Machine Learning Potentials for Material Science

Construction of a Neural Network Potential for Supported Copper Clusters on Zinc Oxide Surfaces

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In Short

- Our group utilizes machine learning based neural network potentials to reproduce the potential energy surface of systems and perform simulations.
- These potentials are trained on thousands of single-point reference *ab-initio* calculations, and are able to approach the accuracy of the reference method with a computational efficiency close to classical force fields.
- Here we apply the potential to a model catalyst system, composed of copper and zinc oxide nanoparticles, utilized in the industrial synthesis of methanol.
- As a first approximation, we study structure and energy properties of copper clusters on zinc oxide surfaces. For this we use the potential to run different simulation schemes such as global optimization, grand canonical Monte Carlo, and constant velocity indentation.
- Computational time is required to extend the current potential to new configurations of the system, sing as a reference method DFT calculations with the VASP code.

Methanol is one of the main base products in the chemical industry. The catalyst used for methanol production in industrial plants from synthesis gas (a mixture of H_2, CO and CO_2) consists of copper nanoparticles, which are supported by zinc oxide. The investigation of the structural properties of the catalyst by computer simulations is very challenging for the presently available theoretical methods, since a direct application of methods such as DFT is prohibitively expensive due to the large system size.

In order to study the large systems involved, efficient interatomic potentials are required. A possible solution to this dilemma is the application of artificial neural networks [1,2], which have been shown to provide potentials with close to DFT quality for the Cu/ZnO system [3]. Neural network potentials (NNP) are able to reproduce the potential energy surface (PES) of a system, combining the high accuracy of electronic structure methods with the efficiency provided by a simple but flexible functional form. NNs have several properties that make them ideal candidates for constructing PESs: they are smooth and continuous, they do not require any knowledge about the underlying functional form, they allow to make and break bonds, and analytic derivatives are available for the calculation of forces. The NNP needs to be trained on hundreds of single point *ab-initio* calculations, using the calculated energies and forces to learn the PES of the system.

An example of one of our neural networks can be seen in figure 2. An atomic neural network calculates the forces on one atom, and the energy contribution of that atom to the total system energy; multiple atomic networks are then added together to obtain the total system energy. The atomic neural network is divided into an input layer, one or more hidden layers, and an output layer. Each layer is composed of nodes, functions of different types, with parameters (multiplicative weights and additive biases) connecting the nodes. In the input layer, the nodes are composed of symmetry functions, which process the environment around the atom of interest to provide a unique numerical fingerprint. In the hidden layers, the nodes are non-linear activation functions (usually hyperbolic tangent) that provide flexibility to the network.

The network is trained by adjusting the parameters connecting all the different nodes, until the outputs of the network (energies and forces) match those in the reference dataset (single-point calculations using a given *ab-initio* method). For this, thousands of single-point calculations are required, each giving a sample of the PES of the system. Once so trained, the network is able to reproduce energies and forces accurately but at a fraction of the computational cost of an *ab-initio* calculation. This allows for longer simulations in larger systems to be performed, but keeping the accuracy of the reference method.

The currently available NNP is able to accurately

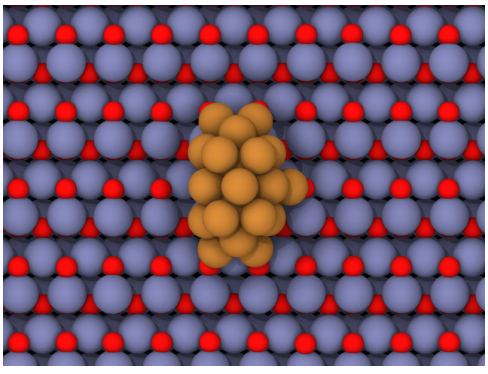

***Figure 1:** Globally optimized 24 atom copper clusters on the $(10\bar{1}0)$ low index surface of ZnO.*

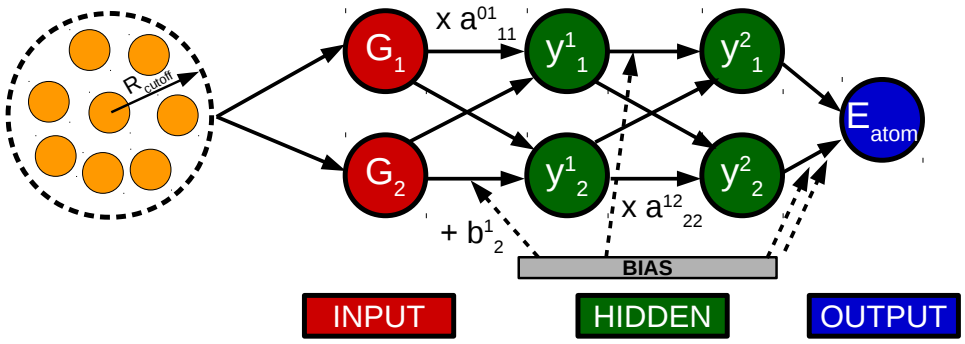

Figure 2: *Scheme of a single atomic neural network. a are multiplicative weights, b are additive biases, G are input symmetry functions, y are activation function nodes.*

reproduce many simple structural and energetic parameters for both bulk and slab structures of copper and zinc oxide. In addition, the potential is capable of successfully globally optimizing copper clusters on the $(10\bar{1}0)$ surface of ZnO with up to 30 copper atoms (and finding low energy minima for larger clusters). An example can be seen on figure 1.

The computation time at the HLRN will be utilized to extend the potential by calculating further structures utilizing as a reference DFT calculations with the program VASP. Three new environments will be considered: copper clusters on different ZnO substrates (ZnO $(11\bar{2}0)$, a stepped surface of ZnO containing interfaces of the two other low-index surfaces ($(10\bar{1}0)$ and $(11\bar{2}0)$), and a ZnO $(10\bar{1}0)$ surface with oxygen vacancies); clusters including oxygen and zinc atoms from the substrate; and partially embedded clusters into the substrate. We estimate that a total of 11500 structures will be required to accurately simulate all of these systems, the equivalent to 120k NPL hours.

With these extra structures, we will be able to perform a range of interesting simulations. In the first place, we will perform further global optimization searches utilizing basin hopping Monte Carlo, allowing us to compare the shape and size of clusters and the nature of the interface with the ZnO support between all the different ZnO surfaces. The simulations will also allow us to perform grand canonical Monte Carlo simulations, to study the distribution of sizes and shapes on large supporting slabs for different temperatures and copper chemical potentials. Including atoms from the substrate in the minimization will allow us to study the property of the clusters under more realistic conditions, and observing the possible shape changes induced by the new atoms. Finally, we will be able to study the embedding of the clusters into the substrate and the possibility of formation of amorphous interfaces and films, as observed in some experiments [4].

WWW

https://www.uni-goettingen.de/de/556198.html

More Information

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