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Predicting vacancy migration energies in lattice-free environments using artificial neural networks

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ABSTRACT

We propose a methodology for predicting migration energies associated to the migration of single atoms towards vacant sites, using artificial neural networks. The novelty of the approach, which has already been proven efficient for bulk materials (e.g. bcc or fcc Fe-based alloys), is to allow for any structure, without restriction to a specific lattice. The proposed technique is designed in conjunction with a novel kind of lattice-free atomistic kinetic Monte Carlo model. The idea is to avoid as much as possible heavy atomistic simulations, e.g. static relaxation or general methods for finding transition paths. Such calculations, however, are applied once per Monte Carlo event, when a selected event is applied. The objective of this work is thus to propose a methodology for defining migration events at every step of the simulation, and at the same time assigning a frequency of occurrence to them (using artificial neural networks), in short computing times. We demonstrate the feasibility of this new concept by designing neural networks for predicting vacancy migration energies near grain boundaries in bcc FeCr alloys.

1. Introduction

Atomistic kinetic Monte Carlo (AKMC) methods are widespread tools to study diffusion-controlled microstructural and microchemical evolution in alloys during thermal ageing and under irradiation (see, e.g. Ref. [1] and references therein). In these models, short time-scale motion, such as thermal vibrations of the atoms around their equilibrium position, is disregarded: only significant transitions are considered, allowing for long time scales (seconds, hours or even years) to be simulated. For convenience, they are often formulated in a rigid-lattice world, where the atoms of the alloy are located on the positions corresponding to the crystallographic structure of interest. In that case, the evolution of the system is driven by the migration of point defects (vacancies and/or self-interstitials), whose position is exchanged with nearest neighbor atoms. The jump to occur is each time selected stochastically, based on the Monte Carlo method. Simulations conducted with these methods can potentially predict in detail, and with high accuracy, phenomena such as precipitation or segregation, which are known to affect the mechanical and chemical behaviors of materials ([1] and references therein), thereby determining their aging when in use. There are two main limitations to the application of AKMC tools: the first one is their high computing cost; the second one is the physical reliability of the method itself. Rigid-lattice AKMC models are indeed of straightforward implementation and use, but it is not easy to make them computationally faster; on the other hand most of the physics is contained in the energy barriers associated with the migration jumps of the point defects, E_m, which must embody both the thermodynamics and the kinetics of the system being studied. The accuracy with which these migration energies are determined represent therefore a key issue. First, AKMC simulations will be the more reliable, the most adequate is the cohesive model used as Hamiltonian (interatomic potential or *ab initio* methods). Secondly, the physics stemming out of the cohesive model will be the most accurately transferred into the simulation, the better the influence of the local atomic environment (LAE) on E_m is included in terms of chemistry and strain field. Generally, however, improving the reliability of E_m calculations entails a significant increase in the computational cost of the algorithm.

In previous works [1,2], we proposed a fast rigid-lattice AKMC algorithm to perform accurate simulations of thermal annealing processes in bulk materials. We could accurately simulate the kinetics of precipitation of either Cr [1] or Cu [2] in bcc Fe, obtaining results in excellent agreement with experimental data. The same method was also extended to more complex alloys (e.g. the FeCrW system [3]), and also to the modeling of the mobility and stability of vacancy clusters [3,4] in irradiated materials. The main

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idea of the model is to rely on a description as a rigid-lattice of the simulated system, and calculate the migration energies associated to the elementary vacancy jumps using the nudged elastic band (NEB) method [5], thereby making as little approximations as possible. Relevant interatomic potentials are used as cohesive energy functions. Artificial Neural Networks (ANN) [6] are designed to predict these migration energies, receiving as input vector a precise description of the LAE. Once trained on the basis of a finite database of relevant examples, the ANN is used in complete replacement of NEB, and the AKMC simulation proceeds very fast.

As such, this ANN-based AKMC algorithm is inapplicable to more complex systems where the rigid-lattice approximation would no longer be valid, such as at free surfaces, grain boundaries, or in the bulk of materials containing dislocations or nano-structural features such as nanovoids and dislocation loops. The limitation is twofold: (a) the impossibility to refer to a "perfect material configuration" at all times complicates or may even prohibit the definition, given the present state of the system, of all possible transitions to a nearby basin of the potential energy; (b) these transitions are *a priori* of different kinds from one event to another, which complicates the formulation of empirical laws, or the design of numerical procedures, to assign a frequency of occurrence to them.

