Seminar

The new class of Moment Tensor Potentials

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Acronyms

AIMD *ab initio* molecular dynamics. 9

ALS active learning state. 8

cfg configuration. 9

DFT density functional theory. 6, 8–10

EAM embedded atom method. 4–7, 10

LAMMPS Large-scale Atomic/Molecular Massively Parallel Simulator. 9

MD molecular dynamics. 4, 5, 8, 9

MEAM modified embedded atom method. 5, 10

MTP moment tensor potential. 6–10

TILD thermodynamic integration using Langevin dynamics. 10

1 Classical potentials and their limitations

Classical potentials, such as embedded atom method (EAM) potentials [2], are widely used to perform molecular dynamics (MD) simulations. A number of different formulations exist, which vary in their amount of fitting parameters and therefore in the type of system that they can describe. Simple, empirical potentials, such as the Lennard-Jones potential, could theoretically be used to run MD simulations. And their analytical form could potentially make for very fast evaluations of energies and forces, and thus for very fast simulations in general. However, with only two free parameters that could be used to fit the potential to a given system, one can not expect to obtain accurate results and therefore quantitatively useful information for any system.

This point is intuitively understandable and valid for any potential parametrization: The higher the desired accuracy of the potential, the more complex the required model, the more free fitting parameters must be available and the more demanding the evaluation of forces and energy during runtime. It is the usual compromise between speed and accuracy that anyone working in the field is so painfully aware of.

The question of what kind of potential is best for a given simulation is by no means a trivial one, as the choice not only depends on the elements present in the system, but also on the physical phenomenon under investigation. The simulation of vacancy formation energy in a metal, in which all atoms can be assumed to be somewhat close to thermodynamic equilibrium may indicate the use of an EAM potential, whereas the simulation of the development of radiation damage may call for the use of screened Coulomb potentials, which may be less accurate in some areas, but describe the force between atoms in very close proximity well.

Generally, interatomic potentials can be classified according to the amount of atoms that are considered simultaneously during force/energy evaluation.

In a **single body potential**, no interactions of atoms are considered at all, which makes such a potential quite useless, since it can only describe external potentials, such as an electrical field, as shown in Figure 1.

The Lennard-Jones potential would be a **two body potential**, since the distance between two atoms is evaluated. Such potentials are, however, by design not capable of describing certain materials properties, such as all three elastic constants of cubic metallic systems simultaneously. As visualised in Figure 2, the interactions of three atoms A, B and C would have to be evaluated consecutively: $E(\mathrm{A}) = E(\mathrm{A–B}) + E(\mathrm{A–C})$.

Three body potentials would evaluate the relative positions of three atoms at a time, as in Figure 3, where $E(\mathrm{A}) = E(\mathrm{A–B–C})$.

Many body potentials, shown in Figure 4, that potentially include direct interactions of an arbitrary amount of particles, also exist.

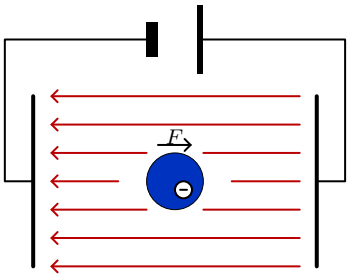
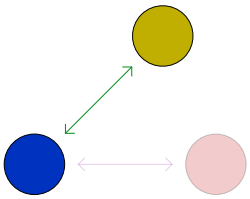
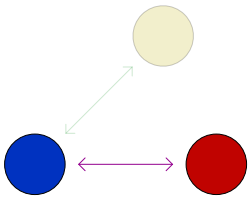


Figure 1: A single body potential can only descibe interactions with an external field, like an E-field.

(a) Two interacting atoms.



(b) First pair interaction.



(c) Second pair interaction.

Figure 2: A two body potential evaluates the interaction of two atoms at a time as shown in **(a)**. If there are more than two atoms, the interactions will still be evaluated pairwise. **(b)** and **(c)** show how the energy of the blue atom is evaluated.

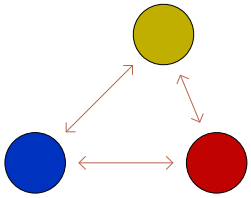


Figure 3: In a three body potential, the interactions of three atoms are evaluated simultaneously.

