Explosive crystallization in phase change thin films

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Latent heat released upon local crystallization of phase change materials can be used to activate the crystallization of neighboring amorphous material resulting in a self-sustained propagation of the crystallization front, known as explosive crystallization (EC). In rewriteable optical disks EC is generally prevented by a heat conducting metal layer. However, in PRAM applications the PC material is mostly surrounded by poorly heat-conducting SiO_X and therefore EC can occur.

In case of EC, the kinetics of the combined melting and crystallization fronts can be described using the Frenkel-Wilson law [1, 2]. Using a 1D thermal balance equation [1] a steady state solution can be obtained that describes the speed of the explosive crystallization front as a function of temperature. By finding the intersection of both descriptions a physically allowed solution is found of the EC front propagation speed. Physically allowed solutions are only found above a critical substrate temperature (Figure 1 left) and a critical film thickness (Figure 1 right). For certain substrate temperatures and thicknesses two intersections occur, leading to two steady state solutions for the same conditions. This might lead to interesting oscillating behavior of the crystallization front speed [2]. During EC the crystallization front will propagate with speeds in the order of 10 m/s. Due to the fast front propagation a high speed camera is used to observe EC.
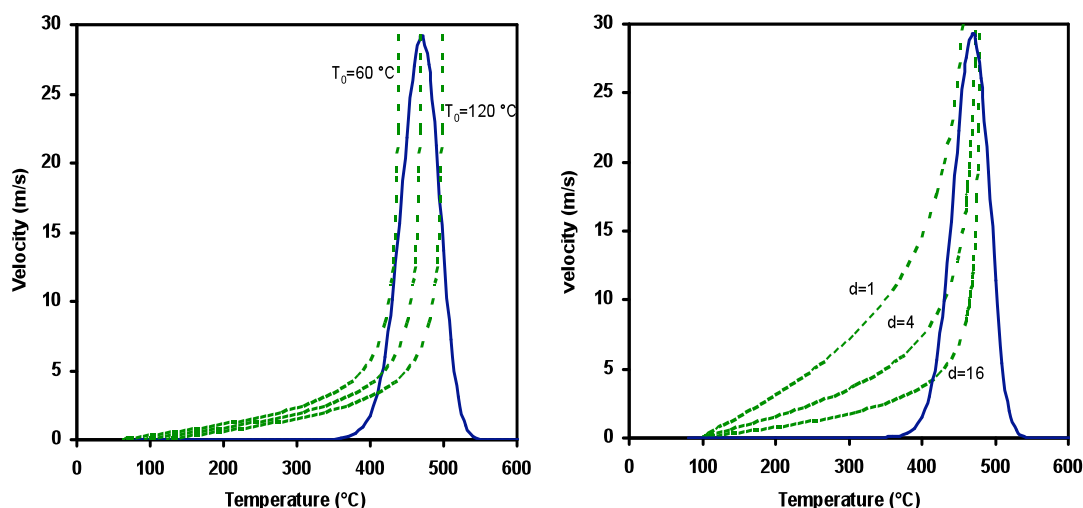

Figure 1. *Crystallization velocity as function of temperature is represented by the blue line for a typical phase change material. Steady state solutions for EC are represented by the green lines. Physically allowed speeds for EC are found above a critical temperature (left) and critical thickness (right).*

Explosive crystallization can be induced in several ways. It has been shown that explosive crystallization can be induced by conductive AFM [2], see Figure 2. By applying an electrical pulse via a stationary AFM tip to a $Ge_2Sb_2Te_5$ film with bottom electrode the film is locally crystallized resulting in further crystal growth that can only be explained by (unstable) EC, because the radius of the crystallized area has become about 100 times larger than the AFM tip radius.

Figure 2. *TEM image of unstable explosive crystallization that occurred after a voltage pulse was applied to a phase-change film using a conductive AFM tip with a radius of ~40 nm [3].*
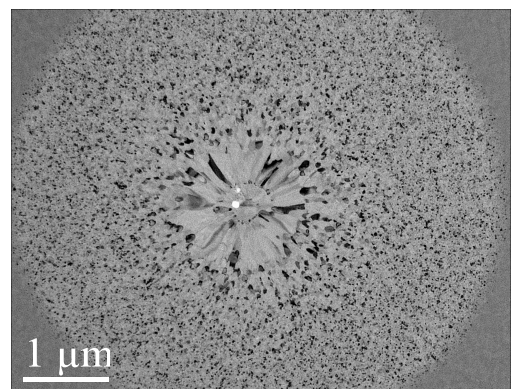
Most experiments on EC are initiated by using a (pulsed) laser [1, 2]. In our experiments we use a 100 mW diode laser wavelength of 655 nm to induce EC. Spot sizes of 5 μm in diameter can be obtained, however this will be improved using better focusing. Also initiating EC using a tapping indenter will be explored.

