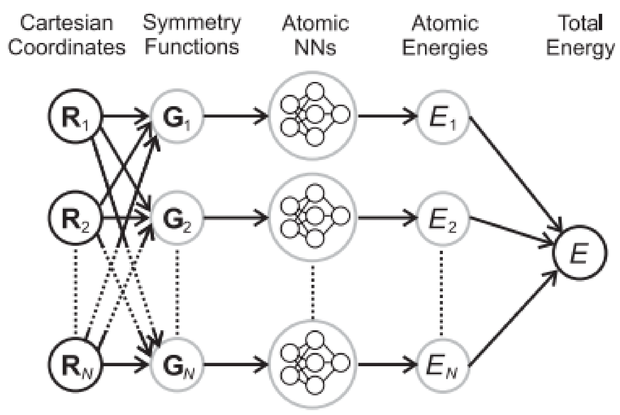


***I. W. Yeu et al.*, Applied Surface Science, 497, 143730 (2019)**

- In addition, the absolute surface energy is crucial to compare dynamics of surface reaction among different surfaces.
- **Problem:** the availability of the absolute surface energy is **restricted to symmetric slabs with two identical surfaces**, calculated by $\boldsymbol{\gamma = (E_{slab} - n_{slab}E_{bulk})/(2A_{slab})}$.

Purpose & Methods

Locality: atomic energy mapping by Atomic NN



$$\mathcal{E}(\{\mathbf{W}_\ell\}) = \frac{1}{2}\sum_{\sigma}^{structures}\left[E^{ANN}(\sigma;\{\mathbf{W}_\ell\}) - E^{\sigma}_{ref}\right]^2$$

$$= \frac{1}{2}\sum_{\sigma}^{structures}\left[\sum_{i}^{atoms} E^{ANN}_i(\sigma;\{\mathbf{W}_\ell\}) - E^{\sigma}_{ref}\right]^2,$$

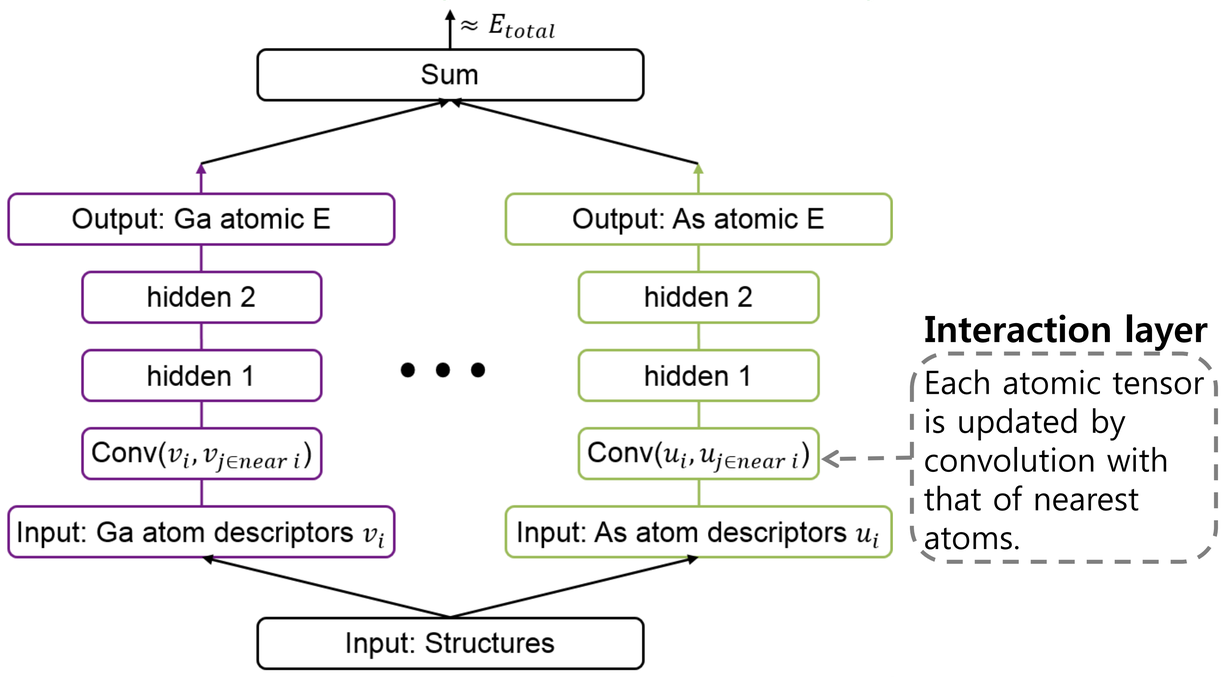
$$\{\mathbf{W}^{opt}_\ell\} = \arg\min_{\{\mathbf{W}_\ell\}} \mathcal{E}(\{\mathbf{W}_\ell\})$$

[PRL 98, 146401 (2007)]

- In general, **the obtained atomic energy is meaningless and not converged** because the Atomic NN training is **the process to minimize the error in total energy not in atomic energy**.
- Using **GaAs (111)AB polar surface as an example of asymmetric slab**, we demonstrate that **the robust extraction of the meaningful local energy** and following surface energy is possible by $\boldsymbol{\gamma = (E_{top;\,slab} - n_{top;\,slab}E_{bulk})/(A_{slab})}$.
- Importantly, it shows possibility of **convergence with respect to the number of datasets, descriptors, and hyper-parameters**.

