

Capturing Irreversible Reactions with Nanosecond-Scale Dynamic TEM Movies: Measuring Crystal Growth Rates During Laser Annealing of Phase Change Materials

M. K. Santala¹, B. W. Reed¹, S. Raoux², T. Topuria³, T. LaGrange¹, G. H. Campbell¹

¹. Condensed Matter & Materials Division, Lawrence Livermore National Laboratory, Livermore, USA

². IBM T. J. Watson Research Center, Yorktown Heights, USA

³. IBM Research Division, Almaden Research Center, San Jose, USA

Multi-frame dynamic transmission electron microscopy (DTEM) has been applied to the study of crystal growth in phase change materials (PCMs). PCMs are used for optical memory (CDs, DVDs) and non-volatile random access memory, which exploit the distinct optical and electrical properties of the amorphous and crystalline phases. For memory applications, it must be possible to switch between the amorphous and crystalline phases in nanoseconds by rapid heating. Thus crystallization kinetics of PCMs are of vital interest as they directly impact device switching speed, but even basic quantities, such as crystal growth rates, are difficult to measure experimentally during highly-driven laser- or current-induced crystallization (and are totally inaccessible with conventional microscopy techniques).

“Single-shot” DTEM has been used to study laser-induced crystallization in PCMs, such as $\text{Ge}_2\text{Sb}_2\text{Te}_5$ [3] and GeTe [4]. In the earlier work on GeTe , only a single 15-ns image was formed during each laser-induced crystallization event. This left open questions about the crystal growth rates since the incubation time before the nucleation of the imaged crystalline grains was unknown. Here we capture nine-frame movies of the laser-crystallization of amorphous GeTe (Figure 1), in which each frame consists of an electron image with 17.5-ns time resolution. Individual growing grains are tracked during different points in their growth and unambiguous measurements of growth rate may be made. Crystal growth rates exceeding 3 m/s are observed and changes in growth rate are observed during a single crystallization event.

Finite element analysis simulations are used to model the rapidly changing spatial and temporal temperature profiles during laser heating. The calculated temperature profiles allow us to fit our measured growth rates to kinetic models of crystal growth. The ability to create multi-frame movies of unique irreversible phase transformation on the nanosecond and microsecond scale, paves the way for understanding crystallization kinetics of PCMs over the whole range of technologically-relevant time scales.

References:

[1] S. Raoux *et al.*, *Applied Physics Letters* **95** (2009), p. 143118.

[2] G. Bruns *et al.*, *Applied Physics Letters* **95** (2009), p. 043108.

[3] M. K. Santala *et al.*, *Journal of Applied Physics* **111** (2012), p. 024309.

[4] M. K. Santala *et al.*, *Physica Status Solidi B* **249** (2012), p. 1907-1913.

[5] This work performed under the auspices of the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

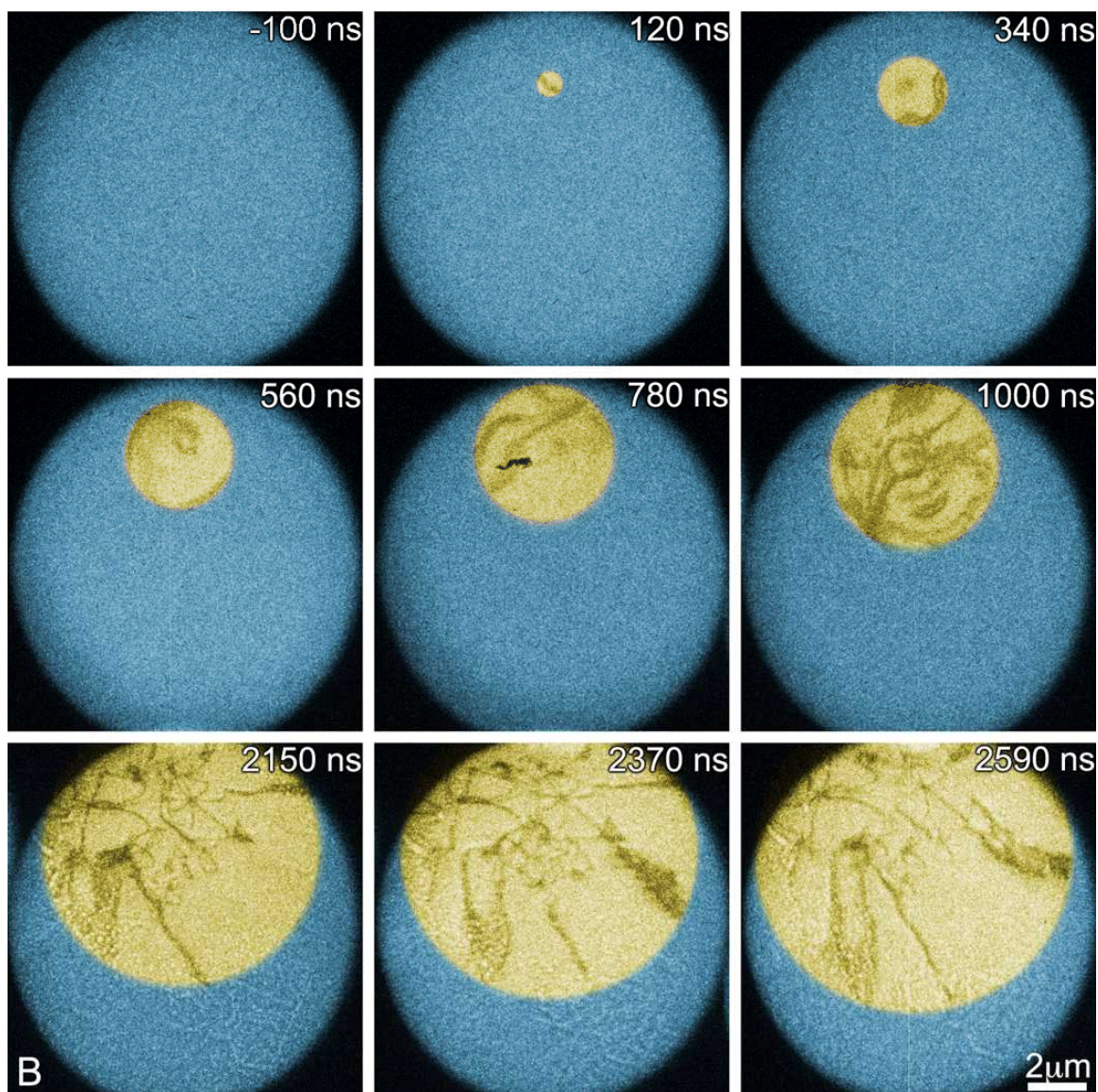


Figure 1. False color images showing the growth of a crystalline grain (yellow) in amorphous GeTe (blue) in a nine-frame movie formed by nine 17.5-ns electron pulses. The time signature in each frame is relative to the time of the peak specimen laser intensity with an uncertainty of ± 3 ns.