Molecular dynamics simulation of the effect of cyclic stresses on nanocrystals with nonequilibrium grain boundaries: the role of the grain size

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Abstract. Grain boundaries (GBs) in bulk nanostructured materials processed by severe plastic deformation (SPD) have a nonequilibrium structure caused by extrinsic grain boundary dislocations (EGBDs) absorbed during deformation. Under external influences (annealing or cyclic straining) these GBs relax towards equilibrium resulting in a release of the excess energy of nanomaterials. Using our earlier developed method, atomic models of nanocrystals with nonequilibrium GBs having grain sizes of 10, 15, and 20 nm are constructed and relaxed by molecular dynamics. Then, the effect of oscillating tension-compression stresses with amplitudes of 2, 3 and 4 GPa on the systems thus obtained is simulated. Relaxation of nonequilibrium GBs by dislocation emission is found and the effect is shown to be independent of the grain size.

1. Introduction
The interaction of grain boundaries (GBs) with lattice dislocations results in the formation of extrinsic grain boundary dislocations (EGBDs) and a specific structure of GBs, which is commonly referred to as a nonequilibrium structure [1]. The nonequilibrium GBs (NGBs) induce long-range internal stress fields and possess an excess energy as compared to equilibrium GBs of the same geometry [2]. The nonequilibrium GB structure was shown to be typical for bulk nanostructured materials processed by severe plastic deformation (SPD) and to largely determine their properties [3]. For better understanding of the effect of NGBs on the properties of materials, it is necessary to know their atomic structure.

Recently, we have developed a method for the construction of initial structures for molecular dynamics (MD) simulations of nanocrystals with NGBs and studied the atomic structure, and energy of a model nickel [112] columnar nanocrystal with NGBs [4,5]. Also, the behavior under cyclic stresses was studied for the case of a nanocrystal with the grain size of \( d = 15 \) nm.

The present paper aims to study the role of grain size in the behavior of a [112] columnar nanocrystal with NGBs under cyclic stresses.

2. Model description and simulation procedure
The geometry of a computation cell containing four columnar grains with crystallographic axes [112] parallel to the \( z \) direction of a coordinate frame is presented in figure 1a. The grains have a regular hexagon-shape cross section with an edge \( a \). The maximum size of the hexagons, \( d = 2a \), will be
taken as the grain size. Grains 1 and 2 have orientations of slip planes (11T) parallel to xOz and yOz planes, respectively. Taking the orientation of grain 1 as a reference, grains 3 and 4 are rotated to angles $\pm \alpha = \pm 25^\circ$ to form a symmetric high-angle tilt GB between them. To initialize the structures with NGBs, the construction method proposed in [4] is used. This method is based on an artificial shear strain of selected grains of the nanocrystal to introduce EGBDs into its boundaries. In order to form EGBDs on the boundaries of the selected grain, it is assumed to be removed from its place, freely deformed by plastic shear $\gamma$, then strained back elastically to $-\gamma$ and embedded into the polycrystal. In figure 1 a, grains 3 and 4 are deformed in this way. Then the system with EGBD precursors is subjected to energy minimization with special precautions preventing the EGBDs’ annihilation. As an example, presented in figure 1 b is the initial structure of a nanocrystal constructed for the grain size $d = 5$ nm and shear strain $\gamma = 0.15$ of grains 3 and 4.

![Figure 1](image)

**Figure 1.** Computational cell construction for [112] columnar nanocrystals with NGBs (a) and initial structure of a nanocrystal with grain size $d = 5$ nm and shear strain $\gamma = 0.15$ of grains 3 and 4 (b).

Using the method described above, the initial structures of nanocrystals with grain sizes $d = 10$, 15 and 20 nm and shear strain $\gamma = 0.10$ of grains 3 and 4 were constructed for further studies. Simulations were carried out for nickel using embedded atom method (EAM) potential developed in [6]. The corresponding lattice parameter is equal to $a_0 = 0.352$ nm and the potential energy per atom $E_{a0} = -4.45$ eV. The period of the computational cell along the z-axis was fixed equal to $H_z = 3a_0 / 2[112]| 1.293$ nm for $T = 0$ K and scaled by a factor of 1.0044 at $T = 300$ K to account for thermal expansion. Periodic boundary conditions with a constant pressure were applied along the other two directions (zero along x and zero or $p$ along y). To obtain relaxed configurations of the systems, an energy minimization was carried out first keeping all atoms having energies $\varepsilon > -4.40$ eV (i.e. atoms on and near GBs) fixed to their positions that allowed one to avoid the annihilation of EGBD precursors. Then, a full relaxation at $T = 0$ K followed by equilibration at $T = 300$ K was carried out.

