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Microarticle

Rotation of micrometer-sized grains in cyclically loaded thin gold films at room temperature

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ABSTRACT

Room temperature grain rotation in polymer-supported gold films subjected to cyclic tensile loading is directly demonstrated by quasi-in-situ electron backscatter diffraction analysis. Grain rotation is activated after saturation of grain coarsening. Rotating grains try to align themselves with their (1 0 0) direction parallel to the axis of applied mechanical strain which corresponds to the lowest elastic modulus.

Grain rotation as a response to applied mechanical loading is a well-known phenomenon which was frequently reported to occur in nanocrystalline metals where traditional plasticity mechanisms based on nucleation and motion of bulk dislocations are restricted [1–8]. Despite the large number of experimental and computational evidences of grain rotation as an alternative plasticity mechanism it is still unclear when and why it occurs. On the one hand, there is a universal answer to the question "Why a particular grain has rotated in a particular way?" which is "In order to reduce the total free energy in a particular thermodynamic state, i.e. in a mechanically stressed state". On the other hand, it is often hardly possible to provide a more detailed answer explaining the exact driving force leading to a particular grain rotation event in a polycrystalline sample. Even if a well-defined mechanical strain is applied, the local stress/strain state in the vicinity of a particular grain is, to large extent, unknown. So for instance, in a detailed in-situ transmission electron microscopy (TEM) experiments on free-standing notched Al films [6] multiple grain rotation events were observed and characterized. However, no clear conclusion about the driving forces leading to the particularly observed rotations was provided.

Here we consider polymer-supported thin gold films with initially ultra-fine grains which are subjected to cyclic mechanical strain. The films were deposited on 50 µm polyimide UPILEX S substrates by electron beam evaporation in a Balzers BAK 550 evaporation machine with the vacuum of 2.1×10^{-7} mbar and using a deposition rate of 0.3 nm/s. The test samples with the width of 4 mm and length of 40 mm were cut using a scalpel out of larger sheets and subjected to cyclic strain by applying a sine strain-time function with the frequency of 0.5 Hz. It was shown in a recent publication that strong room temperature grain coarsening occurs in such films as a response to cyclic strain [9]. The average grain size increases when cyclic strain is applied but, after some number of applied cycles (approximately 5000 for 1% strain amplitude), the grain coarsening stagnates, as demonstrated in the Supplementary Fig. S1. In Fig. 1a and 1b the electron backscatter diffraction (EBSD) grain orientation maps of the same surface area of 500 nm thick gold films after 5000 and 10,000 cycles with 1% strain, respectively, is demonstrated. Although some grain boundary migration occurs between 5000 and 10,000 cycles (an example is marked by the white arrow in Fig. 1a), the average grain size does not change significantly, i.e. the stagnation of grain coarsening is reached. At the same time, several grains have rotated as can be seen by the change in color comparing Fig. 1a with Fig. 1b. Three grains which have changed their orientation most significantly are marked in Fig. 1b by white numbers. As can be seen from the comparison of elastic stiffness maps after 5000 cycles (Fig. 1c) and 10,000 cycles (Fig. 1d), the rotation of these three grains led to substantial decrease of their elastic modulus. The elastic stiffness maps were calculated in EDAX OIM software using the elastic constants for gold ($C_{11} = 193$ GPa, $C_{44} = 41.5$ GPa, $C_{12} = 164$ GPa) from [10].

In order to assure that the observed decrease of local elastic modulus by grain rotation is not an occasional coincidence, large statistical data was collected by comparing the inverse pole figures (IPFs) of different samples with increasing number of applied cycles. The results are summarized in the Supplementary Fig. S2 confirming that there is a general trend of grains rotating towards (1 0 0) direction parallel to the axis of applied strain which corresponds to the orientation with the lowest elastic modulus in gold.

