Dynamic Transmission Electron Microscope Study on the Crystallization of Ion-Bombarded Amorphous Germanium Thin Films

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The structure-properties relationships of amorphous group IV semiconductors such as germanium (a-Ge) have attracted wide research interests over the past decades. The materials have great technological significance thanks to a plethora of applications, and understandings of their structural properties provide key scientific insights for those with similar covalently bounded networks. Pulsed laser induced crystallization is a common method to evolve the nanostructure in a-Ge. In this study, we report the correlations between the differences in the nanostructure of a-Ge thin films created by ion bombardment, and the laser crystallization kinetics measured using dynamic transmission electron microscope (DTEM).

Previously, amorphous silicon (a-Si) prepared by different methods has have been shown to exhibit different medium range order (MRO), which gives rise to different mechanical properties and tendencies to structural relaxation under thermal annealing [1]. Changes in MRO can be measured using a statistical microscopy technique known as fluctuation electron microscopy (FEM), as shown in Figure 1. We sputter deposit ~30 nm of a-Ge thin film, and subsequently alter the nanostructure by Ar⁺ ion bombardment. We then observe the differences in MRO between the as-deposited and the treated samples using FEM.

Amorphous germanium is known to crystallize with rate as high as ~10s of m/s during laser annealing. The movie-mode DTEM at LLNL provides us the unique capability to capture the structural evolution in situ during the crystallization process [2]. From a series of (up to 9) single-shot acquisitions in under ~1 µs, we can track the movement of the amorphous-crystalline boundary (Figure 2) with temporal resolution unavailable in conventional TEM experiments. We collect the crystallization kinetics data of both as-deposited and Ar⁺-treated a-Ge samples, and map the differences to the changes in the MRO.

References
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Figure 1: FEM variances ($V(k)$) versus diffraction vector ($k$) of four different amorphous Si samples: as-sputtered (as-sputt.), sputtered then annealed (ann. sputt.), as-implanted (as-imp.), and implanted then annealed/relaxed (rel. imp.). The differences in $V(k)$ indicate the changes in the medium range order when the samples are subject to different preparation conditions and thermal treatments. Taken from [1], used with permission.

Figure 2: (Left) A series of images collected using Movie-mode DTEM during in situ laser crystallization of amorphous Ge. The images are collected with 20-ns exposure and 95 ns intra-frame time. The crystallization front moves at ~ 20 m/s. (Right) conventional TEM image of the crystallized area after laser annealing.