Analyzing Structural Dynamics in Nanocrystalline Thin Films using In-Situ 4D-STEM: A Statistical Approach

Yuan Tian[1], Yutong Bi[1], Mingjie Xu[2], Evgeniy Boltynjuk[3], Horst Hahn[3], Jian Han[4], David J Srolovitz[5], and Xiaoqing Pan[1,2,6,*]

[1]*Department of Materials Science and Engineering, University of California, Irvine, CA, USA*
[2]*Irvine Materials Research Institute (IMRI), University of California, Irvine, CA, USA*
[3]*Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany*
[4]*Department of Materials Science and Engineering, City University of Hong Kong, Hong Kong SAR, China*
[5]*Department of Mechanical Engineering, The University of Hong Kong, Hong Kong, China*
[6]*Department of Physics and Astronomy, University of California, Irvine, CA, USA*

*Corresponding author: xiaoqing.pan@uci.edu

Traditional in situ transmission electron microscopy (TEM) techniques are highly regarded for their exceptional resolution and the detailed insights into the microstructural dynamics of materials at the nanoscale. However, a significant limitation of these methods is their lack of statistical robustness and representativeness for the materials being studied [1]. In recent years, four-dimensional scanning transmission electron microscopy (4D-STEM) has emerged as a method capable of gathering more comprehensive information across a broader field of view [2] The integration of 4D-STEM with in situ methodologies has made it into a powerful tool for examining material dynamics [3-4]. In this work, we have developed an in situ 4D-STEM technique specifically for the statistical analysis of the structural dynamics in nanocrystalline thin films. The statistical data obtained from in situ 4D-STEM represents a crucial enhancement to traditional in situ TEM methods, providing a more representative understanding of the materials under investigation.

Figure 1(a, b) shows two consecutive orientation mappings, derived from in situ 4D-STEM datasets, of the same area in a Pt nanocrystalline thin film subjected to different times of annealing. Notably, the annealing process induces significant grain growth. By applying intraframe grain segmentation and interframe grain association, we successfully constructed the evolution maps of each grain in the field of view. An example is illustrated in Figure 1(c-f), which demonstrate the evolution of grain morph-ology, diffraction pattern, grain size and in-plane orientation respectively. We can find that the grain shows not only the change of grain size, but also the change of orientation. This suggests the occurrence of grain rotation. Further analysis reveals that over 10% of the grains undergo rotation with the rate of 2 °/min.

With the extraction of evolution maps for all grains, statistical analysis can be conducted on the microstructural dynamics. As shown in Figure 2(a), we plotted the probability distribution of relative grain size changing rate, $\frac{|\dot{r}|}{r}$, for all grains (in blue bars) and for significantly rotating grains (in orange bars). The ratio of them, plotted in red dots, elucidates the correlation between of grain rotation probability and the relative grain size changing rate. Initially, this probability increases, then stabilizes as the rate of grain size change reaches a certain threshold. This pattern suggests that grain rotation is concurrent with grain growth/ shrinkage and reaches a saturation point at specific growth/ shrinkage rates. Additionally, the spatial distribution of rotating grains, analyzed through the pair distribution function (PDF), shows a small bump for the rotating grains. This indicating a propensity for rotating grains to cluster spatially, i.e., grain rotation predominantly occurs among neighbor grains.