The observed direction of grain rotation can be easily explained by basic thermodynamics considerations. With the lowest possible Youngs modulus the elastic strain energy density within the grains will be also minimized thus decreasing the amount of free energy in the system under mechanical load. Although we observe a clear *correlation* between the grain rotation direction and minimization of the elastic modulus, it does not automatically mean the existence of a *direct causal relationship* between them. In other words, we cannot directly prove that grain rotation is not caused by another driving force and the reduction of the elastic modulus is the consequence of it. Nevertheless, the statement that the observed direction of grain rotation leads to

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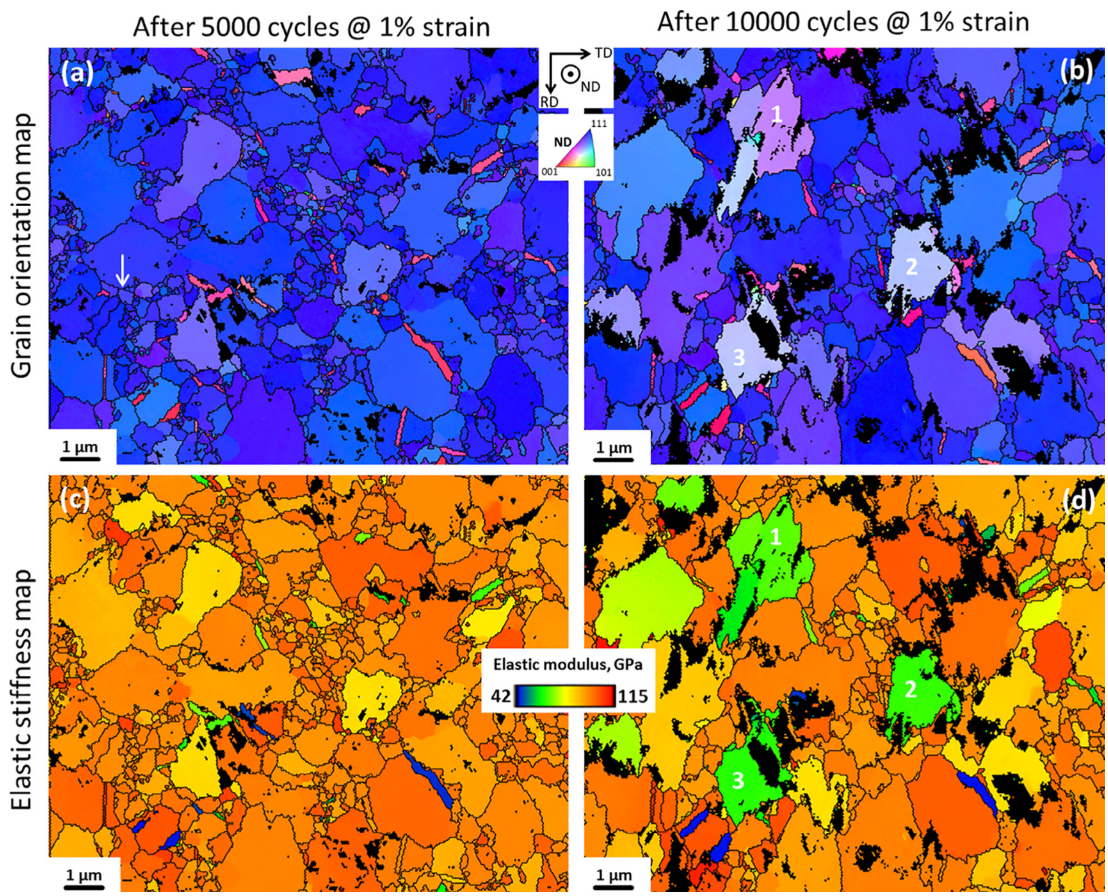

Fig. 1. EBSD grain orientation maps (a, b) and elastic stiffness maps (c, d) of a 500 nm thick Au film thermally evaporated on polyimide substrate after applications of 5000 cycles (a, c) and 10,000 cycles (b, d) with 1% strain. The white numbers in (b, d) depict three examples of rotating grains. The white arrow in (a) shows an example of a grain boundary which has migrated during further cyclic loading. Black areas in all maps correspond to the scan points which were not properly indexed by EBSD software. Higher amount of black areas in (b, d) is caused by the surface roughening due to the formation of fatigue-induced extrusions.

minimization of elastic strain energy density during cyclic loading holds true.

Presented results clearly demonstrate that grain rotation phenomenon is not a property of nanocrystalline materials but is also observed in polycrystals with micrometer-sized grains. Strong correlation between grain rotation and decrease of the individual Young's modulus of the rotating grains suggests that the driving force for the observed grain rotation is minimization of the elastic strain energy in mechanically strained state. On the basis of present results and the results of [9] one can generally conclude that dislocation-based plasticity in ultra-fine grained films under cyclic loading is not restricted to accumulation of dislocation slip events but can appear in the form of grain boundary migration, grain coarsening, and grain rotation.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rinp.2019.102616.

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