Para 2010 – State of the Art in Scientific and Parallel Computing – extended abstract no. 49
University of Iceland, Reykjavik, June 6–9 2010
http://vefir.hi.is/para10/extab/para10-paper-49.pdf

Computation of transition states for extended defects in materials science: issues and challenges from selected exemples

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Abstract The determination at the atomic scale of transition states can be difficult in specific cases, especially when involving extended defects in materials science. We discuss some of these issues (complex mechanism, multiplicity of solutions) using several examples.

Keywords Materials Science, transition state, extended defects

1 Introduction

The knowledge of transition states is often a prerequisite in order to understand the spatial and temporal evolution of a system. It is especially true in materials science at the atomic scale. Although methods such as molecular dynamics allow to monitor this evolution at finite temperature, scale limitations in space and especially in time severely restrict their utility, despite many improvements[1]. To overcome these limitations, a solution is to directly determine the transition states which will govern the system evolution, by identifying the mechanism and computing the associated activation energy. In a multiscale modeling perspective, these data can then be used in other approachs such as kinetic Monte Carlo, which do not suffer from time scale limitations.

To characterize transition states in an effective way, several kind of methods have been proposed [2]. Although they have been shown to be extremely successful in many situations, there are still several cases for which their use is difficult. Our goal here is to show and discuss such cases, issued from recent problems in materials science.

2 Nucleation of extended defects: a complex and rare mechanism

A first example concerns the nucleation of dislocations in materials with nanometric dimensions. Due to the reduced dimensions in these systems, the relaxation of an applied stress can not be done through the multiplication of dislocations, via the Frank-Read source for instance. Instead, the formation of dislocations would occur at the surfaces (Figure 1), which are predominant at nanoscale. The understanding of the nucleation of dislocation from surfaces is therefore an interesting problem, which is especially important for the determination of the plastic properties of nano-objects.

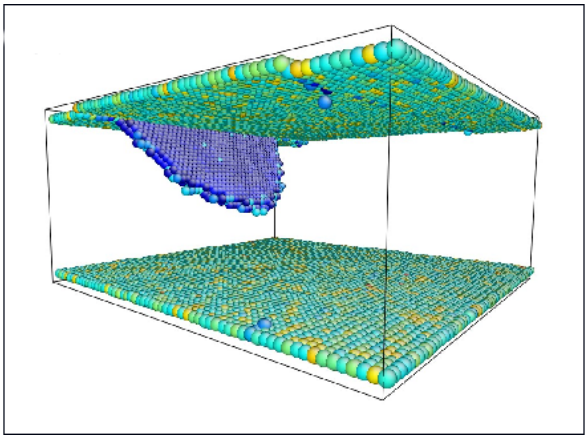

Figure 1: Half-loop dislocation nucleated from a surface in aluminum.

The nucleation process is typically a rare event, since only one is required to initiate the plastic deformation of the object. Because of time scale limitations, the probability to obtain it in molecular dynamics simulations is extremely low, except in conditions very far from experimental ones. We have used here the nudged elastic band method in the case of dislocation nucleation from surface in simple metals, since it is well suited for characterizing this mechanism. Results provide the activation energy as a function of the applied deformation, as well as the critical radius of the nucleated half-loop dislocation [3]. An important issue regarding the use of this method in this case is the need to provide an appropriate initial transition path in the calculations, although the process is complex. Another minor point is the difficulty to define a stable final state in the definition of the mechanism.