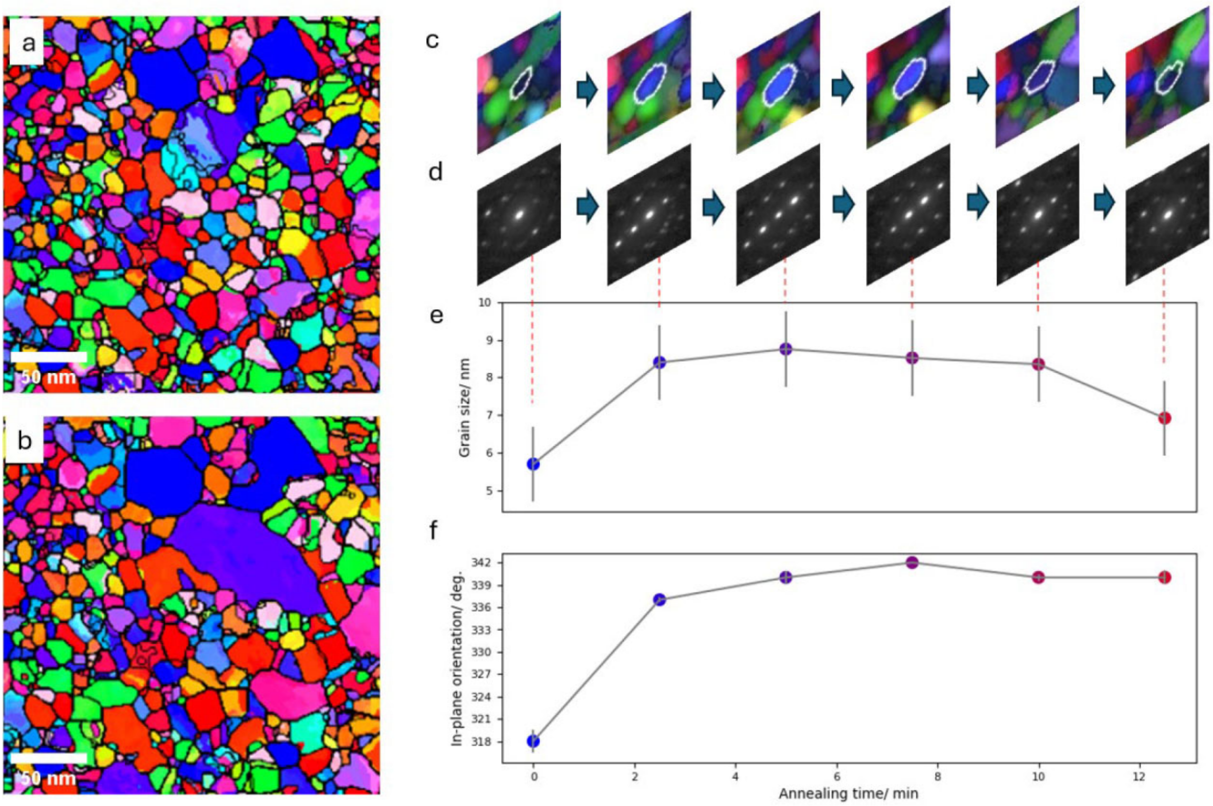

Fig. 1. (a, b) two consecutive orientation mappings taken from the same area of Pt thin film after annealing for 10min and 15min respectively. (c f) The evolution of grain morphology, diffraction pattern, grain size and in plane orientation respectively during the annealing process.

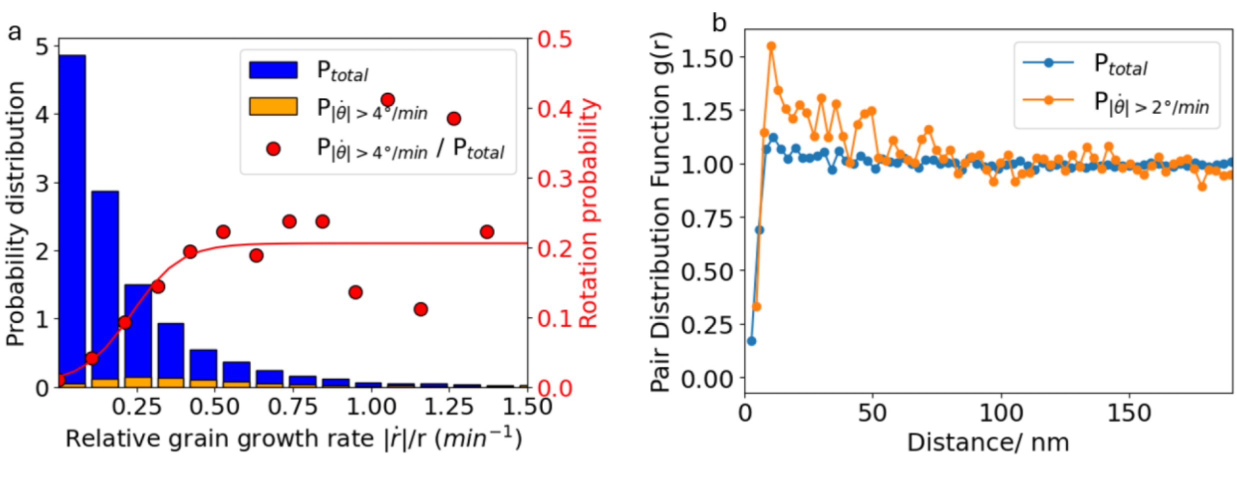

Fig. 2. (a) The probability distribution of relative grain growth rate, $\frac{|\dot{r}|}{r}$, for all grains (blue) and for significantly rotating grains (orange). The ratio of yellow bars and blue bars (red dots) represents the dependence of rotation probability on the relative grain growth rate. (b) The pair distribution function of all grains (blue) and rotating grains (orange).

References

1. M Legros *et al.*, *Nat. Mater.* **23** (2024), p. 20.
2. C Ophus, *Microsc. Microanal.* (2019), p. 563.
3. A Minor, *Microsc. Microanal.* **26** (2020), p. 2019.
4. TC Pekin *et al.*, *Nat. Commun.* **10** (2019), p. 1.