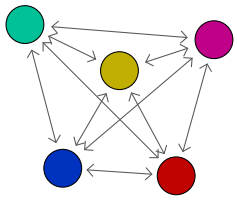


Figure 4: A many body potential simultaneously evaluates the interactions of an arbitray amount of particles.

Let's discuss the properties of one of the most popular interatomic potential parametrizations.

1.1 EAMs

EAMs [2] are very popular to describe metallic systems, in which a kind of electron gas is present, and directed, covalent bonds do not play a role. This type of potential is a member of the class of many body potentials. It is ignorant of any kind of "directions" and therefore cannot describe energy gradients with respect to angles, as would be required to reflect the notion of atomic orbitals necessary for directed bonds.

This parametrization involves three functions that solely rely upon atomic distances, which are shown in Figure 5 [6]:

Pair potential: A function $\phi_{\alpha(i),\alpha(j)}(r_{ij})$ acts as pair potential between the two atoms i and j of elements $\alpha(i)$ and $\alpha(j)$. For every combination of elements in the system, a separate function $\phi_{\alpha(i),\alpha(j)}$ exists.

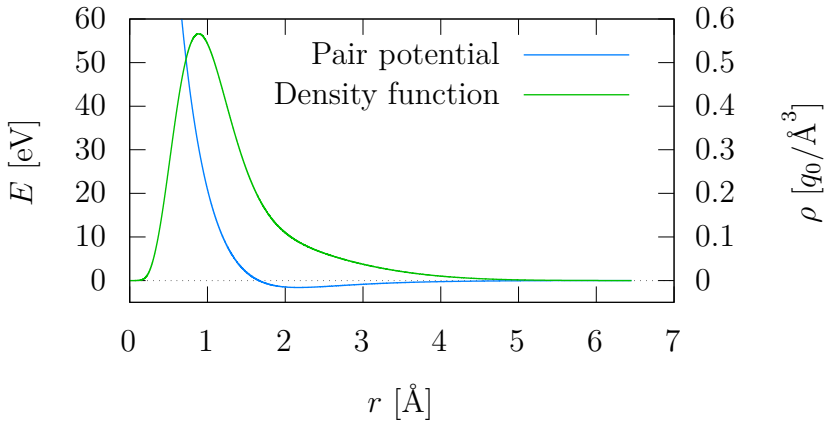
Electron density function: A function $\rho_{\alpha(i)}(r)$ describes the contribution an atom makes to the electron density surrounding it. In practical cases, this function often does not reflect the actual electron density, but is seen purely as a fitting parameter, and can even become negative.

Embedding function: A function $F_{\alpha(i)}(\rho)$ describes the potential energy of an atom due to the electron density at its current position.

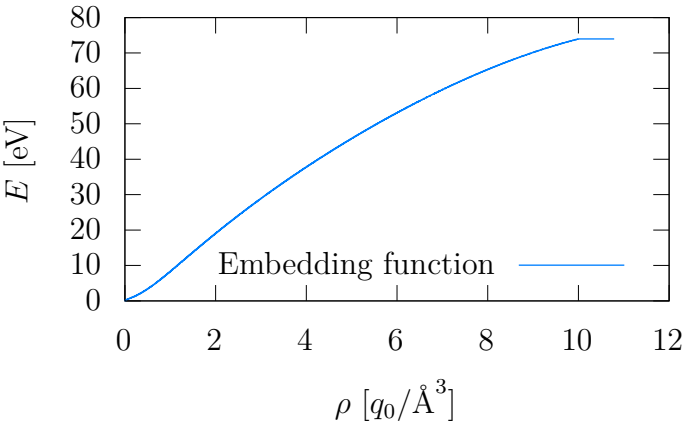
The full energy E_i of an atom i is then given by the formula

$$E_i = F_{\alpha(i)}\left(\sum_{j\neq i}\rho_{\alpha(j)}(r_{i,j})\right) + \frac{1}{2}\sum_{j\neq i}\phi_{\alpha(i),\alpha(j)}(r_{i,j}) \tag{1.1}$$

The many body character of EAMs is fully encapsulated in the interplay of the electron density function and the embedding function.