The effect of cyclic straining was simulated at $T = 300$ K, applying a periodic tension-compression stress with a period of 160 ps along the y-axis. For this, the period was divided into 40 intervals, in each of which the value of the stress changed from $p_{k-1} = p_0 \sin[2\pi(k - 1)/40]$ to $p_k = p_0 \sin[2\pi k / 40]$ in each of the $k$-th interval ($k = 1, 2, \ldots, 40$) and for each interval of the stress 2000 steps of MD run with time step 2 fs were done. Three values of the stress amplitude, $p_0 = 2$, 3 and 4 GPa were taken for simulations.

Simulations were carried out using the LAMMPS MD code [7], visualization and common-neighbor analysis (CNA) [8] of the structures were done by OVITO [9]. Events of lattice dislocation emission, changes in the sizes of computational cells $H_z$ and $H_x$, and of the potential energy of atoms $E_a$ with time were analyzed.
3. Simulation results and discussion

Figures 1 a,b present the results of the CNA of the atomic structures of a nanocrystal with the grain size $d = 10$ nm with the nonequilibrium structure of GBs at 0 K and after ten periods of cyclic deformation with the stress amplitude of 4 GPa at 300 K.

**Figure 2.** Results of a CNA of the atomic structures of a nanocrystal with the grain size $d = 10$ nm: (a) with nonequilibrium GBs at 0 K; (b) after ten cycles of straining with the stress amplitude of 4 GPa at 300 K. Green atoms have a fcc environment, red ones an hcp environment, and white and blue atoms have other types of coordination.

In the figures, red atoms having an hcp environment visualize stacking faults, which are formed due to the glide of partial lattice dislocations across the grains. Therefore, the comparison of the figures shows that under the effect of cyclic stresses GBs generate lattice dislocations. Analysis shows that the latter are emitted when the applied stress has the same sign as the internal stresses induced by EGBDs, and, therefore, these dislocations result in a relaxation of the stress fields of NGBs.

The fact that the generation of dislocations results in relaxation of NGBs is clearly seen from figures 3 a-c, which demonstrate the change of atomic potential energies with time expressed in terms of the number of stress cycles $N_{\text{cycle}}$ for the three grain sizes studied. One can see that the potential energy decreases with the number of cycles. At the stress amplitude of 2 GPa, there is nearly no change of the average energy, since lattice dislocations are not emitted by NGBs. At the amplitude of 3 GPa, dislocation activity occurs in the grains and at 4 GPa it becomes higher. Accordingly, the average potential energy per atom decreases with time of stress cycling.

**Figure 3.** Dependence of atomic energies of nanocrystals with grain sizes $d = 10$ (a), 15 (b) and 20 (c) nm on the number of stress cycles; $p_0 = 2,3,4$ GPa (blue, green and red curves, respectively).

The fact that the average potential energy $E_a$ decreases due to the elimination of long range stress fields induced by EGBDs is seen from figures 4 a,b, where atomic energy maps for a nanocrystal with the grain size of 10 nm before and after cyclic deformation are presented. One can clearly see that the
atoms inside the deformed grains had an enhanced energy due to the long range elastic fields of EGBDs, and after stress cycling the atoms have energies close to that of perfect crystal atoms due to the compensation of the stress fields by the emission of dislocations.

**Figure 4.** Atomic energy maps of nanocrystals with the grain size of $d = 10$ nm in the state with NGBs (a) and after ten cycles of straining with the stress amplitude of 4 GPa (b) and energy scale (c) to demonstrate the relaxation of long range stress fields.

Figure 3 also shows that, despite a two times difference between the minimum and maximum grain sizes of the studied nanocrystals, there is no apparent change of the stress amplitude necessary for initiating the dislocation emission by NGBs: in all three cases, the generation of dislocations occurs at an amplitude between 3 and 4 GPa. Therefore, the stresses necessary for an emission of individual dislocations from GBs, NGBs as well, depend mainly on their local atomic structure rather than on long range stress fields. However, the latter bring an asymmetry to the dislocation emission process under cyclic stresses resulting in the possibility of structure relaxation under completely symmetric stress oscillations.

### 4. Conclusions

Molecular dynamics simulations show that oscillating applied stresses can result in relaxation of the nonequilibrium structure of GBs in nanocrystals by emission of partial dislocations, which compensate the stress fields of EGBDs. This process occurs at the stress amplitudes above a certain lower bound, which for the present case is between 2 and 3 GPa. This lower bound seems to be independent of the grain size of nanocrystals. Therefore, the relaxation of NGBs by dislocation emission is controlled by the local structure of GBs rather than by their long range stress fields.

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