Results & Summary

Model: Interaction layer + conventional FC layer

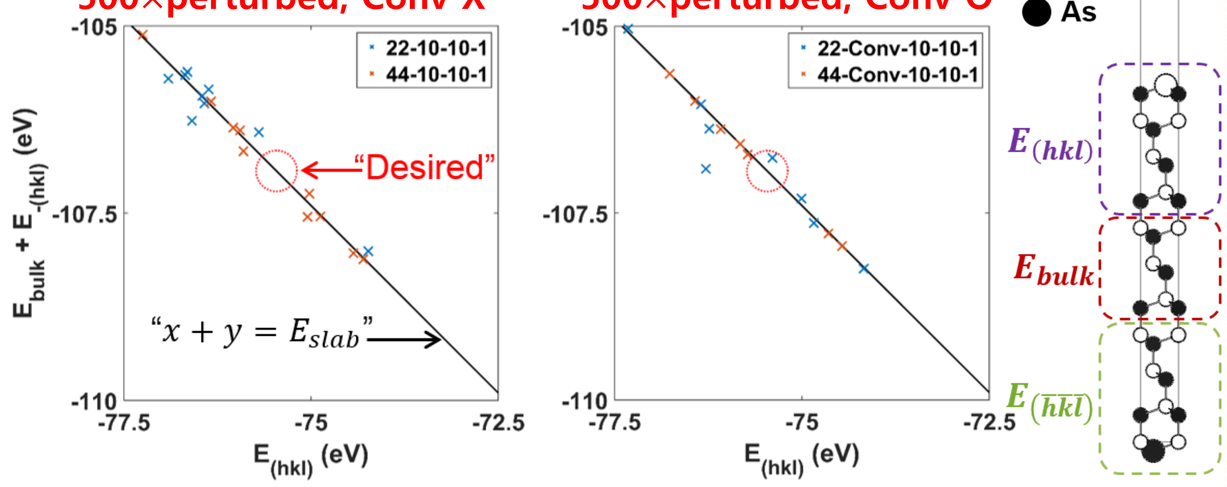


- Hyper-parameters: N_{input}-(Conv)-N_{hidden}-N_{hidden}-1 for each element.
- Datasets: 1×bulk + 1×slab + N×perturbed slab (8:2 as train:valid).
- Descriptor: Chebyshev, [PRB 96, 014112, (2017)].
- Weight initialization: Xavier (fan-average).
- Optimizer: Nesterov (momentum = 0.9).
- Training: early stopped with 500 patience.

Results: Extracted local energy from 10 independent trainings

$$\underbrace{E_{(hkl)}}_{"x\ (eV)"} + \underbrace{E_{bulk} + E_{(\overline{hkl})}}_{"y\ (eV)"} = E_{slab}$$

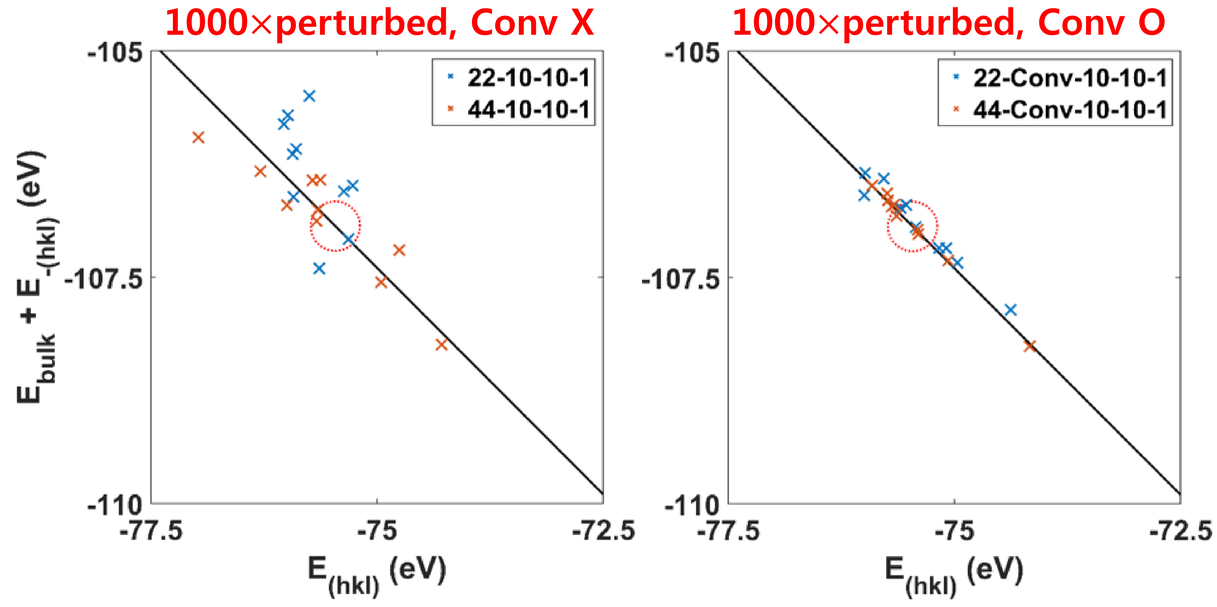
500×perturbed, Conv X | **500×perturbed, Conv O**



Red circle: correct local energy in which surface energy is calculated with error lower than 5 meV/Å^2 → deviation from this region indicates error in local energy.

Black solid line: exact total energy of slab → deviation from this line indicates error in total energy.

1000×perturbed, Conv X | **1000×perturbed, Conv O**



Summary

- **The inherent randomness of atomic NN**, caused by large latent space and random initialization, is a **severe obstacle to extract correct and converged local energy**.
- The flexible model with physically acceptable meaning **automatically adjusts the defined descriptors and shows convergence of the local energy**.