(a) Pair potential (left axis) and electron density function (right axis).



(b) The embedding function

Figure 5: Examples for the three different interaction functions used in EAM potentials. q_0 is the unit charge (charge of a proton). The shown functions correspond to Ni–Ni interactions from a CuNi potential by Onat and Durukanoğlu [6]

An extension to EAM potentials are modified embedded atom method (MEAM) potentials, in which the electron density function is multiplied with a screening function $S_{i,j}$ that depends on the coordination of the atom i, introducing an angular dependency to the formalism.

2 The gap between EAM and DFT

EAM and MEAM potentials are accurate enough to allow qualitative investigation of materials properties of many systems. They are also very efficient, allowing MD runs with several hundreds of thousands of atoms over several nanoseconds (which is great for many things). However, quantitative analyses of materials' properties are seldomly reliable, and potentials are often fit to accurately describe certain scenarios, like surfaces, but are inaccurate in others. So even for one given system one must often choose from a pool of potentials the one that is best suited to describe the phenomenon of interest. Within this inherent inaccuracy, though, it is possible to simulate a wide variety of things, from bulk properties, to surfaces, grain boundaries, defects such as dislocations, and more.

To achieve higher accuracy than provided by EAMs, one usually had to resort to much more computationally expensive methods such as density functional theory (DFT), which attempts to accurately describe the Quantum Mechanics at play during atomic movement. This involves applying the Born-Opponheimer approximation to generate the wave function of at least the outer electrons of each atom, increasing the number of particles that need consideration drastically and requiring a much more involved theoretical treatment. As a result, the system scales with $\mathcal{O}(N^3)$ of the number N of *particles* in the system (classical potential: $\mathcal{O}(n)$, $n \cong$ number of atoms), making computations involving more than a few tens of atoms infeasible.

As one can see, there exists an enormous gap of accuracy and efficiency between the two approaches, leaving scientists with the crude choice between complex systems at low accuracy and tiny ones with very high accuracy. Little could be done to bridge this gap.

In this work I present a new approach. Moment tensor potentials (MTPs), a new class of classical interatomic potentials with an arbitrary amount of fitting parameters and machine learning capabilities, are already relied upon to provide near DFT accuracy at computational cost of about 30 to 50 times that of EAMs. An investigation of the accuracy and stability of these potentials can be found in my Master's thesis [1].

3 What are MTPs

3.1 Mathematical structure

The precise mathematical formulation of MTPs is very involved and will therefore not be explained in this work. For more details, the reader is referred to Novikov et al. [5]. However, let's have a look at the general requirements of an interatomic potential and see how well they are met by MTPs as compared to EAMs.

3.1.1 Smoothness of the energy surface with respect to atoms leaving or entering the cutoff radius

For efficiency reasons, not all interactions of atoms in the simulation are considered when evaluating the energy of a single atom, since that would mean that the computational cost for energy evaluation for a single atom increased with the system size, which would quickly outgrow our computational resources. Also, in most systems, this approximation is a very good one, since interactions usually become negligible between atoms with a sufficient distance. Therefore, only atoms that are closer to each other than a certain cutoff radius $r_{\text{cut}} \approx 5\,\text{Å}$ are considered.

The requirement of smoothness essentially means that the interaction between atoms as described by the potential must drop to zero before the atoms leave the cutoff radius. Otherwise, since forces are proportional to the gradient of the potential energy surface, an artifical, extremely large force would be introduced whenever atoms reach the cutoff radius, leading to large errors in any atomistic simulation in which sufficient atomic movement is present.

By the choice of appropriate radial basis functions, this reqirement is easily fulfilled both by EAMs (as can be seen in Figure 5) and MTPs.

3.1.2 Invariance with respect to rotation, reflection and translation

Intuitively understandable, the energy of an atomic configuration should not depend on the angle or the distance from where it is observed, but only on local environments. That means that within a chosen coordinate system the energy of an atomic configuration must not change if it is being moved as a whole, rotated or reflected, as shown in Figure 6. Depending on the complexity of the mathematical model chosen to describe atomic environments, this may be challenging to achieve, or at least to prove. In the case of EAMs it is straightforward since, once again, all energies effectively only depend on atomic distances which are invariant with respect to the aforementioned operations. In the case of MTPs, this requirement is also fulfilled, yet its proof once again exceeds the scope of this work and the reader is again referred to Novikov et al. [5].

