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Diffusion of yttrium in bcc-iron studied by kinetic Monte Carlo simulations

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ABSTRACT

The formation of oxide nanoclusters in oxide dispersion strengthened steels is controlled by the diffusion of yttrium. Yttrium atoms and other oversized solutes show a high binding energy to vacancies and a considerable relaxation from their lattice site towards a neighboring vacancy. In the case of yttrium the relaxation is so prominent, that the resulting situation may also be considered as an interstitial atom sitting in between two vacancies. We calculated the yttrium-vacancy binding energy and the migration barriers of vacancy jumps in the vicinity of a yttrium atom by means of nudged-elastic band calculations using density functional theory calculations. These barriers were used in a kinetic Monte Carlo code to calculate the diffusivity of yttrium and investigate the diffusion mechanism of yttrium in bcc iron with focus on correlation effects. The results reveal that the diffusion of yttrium is due to a sequence of vacancy jumps between the nearest and third nearest neighbor shell of the yttrium atom.

1. Introduction

Oxide dispersion strengthened (ODS) steels are considered promising materials for the next generation of fission power plants and future fusion power plants due to their outstanding combination of mechanical properties and resistance to radiation damage [1]. The addition of thermally stable yttrium containing oxide nanoparticles drastically improves the creep resistance of ferritic steels which enhances the reactor efficiency [2]. The properties of ODS steels are heavily influenced by the size, distribution and composition of these particles [3–5]. Optimization of the processing conditions of ODS steels was therefore in the focus of several experimental [6–9] and theoretical [10–13] studies. Since the precipitation of oxide particles as well as the behavior under radiation is directly tied to the diffusion of solutes and point defects, a detailed understanding of the intrinsic diffusivities of the relevant species, in particular of yttrium, is a prerequisite for further optimizing ODS steels.

It has been shown, that there is an exceptionally high binding energy between the yttrium atom and a vacancy due to the considerably bigger size of the yttrium atom [14]. An yttrium atom with a vacancy in a nearest neighbor position relaxes from it's lattice position towards the vacancy [15]. The resulting configuration can be either considered as a yttrium-vacancy pair or as an interstitial yttrium atom in between two vacancies. Fig. 1 illustrates this interesting feature. As a result of this, the barrier for the yttrium-vacancy exchange is negligible [14,16] or not present at all [17] and not relevant for the diffusion mechanism. This raises the question of how the yttrium diffusion actually happens. Does the diffusion of yttrium require the separation of the yttrium-vacancy pair, which consumes a considerable amount of energy, or is there an other mechanism?

Due to their influence on radiation resistance of steel and the growth of precipitates the diffusion of solutes in bcc-Fe has been the objective of several computational studies [18,19]. The diffusion of yttrium is the limiting factor for the formation and growth of yttrium containing oxide particles in ODS steel [20,21]. It has been investigated by Murali et al. [16] and Gao et al. [14] using density functional theory (DFT) and Le Claire's nine frequency model, as well as by Bocquet *et al.* [17] who used an analytical model. Considering all these groups calculated a considerably lower activation energy for diffusion than Hin et al., where a precipitation growth model was fitted to experimental data [22], possible explanations could be, that Le Claire's model breaks down for highly correlated jumps or the restriction of the yttrium-vacancy interaction to the second nearest neighbor shell is to narrow.

Thus, in this work, the diffusion of yttrium in iron was simulated

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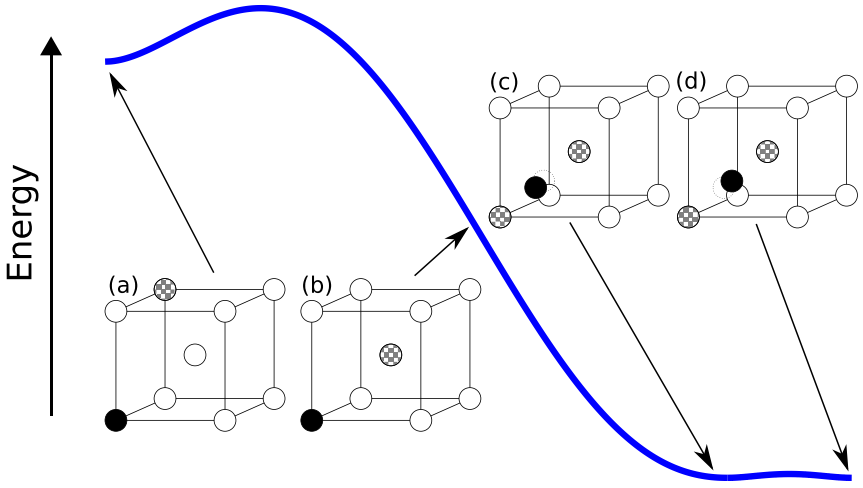

