

In-situ Observations of the Complex Crystallization Processes Occurring Laser Heated Amorphous Germanium Films

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The crystallization of amorphous Ge films induced by laser [1,2] heating is quite complex, creating an exotic microstructure that forms over time scales ranging from a few nanoseconds to several microseconds after pulsed laser excitation (see Figure 1). High quality information on the final post-mortem structure can be obtained with a variety of techniques. However, structural characterization of short-lived nonequilibrium states has shown to be very challenging. Revealing the dynamics of the structural transformations in materials requires direct observations with at most nanosecond temporal resolution with a spatial resolution of few nanometers [3]. The recently developed dynamic transmission electron microscope (DTEM) at LLNL permits *in situ* observation of laser induced crystallization behavior with ~10 nm spatial resolution and 15 ns temporal resolution [3].

We used the single-shot nanosecond imaging capabilities of the DTEM to capture the complex crystallization behavior of a-Ge films. During crystallization, three distinct microstructural regions form, (1) nanocrystalline region, (2) radial dendritic-like grains, and (3) azimuthally orientated grains (see Figure 1). Crystallization initiates at times less than 20ns and zone (1) fully crystallizes within 55 ns (see reference [2] for details). The large dendritic grains emerge from the nanocrystalline zone that act as nuclei and grow at rates exceeding 10 m/s to lengths of several microns (see the time resolved images in Figure 2). After delays longer than 1 μ s from the laser heating pulse, the growth mode at the periphery of the laser irradiated zone changes from radial along the direction heat diffusion to tangential, perpendicular to the thermal gradient, forming zone (3) that consist of “azimuthally orientated” layers comprised of large faceted crystals (white contrast) and nanocrystals (dark grey). The time-resolved images in Figure 3 clearly show this tangential growth mode that develops over a period of ~10 μ s. The complexity of the growth dynamics reveal the underlying importance of interface energy on the crystallization kinetics. To discern the influence of this anisotropic growth behavior on the crystallization process, we have studied the local crystallography, post mortem in the TEM, using the novel NanoMEGAS ASTAR automated orientation imaging tool. In general, the dendritic grains exhibit a weak <100> fiber texture common for dendritic growth in cubic systems and are highly faceted containing many twin defects (see Figure 4). This presentation will discuss the crystallography of laser devitrified Ge films in terms of the crystallization mechanisms elucidated by the time-resolved studies using the DTEM.

References:

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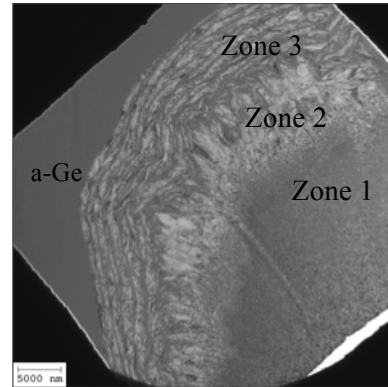


FIG 1. Low magnification micrograph of the laser-crystallized region (90 μm in diameter).

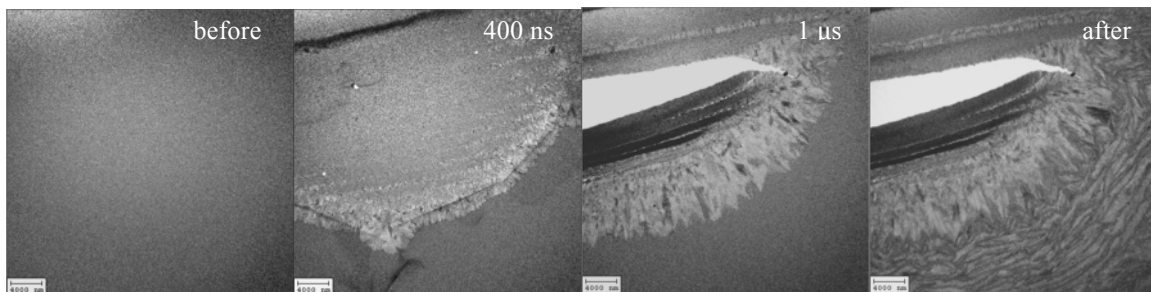


FIG 2. Time-resolved images with a 15ns exposure time showing the growth dynamics of the radial, dendritic grains in Zone (2) at two different time delays, 400 ns and 1 μs .

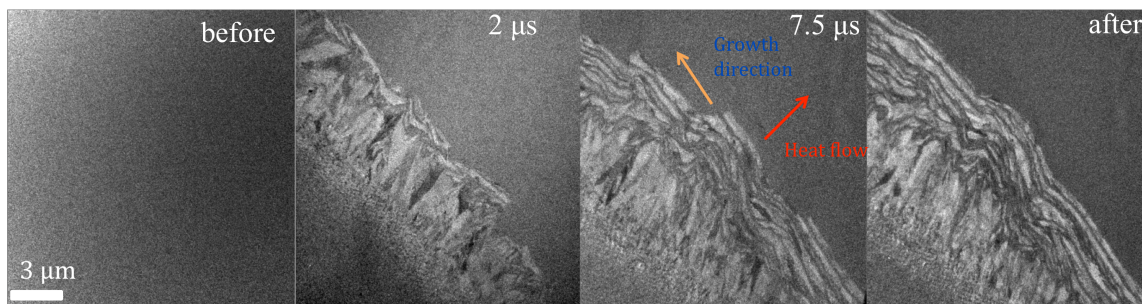


FIG 3. Time-resolved images with a 15ns exposure time showing the growth dynamics of the “azimuthally” orientated layers in Zone (3) that grow perpendicular to the heat flow.

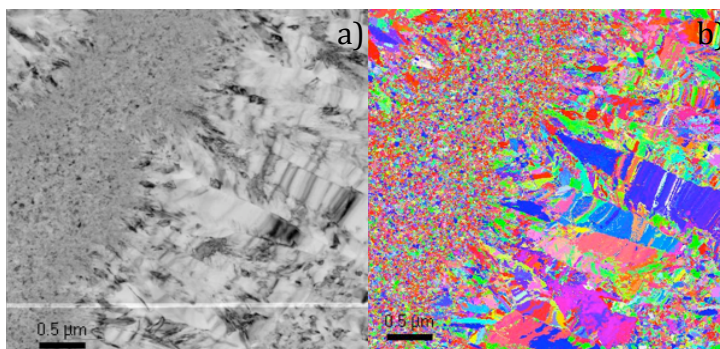


FIG 4. Post mortem images of the laser crystallized microstructure at interface between Zone (1) and (2) taken using the NanoMEGAS, inc. ASTAR® automated orientation mapping system, a) virtual brightfield, b) crystallographic orientation map of the same region.