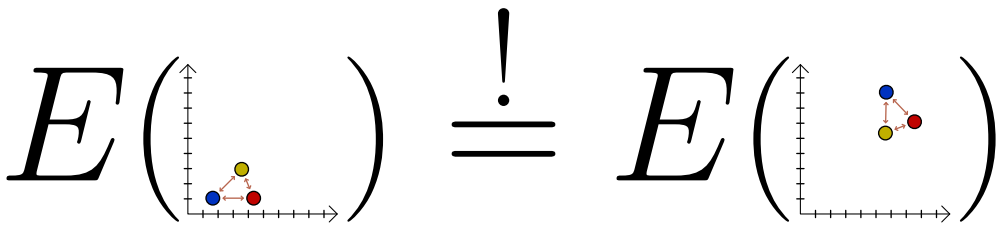


Figure 6: The energy of an atomic configuration must not be affected by the choice of coordinate sytem.

3.1.3 Complete description of local environments

To describe the energy of an atomic configuration accurately, one must first describe the configuration itself accurately. This means that any local environment of atoms should be fully and unambiguously described by the arguments of the potential (its descriptors), and that any possible configuration must be describable in the first place. The accuracy of the entire potential, no matter how many fitting parameters it has, will always be limited by this requirement.

In EAMs, for example, this requirement is not fulfilled, which is the method's greatest limitation. Because it only describes distances, the energy of a centered atom in a linear arrangement of, say, three atoms will *always* be the same as in a triangular arrangement, provided the two neighbors keep their distance to the central atom (but not necessarily to each other), as shown in Figure 7. This is an inherent limitation built into the method.

In MTPs the descriptors (the mathematics behind describing a local environment) are based on radial basis functions, analogous to EAMs, but are combined with angular descriptors, called moment tensors. They provide a full basis for local atomic environments and therefore fully satisfy this requirement. In conjunction with the fact that the amount of basis functions used to describe local environments can be arbitrarily varied, within the usual constrains of computational complexity, this allows for the construction of interatomic potentials of arbitrary precision for systems that fulfill the assumptions of classical MD-runs. Quantum mechanical effects, like the electronic structure of an atom, cannot be captured, though.

By fulfilling all three of these requirements the MTP provides a means for a highly accurate description of atomic interaction. Combined with the fact that the number of free parameters used to describe the the local atomic configurations can be arbitrarily varied, MTPs can reach very high accuracies, though this is associated with slower computations.

(a) Linear configuration. **(b)** Triangular configuration.

Figure 7: When using an EAM model, the blue particle in **(a)** and **(b)** would always have the same energy, since all positions on the circles surrounding it would be equivalent. This is a significant limitation of the EAM.

3.2 Additional features

At this point we have discussed the properties and quality an MTP can reach, if it is properly fitted to a system of interest. However, there exist other formulations of interatomic potentials that may reach similar accuracy through other means [8], and accuracy is not the only thing that matters in research. The use of any methodology must also be *practical*. And here, MTPs also have their perks!

Having a tool that describes a system well and effeciently is great once you actually have a fitted version of it, but getting to this point may be a challenge of its own.

The challenge arises in the form of choice of training set, which will determine the outcome of the fit, and therefore be decisive of whether or not the potential will describe the system you want, in the scenario you want, or not. Assume, for example, you fit your potential only to configurations of a bulk simulation box, and then attempt to simulate surfaces with it. Most likely, that won't work. This is an extreme example, and in reality other, perhaps much subtler effects may play a role, including those which are shown in Figure 8. In many cases, this means that a researcher has to fit a potential to a set of training data that he assumes is appropriate, and then find a way to validate this fact, or publish uncertain results. This process by itself may take months of manual work. With MTPs, the question of whether the trained potential is up to the task of accurately describing a given atomic configuration can be rationalized by a mathematical construct, called the D-optimality criterion [8]. This method relies solely on the current state of the MTP and the geometry of the configuration in question and does not require the use of any outside software, in particular no expensive DFT calculation.

