

## In-situ Observations of Amorphous Germanium Nucleation and Nanocrystallization by Dynamic Transmission Electron Microscopy

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Crystallisation behaviour upon laser annealing of group IV semiconductors holds great interest from a technological point of view since the final structure determines the properties of the material and functionality of devices created with it. High quality information on the final structure after annealing can be obtained with a large variety of techniques. However, structural characterization of short-lived nonequilibrium states has been very challenging. Revealing the dynamics of the structural transformation of semiconductor materials requires direct observations with nanosecond-scale temporal resolution and spatial resolution of a few nanometers [1].

The transmission electron microscope (TEM) offers high spatial resolution (as low as 0.5Å [2]) and is an indispensable tool for characterisation in materials science. However, due to the poor temporal resolution of conventional TEMs it is rarely used for in situ direct imaging of structural transitions. In this work we present recent results obtained using the enhanced temporal resolution of the Dynamic Transmission Electron Microscope (DTEM) at Lawrence Livermore National Lab (Figure 1) [3], [4]. This instrument enables direct real-space observations of irreversible structural transitions on the nanosecond and nanometer scales.

We have used this new capability to study the crystallization dynamics of amorphous germanium (a-Ge) thin films. Crystallization has been induced by 532nm nanosecond laser pulses of variable fluence. Nucleation and crystal growth have been observed with a temporal resolution of 20 ns. We also observed evolution of the mean crystalline grain size over time at fixed fluence of 128mJ/cm<sup>2</sup>. Time-resolved TEM images have shown that the crystallisation process begins at about 20 ns and the process finishes in about 300ns (Figure 2). The results suggest that the nucleation and the crystal growth processes overlap in the time range of 20-30 ns after the laser pulse arrives. The nucleation finishes at around 30 ns. The grain growth is fast from 50 ns to 100ns but slows thereafter.

Dynamic TEM observations have permitted real-space counting of the nucleation sites and experimental estimation of the density of nucleates. Calculation of the nucleation rate in the interval 20 ns to 30 ns gives a value of  $N \approx 3.6 \times 10^{23} \text{ cm}^{-3} \cdot \text{s}^{-1}$  with mean size of 96 nm. The results are in good agreement with the theoretically predicted values [5].

The process of laser induced nucleation and crystallization of a-Ge has been observed for first time *in-situ* with nanometric spatial and nanosecond temporal resolution.

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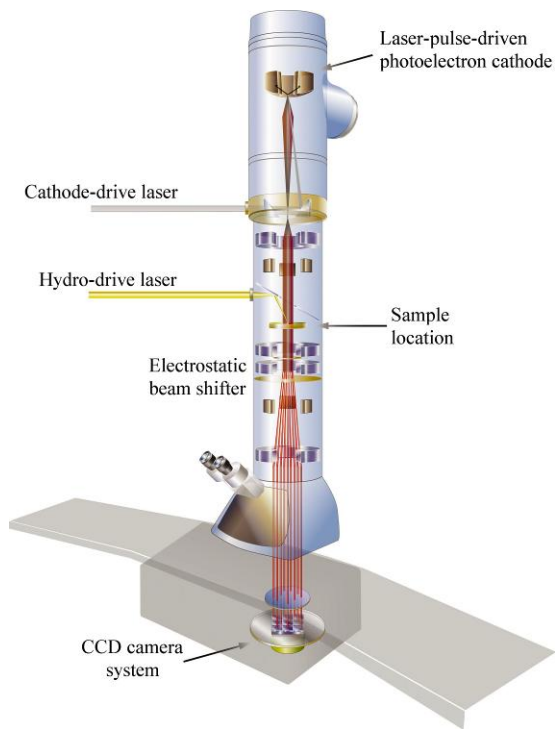


Figure 1. Schematic representation of the DTEM

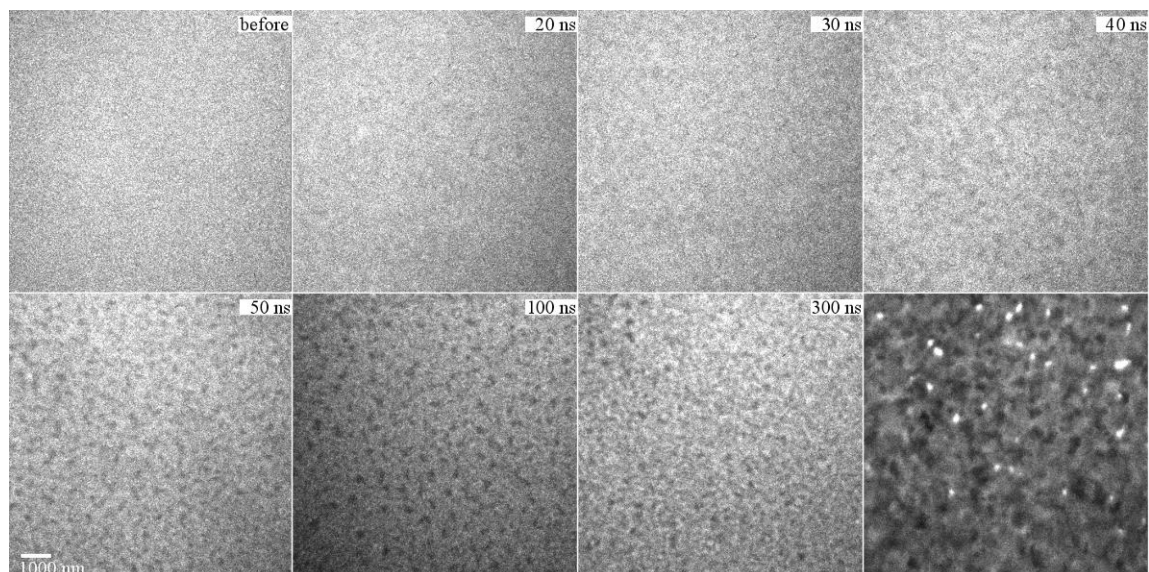


Figure 2. Evolution of the microstructure of the a-Ge film as a composite of independent measurements. The before image presents the sample structure before initiation of the transition followed by different time delays. The final image in the right low corner of the figure presents the post-mortem microstructure i.e. in the end of the transition. The scale is the same for all the images.