Fig. 1. Different configurations of yttrium (black circle) and vacancy (checkerboard circle) positions on a bcc-Fe (white circle) lattice and the corresponding energy landscape for transitions between these configurations. Configuration (a) shows yttrium and vacancy in third-nearest neighbor positions. Configuration (b) shows yttrium and vacancy in the nearest neighbor lattice positions. This configuration is not stable and the yttrium relaxes towards the vacancy which leads to configuration (c). Configuration (d) is the equivalent nearest neighbor position resulting from relaxing configuration (b) with swapped yttrium and vacancy positions. There is only a negligible barrier between configurations (c) and (d), so the yttrium atom might also be considered as occupying an interstitial position.

using a kinetic Monte Carlo (KMC) code [23]. This allows to investigate the diffusion without any assumptions as all correlation effects are automatically considered in KMC simulations. Both possible diffusion mechanisms were investigated: The 'classical' vacancy mechanism that is the basis of Le Claire's model and the 'interstitial' mechanism that considers jumps to and from the interstitial position between two vacancies as the relevant jumps. The 'interstitial' mechanism was also the basis for the analytic model by Bocquet et al. [17] Fig. 2 shows the two mechanisms. The rates of possible diffusion events were determined from migration barriers calculated by density functional theory (DFT) calculations. The KMC simulations shed light on the diffusion mechanism and allow to calculate the diffusivity of yttrium which can be compared to the value calculated using Le Claire's nine frequency model.

The paper is organized as follows. In Section 2 we discuss the DFT and KMC methods that are used in this work. Section 3.1 deals with the calculation of input data for the KMC calculations, namely the binding energies and migration barriers. The results of the KMC calculations are presented in Section 3.2 and discussed in Section 4. Our conclusions are presented in Section 5.

2. Calculation

2.1. Vacancy jump rates

The Vienna Ab initio Simulation Package [24–27] (VASP) was used to determine the migration barriers. The calculations were performed in a plane wave basis set with pseudopotentials from the VASP library based on the projector augmented-wave (PAW) method [28,29] and within the generalized-gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) parameterization [30]. The Brillouin zone sampling was done on a $3 \times 3 \times 3$ k-point grid and the plane wave energy cut-off was set to 500eV. The convergence criterion for the relaxations was achieved when all forces were less than 1 meV $Å^{-1}$. All 127 atom positions in the $4 \times 4 \times 4$ supercell were fully relaxed. The climbing image nudged elastic band method [31] was employed to determine the minimum energy path from one vacancy position to the next.

The binding energy E_i^b between yttrium atom and vacancy in position i was calculated by

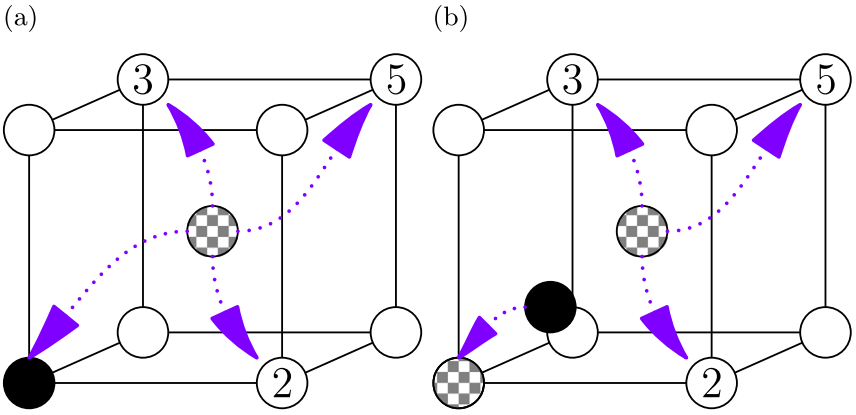

Fig. 2. Schematic of the diffusion mechanisms. In (a) the 'classical' vacancy mechanisms is shown with substitutional atom and vacancies on the lattice positions. The vacancy can either jump to a second, third or the fifth nearest neighbor position or it can change place with the solute atom. The 'interstitial' mechanism is shown in (b). Both of the vacancies can jump to second, third or fifth nearest neighbor positions which will push the solute atom to the position of the other vacancy.