The availability of a method to cheaply and automatically determine if the potential is accurate enough to describe a given configuration opens the door to efficient machine learning methodologies that elevate the potential from one that can be well fit to one that can *actively learn* to describe a system!

So how does that work?

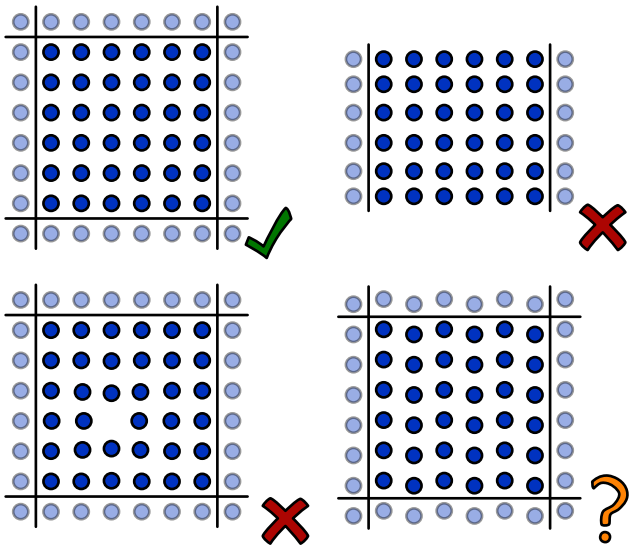


Figure 8: A potential fitted only to bulk properties is likely to accurately describe the bulk, but less apt to describing structures that deviate from it, like surfaces, vacancies or different crystal structures.

3.2.1 Learning-on-the-fly

The mathematics that describe the D-optimality criterion can be found in [8]. For now, just assume that we have a function γ(ALS, cfg) that depends only on the current active learning state (ALS) of the potential (the current state of the MTP and the configurations in its training set) and the configuration (cfg) in question, and that outputs a positive, real number.

The meaning of this number is as follows: If the cfg is well described by the potenital, i.e. it *interpolates*, $0 < \gamma \leq 1$. If $1 < \gamma \leq \gamma_{\text{select}} \approx 2$, the potential extrapolates, but this extrapolation can be assumed to be accurate. If $2 \approx \gamma_{\text{select}} < \gamma \leq \gamma_{\text{break}} \approx 5$, the cfg will still be described reliably, but would improve the quality of the MTP if it was added to its training set. Lastly, if $5 \approx \gamma_{\text{break}} < \gamma$, the extrapolation is unreliable.

Based on this, a full-flexed active learning-on-the-fly scheme can be built [4] (details may be adjusted to fit a given scenario, as this scheme is commonly pieced together using shell scripting):

I **Sampling:** Set up an MD run using an MTP. If a configuration with $\gamma > \gamma_{\text{select}}$ comes up, add that cfg to the set of sampled cfgs. If $\gamma > \gamma_{\text{break}}$, break the simulation and go to the selection stage. If the simulation does not need to be broken, your potential has become stable enough for the simulation to finish, and you can now get your results.

II **Selection:** Select a number of configurations from the sampled cfgs, which are as different from each other as possible.