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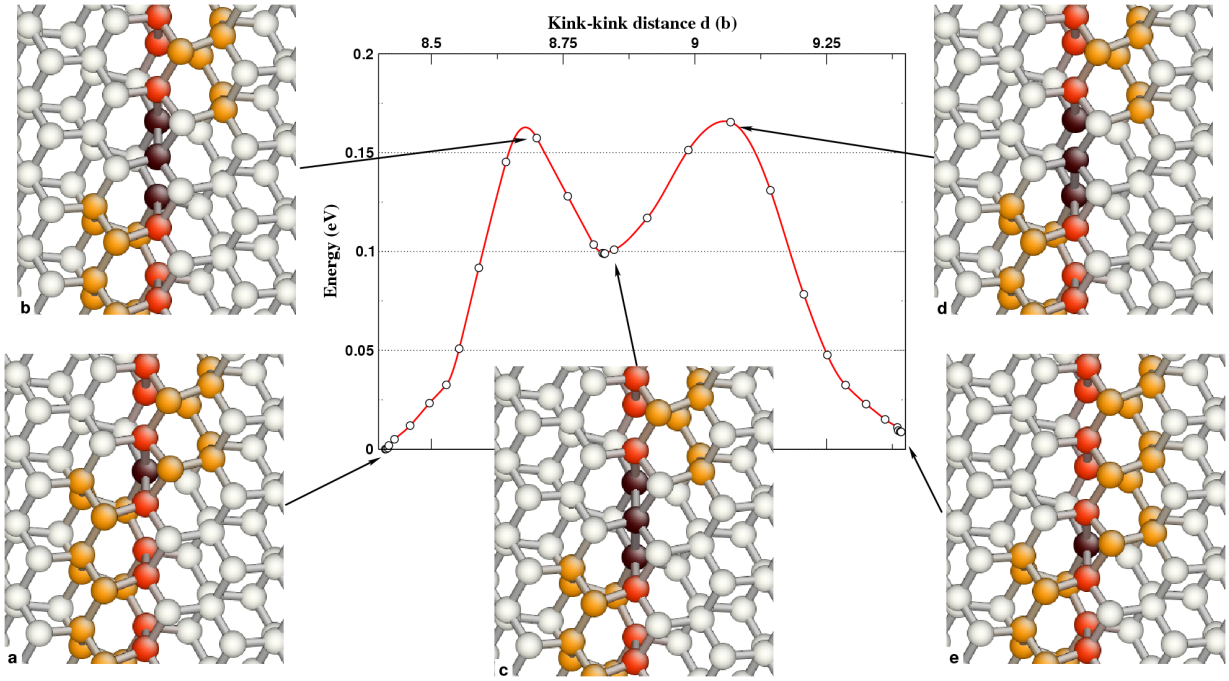

Figure 2: Minimum energy path and corresponding atomic configurations for the migration of one kink along a non-dissociated screw dislocation in silicon.

3 Mobility of dislocations: Multiplicity of mechanisms

Another example will illustrate the issue of multiple solutions for transition state. In a covalent material like silicon, it is known that dislocations are moving thanks to the formation and migration of kinks on the dislocation line. These atomic-scale processes determine the speed of the dislocation at the mesoscale, and therefore the plastic strain rate in deformation experiments at the macroscale. The problem here is the large number of possible kink structures, since the dislocation core geometries can be rather complicated, and kinks can interact with various defects along the dislocation line. It is therefore hard to determine all possible configurations, in order to select the ones with the lowest energies. Transition state methods such as the Dimer method could be used for an extensive search, but they are usually used in combination with a semi-empirical potential description which gives contrasted results in covalent materials.

This problem is first illustrated in the case of a non-dissociated screw dislocation in silicon. An exhaustive search for determining all the possible migration mechanisms of kinks has been made, using nudged elastic band and the Dimer method [4, 5]. A typical result is shown in the Figure 2. We also discussed the case of kinks migration on partial dislocations, a situation where the issue of configuration multiplicity is even more important.

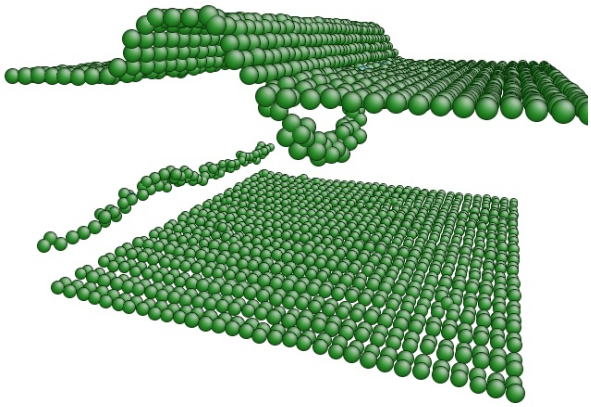

Figure 3: Half-loop dislocation nucleated from a ledge in silicon.

4 An example combining the two previous issues

The next challenge appears when we investigate a system for which the two issues above are present: the nucleation of dislocation, a rare and complex event, from a surface of silicon, a material where the number of candidate configurations is rather large. We present molecular dynamics simulations of such processes [6], obtained in conditions of temperature, strain, and strain rate all much higher than in experiments (Figure 3). To our knowledge, there is no available solution to overcome the current limitations.

5 A mechanism with no activation energy and a ”stationary” transition state ?

At last, we present a curious and specific problem related to extended defects. Some of us have recently shown that a dislocation in silicon with an orientation of 60° is mobile only when its core structure is in an unstable state [7]. The dislocation has to be nucleated first, and is displaced by stress only since thermal activation does not occur. This situation is somewhat equivalent to a process going from a state with no dislocation to a stable state where the dislocation has been transformed. The issue here is the determination of the stationary state, since both initial and final states can be very different, and cannot be used as guide to guess the structure.

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