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A modified synthetic driving force method for molecular dynamics simulation of grain boundary migration

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ABSTRACT

The synthetic driving force (SDF) molecular dynamics method, which imposes crystalline orientation-dependent driving forces for grain boundary (GB) migration, has been considered deficient in many cases. In this work, we revealed the cause of the deficiency and proposed a modified method by introducing a new technique to distinguish atoms in grains and GB such that the driving forces can be imposed properly. This technique utilizes cross-reference order parameter (CROP) to characterize local lattice orientations in a bicrystal and introduces a CROP-based definition of interface region to minimize interference from thermal fluctuations in distinguishing atoms. A validation of the modified method was conducted by applying it to simulate the migration behavior of Ni ⟨100⟩ and Al ⟨112⟩ symmetrical tilt GBs, in comparison with the original method. The discrepancies between the migration velocities predicted by the two methods are found to be proportional to their differences in distinguishing atoms. For the Al ⟨112⟩ GBs, the modified method predicts a negative misorientation dependency for both the driving pressure threshold for initiating GB movement and the mobility, which agree with experimental findings and other molecular dynamics computations but contradict those predicted using the original method. Last, the modified method was applied to evaluate the mobility of Ni Σ5 ⟨100⟩ symmetrical tilt GB under different driving pressure and temperature conditions. The results reveal a strong driving pressure dependency of the mobility at relatively low temperatures and suggest that the driving pressure should be as low as possible but large enough to trigger continuous migration.

1. Introduction

Grain boundary (GB) migration plays a key role in governing the microstructure evolution in polycrystalline materials during recrystallization and grain growth [1]. While experimental studies have provided remarkable insights into various migration behavior and mechanisms [2–8], they are often inadequate for a comprehensive evaluation under various conditions. This is essentially related to difficulties in preparing pure bicrystal specimens with specific crystallographic orientation and boundary geometry, and difficulties in exploring migration kinetics under controlled driving forces [1]. Atomistic computer simulation based on molecular dynamics (MD) can overcome such deficiencies and it has been well received as an effective means to complement experiments in understanding GB migration behavior.

Existing MD techniques for GB migration simulations can be sorted into two categories: "fluctuation" methods and "driving force" methods. In the first category, investigation into GB migration is realized by analyzing thermal fluctuations of a boundary, in the absence of an external driving force [9,10]. This kind of method may not be applied to low temperature and is not suitable for investigating migration kinetics [11,12]. In the second category, a boundary is driven to move under controlled driving forces, which can arise from GB curvature [3,13,14] and elastic anisotropy [15–17] or be related to crystallographic orientations [18–20]. With a curvature-driven technique, a U-shaped GB is often constructed to provide the driving force for its motion. This technique accounts for only three out of the five crystallographic parameters (i.e. misorientation) of a GB and extracts a reduced GB mobility. By contrast, the other two driving force-based techniques consider all five crystallographic parameters of a GB. Simulation of GB migration due to elastic anisotropy imitates a real physical process, but the applied elastic strain cannot provide driving force for symmetric boundaries [12,21]. In simulations utilizing crystallographic orientation-correlated driving force (OCDF) [18–20], the driving force arises

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from artificial potential energies added to atoms according to their local orientations in a bicrystal system. It has been shown that OCDF-based methods can be conveniently applied to simulate GB migration under desired temperature and driving force conditions [22].

The implementation of OCDF by Janssens et al. [20], which is often known as the synthetic driving force (SDF) method, has been applied to investigate various aspects of GB migration, e.g. boundary roughening [23], shear coupled migration [22,24] and grain growth stagnation [25]. In this implementation, the driving forces for boundary motion are imposed after distinguishing atoms in grains and GB, according to their local orientations characterized by order parameters in conjunction with two adjustable cutoff values [20]. Despite the revealing results obtained with this method, cautions have been raised about its limitation in applications and defects of the technique to distinguish atoms. Olmsted et al. [22] warned that this method is effective only when the boundary misorientation is large enough such that the local structural difference between the two perfect crystals is greater than the typical differences due to thermal fluctuations. Similarly, Rahman et al. [26] advised that considerable cares should be taken in applying the SDF method to low-angle GBs, especially at high temperatures. More specifically, Zhou and Mohles [27] pointed out that the limitation of this method could be related to improper distinction of atoms in grains and GB, which may lead to flawed energy and force distributions in the bicrystal.