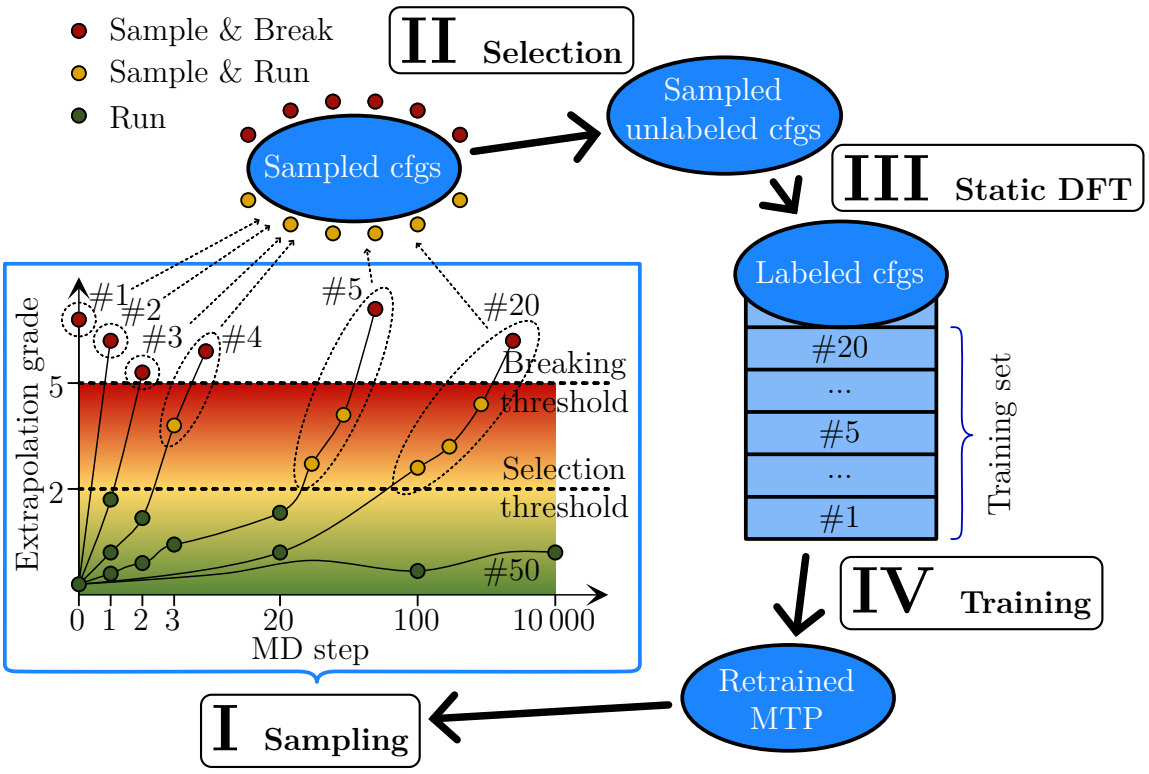


Figure 9: The learning-on-the-fly scheme. Adapted from Gubaev et al. [4].

III **Static DFT:** Determine the energies, stresses and forces associated with those cfgs using a static DFT calculation and add the then labeled cfgs to the MTP's training set.

IV **Training:** Retrain the MTP on the new configurations. Go back to I.

3.3 Implementation

Functions for all aspects of potential fitting, active learning, validation and some pre-processing are implemented in the `mlip` package [5]. It also provides an interface that allows the compilation of the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) with MTPs enabled.

4 Capabilities and potential use cases

The use cases of MTPs can be directly derived from their properties of being very accurate interatomic potentials that may need 30 to 50 times longer to compute than EAMs (according to experience). Due to their seamless integration with LAMMPS, they can

be used for any task that was formerly done with other potentials, though it may often require more computational resources.

At the institute of Materials Design at the University of Stuttgart, MTPs chiefly serve as a bridge between simple, analytical models of crystals, such as harmonic or quasi-harmonic approximations, and DFT.

In the calculation of thermodynamic properties of systems one uses thermodynamic integration using Langevin dynamics (TILD) [3] from said approximations to DFT to efficiently calculate free energy surfaces $F(V, T)$. These calculations run much quicker, if an intermediate system described by a classical interatomic potential is introduced. In the past, EAMs or MEAMs were applied to this purpose. However, the higher accuracy of MTPs allows for a further significant increase in efficiency if they are applied instead. Details about current research on this topic can be found in [1].

5 Summary

The class of moment tensor potentials (MTPs) is a highly accurate classical interatomic potential formulation. It's descriptors can capture any atomic configuration accurately and an arbitrary amount of parameters can be chosen to be used for regression, leaving the choice of balance between accuracy and efficiency entirely in the user's hands. While generally more computationally demanding than the embedded atom method (EAM), the possible gain in accuracy approaches that of density functional theory (DFT), opening the doors for previously inaccessible methodologies.

It exists a program that can be used to fit the potentials (the `mlip` package) [5], and they can be seamlessly integrated into Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [7], making the transition as simple as a specification of a different potential in existing codes.

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