With our current set-up we are able to write marks in amorphous thin films. A 1 mm^2 crystalline mark was created using 10,000 laser pulses with a pulse duration of 200 μs. At both 10,000 frames per second (fps) and 45,000 fps the crystalline mark was still clearly visible, see Figure 3. With the current lens the minimum feature visible with the high speed camera is ~20 μm. With the crystalline mark still clearly visible at high recording speeds we have shown that we are able to distinguish the two phases and thus will be able to record the front propagation during explosive crystallization.
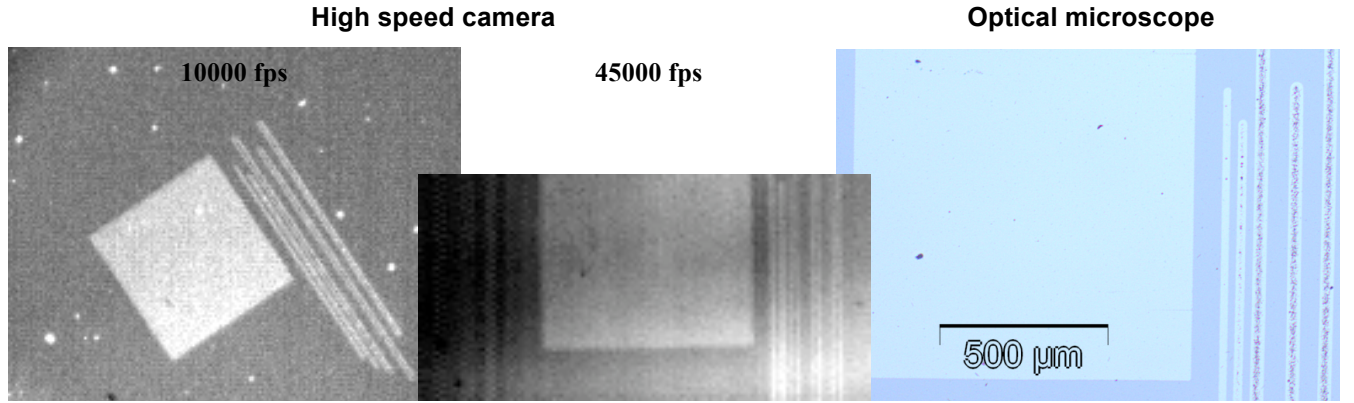
High speed camera   **Optical microscope**

Figure 3. Left and middle: *Images obtained from high speed camera recordings of an amorphous mark written using 100 μs laser pulses.* **Right:** *The same mark as observed using an optical microscope.*

The first studied composition is $In_3Ge_5Sb_{21.2}Te_{70.8}$, a material suitable for PRAM cells. Samples consisted of a glass substrate with a 100 nm amorphous phase change film protected by a 10 nm ZnS-SiO_2 capping layer. The samples did not show stable EC up to a substrate temperate of 180 degrees Celsius. A growth in spot size was seen with increasing temperature, which is attributed to decreased heat loss to the substrate (see Figure 4). Currently, we are studying film thicknesses of 200 nm and 400 nm. Different phase-change film compositions will be also investigated in the near future.
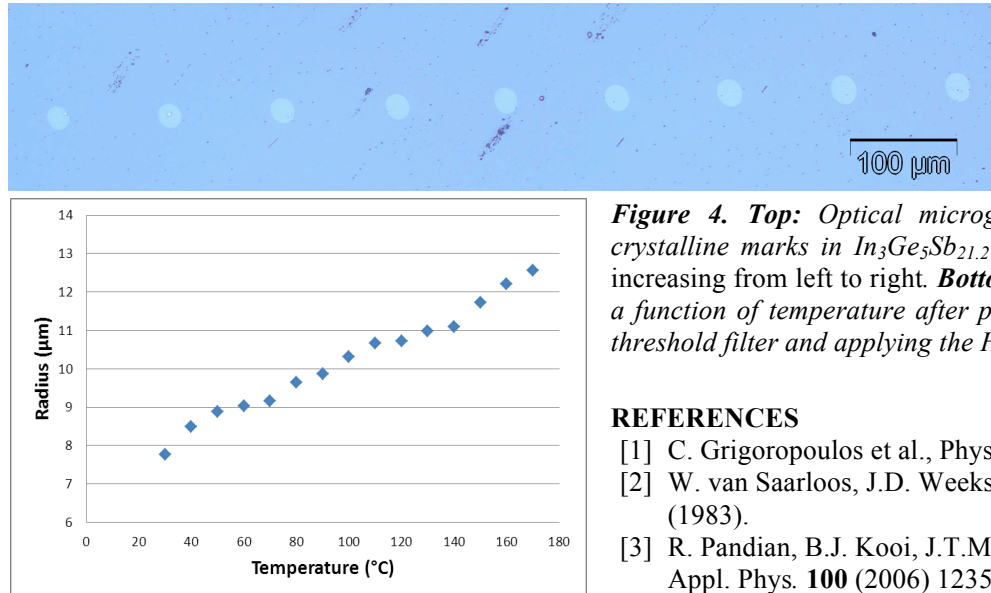

Figure 4. Top: *Optical micrograph showing laser induced crystalline marks in $In_3Ge_5Sb_{21.2}Te_{70.8}$ at various temperatures, increasing from left to right.* **Bottom:** *Crystalline mark radius as a function of temperature after processing using a median and threshold filter and applying the Hoshen-Koppelman algorithm.*

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