Several attempts have been made to overcome the deficiencies of the SDF method stated above. First, Zhou and Mohles [27,28] suggested that using only one set of order parameters (as adopted in the original SDF method) is insufficient to distinguish thermal displacements from those caused by crystal reorientation and they proposed to use two sets of order parameters. Although their modified SDF method was criticized for its controversial definitions of atomic energy and forces [29,30], it seems to bring some improvement in avoiding misidentification of atoms for some cases. In a follow-up work by Ulomek and Mohles [31], it was argued that the original SDF method with only one set of order parameters results in an asymmetric transition of order parameter in the GB area and hence an asymmetric profile of the artificial energy in the bicrystal. They applied an order parameter analogous to that considered in the capillary fluctuation method [9] to generate the so-called symmetrical driving forces for GB migration. However, their method yielded the same mobility results as the original approach within the margin of error [31]. Moreover, similar to that of the original SDF method, the GB migration results obtained using these modified methods [27,28,31] depended strongly on the cutoff values that were arbitrarily chosen without necessary justifications. Furthermore, for cases that atoms in grains and GB have similar order parameter values, which are typical at high temperatures due to significant thermal fluctuations, it is impossible to distinguish these atoms by comparing only their order parameters with the cutoff values. A robust technique with sufficient generality to identify atoms in grains and GB under the influence of thermal fluctuations, which is critical in developing the SDF method as a dependable tool to investigate GB migration, is still lacking.

Another major concern in applying the SDF method for GB migration simulations is the magnitude of applied artificial driving pressure (i.e. a global value of the individual driving orces). In experimental and force-driven computational studies [1,13,16,17], the GB mobility M is often extracted by assuming a linear relation between migration velocity V and driving pressure P in the low limit of P (i.e. $M = \mathrm{d}V/\mathrm{d}P|_{P\to 0}$). However, due to constraints on the computational expense in typical MD simulations, the driving pressure generally applied in the SDF method is around 10–400 MPa [20,22,23,26,32,33], which are orders of magnitude higher than experimentally applied values (10^{-4}–1 MPa) [1,34]. Under such high driving pressures, the migration velocity can be as large as several hundreds meters per second (typical experimental value is 10^{-8}–10^{-3} m/s [1,34]) and it often varies nonlinearly with the driving pressure. The resulting activation energies for GB migration are often significantly lower than those determined from experiments [22,23,26,32,33]. Moreover, an unrealistically high driving pressure has also been observed to alter GB migration mechanism [11] and lead to somewhat surprising results, e.g. zero mobility for $\Sigma 3$ boundary with plane normal $\langle 1\,1\,1\rangle$ while quite high mobility for other $\Sigma 3$ boundaries [20,22]. Under these circumstances, it is no longer certain that the mobility obtained from the MD simulations is a reasonable approximation of the intrinsic mobility. It is necessary to investigate the proper range of the driving pressure to consider in practical applications of the SDF method.

In the present work, a modified SDF method has been developed after evaluating the deficiency of the SDF method in distinguishing atoms. The modified method was applied to investigate the migration behavior of several Ni $\langle 1\,0\,0\rangle$ symmetrical tilt GBs (STGBs) and Al $\langle 1\,1\,2\rangle$ STGBs. The results are compared with those obtained using the original method to reveal the significance of the present modification. Principles for selection of driving pressure in extracting intrinsic mobility are discussed based on a detailed examination of the effect of driving pressure on the mobility of Ni $\Sigma 5$ $\langle 1\,0\,0\rangle$ STGB at different temperatures.

2. Method

2.1. Principles of SDF-based method for GB migration

During recrystallization and grain growth, boundaries are driven to move under forces arising from the free energy difference between two neighboring grains [1]. In a bicrystal system structured with a MD technique, the free energy difference caused by the structure difference between adjoining grains and thermal noises are insufficient to drive a boundary to migrate continuously, if no other sources of free energy difference (e.g. chemical or mechanical ones) are available.

In the SDF method [20], the free energy difference is enlarged by artificially adding an orientation-dependent potential energy difference to the bicrystal, e.g. adding a potential energy to one grain while not to another, and adding an intermediate energy to GB. A clear distinction of atoms in grains and GB indisputably is necessary before adding the artificial potential energy and thus providing driving forces for GB migration. To this end, Janssens et al. [20] defined an order parameter ξ_i for each atom to characterize the deviation of an actual local structure from a reference structure. In a bicrystal consisting of grain A with orientation I and grain B with orientation J, the order parameter for atom i is defined as [20]

$$\xi_i = \sum_{j=1}^{n} |\mathbf{r}_j - \mathbf{r}_j^I|, \tag{1}$$

where $\mathbf{r}_j$ is a position vector of nearest-neighbor atom j of atom i, and $\mathbf{r}_j^I$ denotes the ideal relative vector in the reference grain A. n is 12 for face-centered cubic materials. ξ_i is expected to be small for atoms in the reference (or favored) grain, large for atoms in the non-reference (or unfavored) grain, and intermediate for atoms in the GB. Then, atoms in grains and GB are distinguished based on their order parameters by introducing a pair of cutoffs [20], $\xi_{low} = f\xi_{IJ}$ and $\xi_{high} = (1-f)\xi_{IJ}$, where ξ_{IJ} characterizes quantitatively the orientation difference between the two grains at 0 K ($\xi_{IJ} = \sum_{j=1}^{n} |\mathbf{r}_j^J - \mathbf{r}_j^I|$) and f is a parameter to adjust the cutoffs.