

Effect of Medium Range Order on Crystallization Kinetics of $\text{Cu}_x\text{Zr}_{1-x}$ Thin Film Metallic Glasses

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In this study we investigate how subtle changes in medium range order (MRO) in diffraction-amorphous $\text{Cu}_x\text{Zr}_{1-x}$ alloys alter their crystallization kinetics. CuZr metallic glasses (MG) have gathered much attention due to their unique mechanical properties. It is known that although lacking long-range order, CuZr MG possesses ordered regions in the 1 – 3 nm length scale [1], or MRO, which correspond to the near-critical nuclei as predicted by classic nucleation theory. We use fluctuation electron microscopy (FEM) operated in the nanoprobe diffraction mode to measure the changes in MRO in the materials. We then correlate the MRO to the crystallization kinetics of the materials during pulse laser crystallization measured *in-situ* using the dynamic transmission electron microscope (DTEM). The structural-property relationship provides insights to the fundamentals of crystallization process, which serve a greater scientific community beyond the particular material systems.

We sputter deposit ~30 nm of $\text{Cu}_x\text{Zr}_{1-x}$ thin film MG ($x = 32\% - 80\%$) in a high vacuum chamber by varying the sputtering power during co-deposition. The crystallization pathways of this family of MG materials have been shown alter significantly across the compositional gradient [2]. We hypothesize that the changes are due to a composition-induced variation of MRO (Figure 1), which we measure using FEM on a Zeiss Libra TEM at the National Center for Electron Microscopy. We then measure the crystallization speed *in-situ* during pulsed laser annealing using the DTEM at Lawrence Livermore National Laboratory. The DTEM provides a 9-frame movie during the crystallization process with both nanosecond temporal and nanometer spatial resolution [3]. Operating the DTEM in both imaging and diffraction mode, we can identify formation of various crystalline phases in real time and correlate the observation to the MRO in the material. [4]

References:

- [1] J. Hwang *et al*, Physical Review Letters **108** (2012) 195505.
- [2] M. Apreutesei *et al*, Journal of Alloys and Compounds **619** (2015) 284-292.
- [3] G.H. Campbell, J.T. McKeown, and M.K. Santala, Applied Physical Review **1** (2014) 041101.
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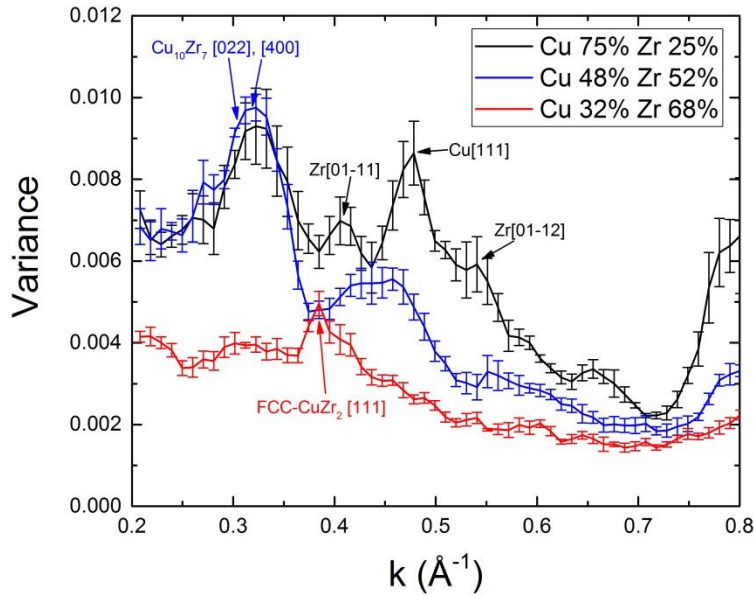


Figure 1. Fluctuation electron microscopy variance spectra versus diffraction vector (k) of various $\text{Cu}_x\text{Zr}_{1-x}$ thin films. Variance peak positions change for the different compositions. The peak positions can be matched to various Bragg conditions of the different intermetallic phases, which allow us to identify the medium range order in the material before crystallization.

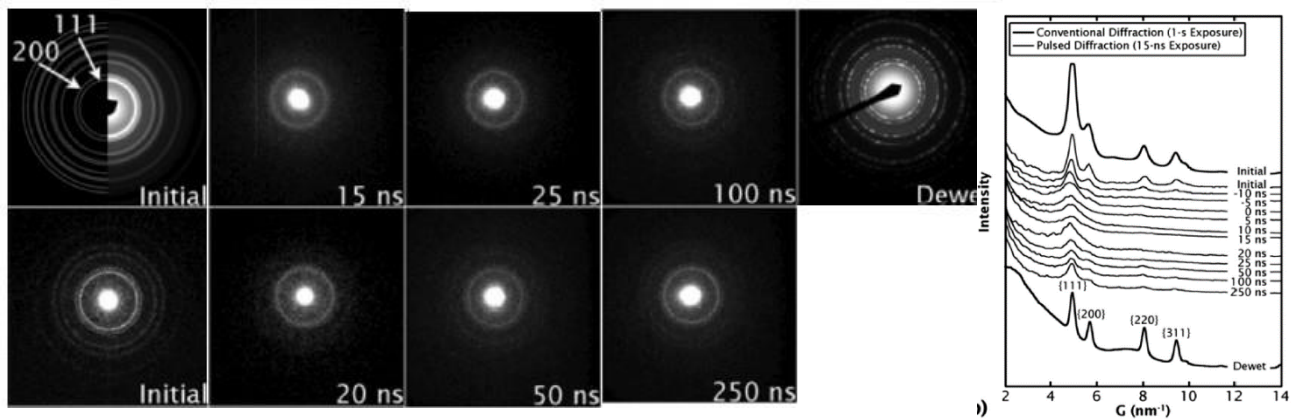


Figure 2. Time-resolved diffraction patterns captured using the DTEM during pulse laser dewetting of polycrystalline Ni. Radially averaged diffraction intensity clearly shows the evolution of melting and re-crystallization. We can determine the crystalline phases as they form, and correlated the result to observed MRO.