

**Reaction pathway analysis of homogeneous dislocation nucleation in solids:
Influence of crystal structure**

(固体における転位の均質核生成の反応経路解析
—結晶構造の影響—)

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Dislocations are responsible for crystalline solids' fundamental mechanical properties of ductility and strength. Continuum based techniques such as Dislocations Dynamics (DD) are adept at dealing with dislocation mobility but cannot handle dislocation nucleation, which, being an atomistic phenomenon, can be best approached by atomistic studies. To date, most of the atomistic level simulation work done on dislocation nucleation has been on heterogeneous systems. However, there are an infinite number of configurations of heterogeneous systems, each with its own peculiar complexities, making it difficult to organize the subject in a coherent manner. The transition of dislocation nucleation cannot be studied at a fundamental level while focusing on inhomogeneous systems because the results are subject to influence by complex effects originating from heterogeneities such as stress fields, surfaces, interfaces, and other defects, which distort the physical picture. Also, the activation energies reported in such studies are presented as functions of nominal stresses, whereas the actual stresses at the dislocation core are much higher than those reported because of the presence of heterogeneities. To observe the phenomenon of dislocation nucleation in a fundamental way, free from complex effects such as stress concentrations, surfaces, etc., analysis of homogeneous dislocation is necessary. Homogeneous dislocation nucleation has also assumed great importance in view of the recent focus on nanoindentation as a means of ascertaining the onset of plasticity in single crystals. From this point of view, there have been efforts of understanding dislocation nucleation using Molecular Dynamics (MD). A serious drawback of MD, however, is the limited time-scale at its disposal. Consequently, very high strain-rates become necessary, resulting in a wide separation from the experimental time-scale of dislocation nucleation, which, being a stress-mediated thermally activated transition, occurs at stress levels significantly lower than the athermal stress.

The Nudged Elastic Band (NEB) method is a reaction pathway sampling algorithm that makes it possible to directly determine activation parameters (activation energy and activation volume) by searching the minimum energy path (MEP) of the transition. The highest point on the MEP is called the saddle-point and the energy difference between the saddle-point and the initial configuration is

the activation energy input required in the form of thermal fluctuation if the transition is to occur.

In this study, using the NEB method we focus on homogeneous crystals of various classes of materials and this allows us to examine the problem of dislocation nucleation at a fundamental level, where complex surface and stress concentration effects are not allowed to distort the physical picture. Once a coherent methodology is established for predicting the favorability conditions for dislocation nucleation, the nucleated dislocation can be handed over to DD for further analysis, in the field of semiconductor and MEMS design.

The first application of the NEB method in the domain of homogeneous dislocation nucleation was Boyer's work on perfect crystal Cu. After reproducing Boyer's work on Cu, reaction pathway analysis for homogeneous dislocation nucleation in Si and Mo is carried out. Activation energy, activation volume, and the mechanics of the dislocation core are examined for all cases with the aim of finding out the influence of lattice structure on the phenomenon of dislocation nucleation. Activation volume assumes great importance because of the fact that unlike activation energy, it can be determined experimentally. The second aim of this research is to compare the atomistic results obtained with results based on the Peierls-Nabarro (PN) model in order to ascertain the limitations of approximate solutions such as the PN framework. Thirdly, this research seeks to link homogeneous dislocation nucleation with heterogeneous dislocation nucleation by comparison with one kind of heterogeneous system, namely dislocation from a sharp corner. Finally, this research also aims to deepen insight into the old but recently reinvigorated glide-shuffle debate in Si.

In total, this thesis is composed of 7 chapters. The contents of each chapter are summarized as follows.

Chapter 1: Introduction

In Chapter 1, the background of dislocation nucleation in which the present research is set is introduced, the aims of the thesis are listed, and the structure of the thesis is explained.

Chapter 2: Method

In Chapter 2, the simulation tools used in this research are introduced. The major simulation algorithm used in this research is the NEB method and other extended schemes, which are explained. However, classical atomistic calculation tools such as molecular dynamics (MD) and Conjugate Gradient (CG) relaxation are also extensively employed for preparing input images for the NEB algorithm.

