Detecting Nuclei in Phase Change Materials by Fluctuation Electron Microscopy (FEM): *An Experimental Proof of Nucleation Theory*

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ABSTRACT

Phase change materials such as $Ge_2Sb_2Te_5$ (GST) and AgInSbTe (AIST) exhibit rapid and reversible amorphous-to-crystalline phase transformations, which are the basis of rewritable CDs/DVDs and non-volatile phase change random access memories. The speed of these devices depends on the kinetics of phase transformation, especially crystallization. One long standing puzzle is that, for a fixed composition, different processing conditions afford amorphous states with significantly different crystallization rates, but these kinetically different states all appear amorphous and indistinguishable in conventional diffraction experiments. Here, we demonstrate that Fluctuation Electron Microscopy (FEM) can detect the existence of nuclei in an amorphous matrix, and that the quantity of such nuclei correlates directly with crystallization kinetics.

FEM employs statistical analyses of nanodiffraction patterns or dark-field images to quantify structural order on the 1-3 nm scale in amorphous materials. From FEM data we find that the population of large nuclei coarsens significantly by thermal pre-annealing under the crystallization temperature. The corresponding change of transformation kinetics is studied by pulsed laser heating with real-time monitoring of optical reflectivity. The incubation delay before crystallization is reduced (by the presence of subcritical nuclei) or eliminated (by supercritical nuclei) by pre-annealing. This is exactly what classical nucleation theory expects. Such effect is much greater in the case of AIST, where nucleation is relatively difficult compared to growth (growth-dominant), than in GST (nucleation-dominant). We also reveal the difference between as-deposited and melt-quenched amorphous states; the melt-quenched state may have nanocrystallites of several nanometers that act as supercritical nuclei, provided that the cooling rate from melt allows the formation of such nuclei.

Thus, FEM provides a quantitative means to “see” the existence of nuclei in an amorphous material. It distinguishes the amorphous phases from different origins and, combined with kinetic study, provides a beautiful proof of the postulations of classical nucleation theory.

Key words: Nucleation, subcritical nuclei, Fluctuation electron microscopy, transformation speed

Biography

Dr. Bong-Sub Lee received his Ph. D. in Materials Science and Engineering at University of Illinois at Urbana-Champaign in USA, and his M.S. and B.S. with honor at Seoul National University in Korea. He received several scholarships, and also a department honor for excellence in academic performance at Seoul National University. Currently he is a postdoctoral research associate at University of Illinois at Urbana-Champaign. He also works in close collaboration with IBM Almaden Research Center, where he was invited as visiting scientist for two occasions. His interest on phase change materials started from the optical and electronic properties, and now he is mainly studying their nanoscale structure and transformation speed.