Lattice-free AKMC models, designed to handle very general cases without being bound to restricting hypotheses, do exist. At every step, they explore the local curvature of the potential energy surface, and find transitions to nearby basins looking for saddle-points, as specified by the transition-state theory [7]. For example, in Ref. [8], Henkelmans and Jonsson proposed to use the Dimer method [9], which in theory fulfills the objective to search for all possible transitions. Other authors proposed different schemes, based on other search methods though having the same finality, e.g. the ART method [10] in Ref. [11]. In these schemes, the choice of the migration events, together with the calculation of their corresponding migration barriers, is made on the fly, allowing, given the initial state, to find all possible transition paths to other nearby local minima in the potential energy surface. The advantage is clearly the flexibility with respect to the simulated system. The main drawback, however, is the complexity of the method: any Dimer-like method requires appropriate parameterization to guarantee that most of possible transitions are found, which can be delicate for some systems. Also, as a consequence of the systematic repetition of search for saddle points, the required computing time is large, inherently limiting the practical application of the method to no more than a few thousands of events. This is certainly far insufficient to study long-term and slow processes such as precipitation or depletion of solutes at interfaces.

In this work, we develop the premises of a new kind of AKMC model that intends to be a compromise between the afore-discussed extremes. On the one hand, we aim at allowing for large-scale perturbations of a regular crystallographic structure. Here, we consider a simple example of interfaces: grain boundaries in bcc materials. On the other hand, the model is designed in such a way that heavy atomistic calculations are limited to the strict minimum, in two ways: (a) only static relaxation is necessary, and only once per MC event; (b) in the same line of ideas than our previous works, the migration energy associated to the MC events are very rapidly estimated using ANN's. As a first step towards this ambitious objective, our goal here is to demonstrate the viability of the model, by showing how these ANN's can be designed to predict the vacancy migration energy, in any position of the simulated material, i.e., close to the grain boundary and far from it.

The paper is organized as follows: in Section 2, we start, to fix the ideas, by describing in general terms the new lattice-free AKMC model. The goal is to discuss the hypotheses and approximations that are made, and most importantly to define the exact purpose of the ANN in the model. Next, in Section 3, we extensively describe how these ANN's are designed to accurately predict the migration energies in any structure, which is the bedrock of our model. Last, in Section 4, we show an application of the proposed methodology to a simple case of interfaces: grain boundaries in bcc FeCr alloys.

2. Lattice-free AKMC model based on artificial neural networks

Lattice-free AKMC models are aimed at replacing fully detailed atomistic simulations (molecular dynamics) by a larger-scale model based on transition-state theory [7]. The requirement is thus, at every step of the simulation, to:

1. Make a list of possible transitions from the present state, assumed to be an equilibrium position in the potential energy surface, towards nearby other stable positions.
2. Assign a frequency of occurrence Γ to all these possible transitions, calculated as a thermally activated event.

$$\Gamma = \Gamma_0 \cdot \exp\left(\frac{-E_m}{k_B T}\right) \tag{1}$$

Here, E_m is the migration energy and Γ_0 is an attempt frequency (which can be obtained using Vineyard's procedure [12]). Using general and non-approximate techniques, such as e.g. the Dimer [8], Monomer [13] or the activation-relaxation technique (ART) [10], these two steps are performed at the same time by exploring the local curvature of the potential energy surface around the present state. It is a complex operation, requiring a (very) long CPU time, because static relaxation and curvature calculations are involved.

As an alternative, we propose in this section a new concept of AKMC algorithm, where the above-listed operations are implemented in such a way to reduce the CPU time requirements to the extreme. This is explained in the two subsections below.

2.1. Generic procedure for defining transitions

To respect the transition-state theory, migration events in AKMC simulations should strictly correspond to the transition from the present state of the system towards a nearby metastable state. The migration path, from the initial state A towards the final state B should be, consequently, qualitatively similar to the one illustrated in Fig. 1(a): at the onset of the transition, the total energy of the system smoothly increases, until it reaches a local maximum. The latter is called saddle point, because it is characterized by a negative curvature in the direction of the transition, and a positive curvature in all others. Once the saddle point is crossed, the system falls into state B.

General methods for searching transitions (Dimer, Monomer, ART, etc.) directly search for saddle points from the initial state. Their contribution to the AKMC is thus twofold: (a) they perform themselves the identification of directions of migrations for the system towards the nearest saddle points, thereby establishing the list of events for the MC; (b) at the same time, they provide the value of the corresponding migration energy, necessary to calculate the events frequencies using Eq. (1).

Since we aim at avoiding these highly complex operations, we propose an alternative in two steps:

- Directions of migration are determined using an "event definition generic procedure" (EGP), where a complete exploration of the potential energy surface, searching for saddle points, is