Chapter 2 also contains details of artificial loop insertion, the methodology of applying the resolved shear stress on the slip-plane, interatomic potentials employed, and the visualization technique used throughout the thesis to show the mechanics of atomistic saddle-point configurations.

Chapter 3: Models and results

Chapter 3 contains the simulation results along with the model details for dislocation nucleation in face-centered cubic (FCC) Cu, body-centered cubic (BCC) Mo, and nucleation of dislocation of the shuffle-set and glide-set in diamond-cubic (DC) Si. In this chapter, simulation results of all calculations are presented without analysis, while the analyses are carried out using these results in the subsequent chapters that deal with the different aims of this thesis.

Chapter 4: Comparison between Si, Cu and Mo

Chapter 4 documents the comparison of homogeneous dislocation nucleation in three representative materials corresponding to three common lattice structures: FCC Cu, BCC Mo, and DC Si. The comparison is presented in the form of the activation energies, the mechanics of the saddle-point configurations and the activation volumes.

Atomistic results show that approximate indicators thought to be closely related to dislocation nucleation, such as the product of the unstable stacking fault energy and square of the Burgers vector, can be misleading even in the qualitative sense because they give higher activation energy for dislocation nucleation for Si than Mo, which is contrary to the atomistic results. Atomistic calculations are therefore necessary for accurate description of the phenomenon. Also made clear by atomistic calculations is that dislocation nucleation is a two-plane phenomenon in Cu, whereas depending upon the stresses involved it is accompanied by considerable atomistic displacements in planes above and below the slip plane in Mo as well as Si. Therefore, while the structural definition, which consists of integrating the contribution of activation volume from particles confined to the slip-plane, gives accurate results for Cu, its results can be misleading for Mo and Si.

Chapter 4 also presents the comparison of homogeneous dislocation nucleation with heterogeneous dislocation nucleation with an aim to improve the understanding about the link between the two fundamental types of dislocation nucleation. Since there are an infinite number of scenarios of heterogeneous dislocation nucleation, a few representative ones are chosen from literature and compared with homogeneous systems in terms of activation volumes and mechanics of the dislocation cores. The activation volumes are of the same order of magnitude for the two types of dislocations, with those for homogeneous dislocation nucleation slightly higher than the heterogeneous case. The reasons behind this fact are the difference between the shapes and radiuses of dislocation loops, as well as the surface effects.

Chapter 5: Comparison with PN based models

In Chapter 5 the atomistic results from the last chapter are compared with results of Peierls-Nabarro (PN) based hybrid model for homogeneous dislocation nucleation. The limitations of the PN model

are highlighted along with the information that can only be made available by fully atomistic approaches. The P-N model results overestimate the atomic displacements at the saddle-point, and overestimate the activation energy requirements by a factor of 2 for Si and 5 for Cu.

The P-N based models, not being fully atomistic in nature, cannot accurately provide discrete atomistic details such as shape of the dislocation core, the presence or otherwise of extra slip-plane displacements and the actual directions of atomistic displacements. Atomistic simulation is therefore irreplaceable in order to get insight into the phenomenon of dislocation nucleation.

Chapter 6: The shuffle-glide controversy in Si

Chapter 6 deals with the shuffle-glide controversy in Si in terms of the activation energy, activation volume, and the mechanics of the dislocation cores in homogeneous dislocation nucleation. The role of mobility is also touched upon, along with comparison with results for heterogeneous dislocation nucleation regarding the shuffle-glide debate. The presence of two nucleation regimes is confirmed, with the cross-over point at 6.5 GPa. Nucleation accounts for bulk of the reaction pathway in the shuffle-set. In glide-set on the other hand, nucleation accounts for minor portion of the reaction pathway, but the low mobility of glide-set results in undulations in the energy curve. The inadequacy of the structural definition of activation volume is again proved, even more in the case of shuffle-set than the glide-set, in this chapter.

Chapter 7: Conclusions and future directions

In this chapter, the conclusions drawn from the thesis are summarized along with possible directions of future research and applications of atomistic simulation in the field of dislocation nucleation.