## **Supporting Information**

## Understanding the Crystallization Behavior of As-Deposited Ti-Sb-

## Te Alloy through Real-Time Radial Distribution Function

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**Figure S1** Sheet resistance as a function of temperature for Sb<sub>2</sub>Te<sub>3</sub> and Ti doped Sb<sub>2</sub>Te<sub>3</sub> films. The experiments were carried out by Linkan LMP 95 hot stage. As shown in this figure, with the increase of annealing temperature, a slow and gradual drop of sheet resistance ( $R_s$ ) is observed for as-deposited ST film. This is because that the ST film has partly crystallized during the sputtering process due to its low  $T_c$  (< 100 °C). Unlike the *RT* curve for ST, after initially monotonic decrease, sudden drops of  $R_s$ , corresponding to  $T_c$ , happens at 175.6 °C, 185.5 °C and 195.9 °C for T<sub>0.32</sub>ST, T<sub>0.43</sub>ST and T<sub>0.56</sub>ST, respectively. These values are much higher than that of pure ST and conventional GST (~150 °C). The high  $T_c$  can inhibit the spontaneous crystallization and make the amorphous state more stable.



**Figure S2** *In-situ* crystallization of  $Ge_2Sb_2Te_5$  film by electron-beam irradiation, characterized by FEI Tecnai F20 TEM at 200 KeV in (a) TEM and (b) HRTEM modes. The diameter of beam spot is about 180 nm. To insure that the same area  $(a_{ei})$  is irradiated by the high-energy electrons, marked by black dashed circle in the figure,

a margin location of the film is used (indentified by dark region in the top left corner). From the figure, we can see that, before electron irradiation, the as-deposited film is amorphous without any lattice, and the surface is very smooth and uniform. After irradiating for 7 minutes, no difference is observed in the morphology. First visible change is found under irradiation for 14 minutes, when some domains of the aei becomes dark, especially in the center. As the irradiation time goes on, the contrast between the a<sub>ei</sub> and surrounding becomes more and more obvious, and the central dark area expands outwards the non-irradiation region. From the HRTEM images of the a<sub>ei</sub>, we can find the reasons for the morphology evolution. No lattice fringe is observed before 14-minutes of electron irradiation. However, after then, ellipticshaped nanoparticles of various sizes appear. Under longer electron irradiation, the grains get larger and more grains come into sight. Thus, the GST film is crystallized under the long-time electron irradiation, and the appearance of grains is responsible for the morphology evolution. It is necessary to state that the area in HRTEM images is not the same because that it's too small to be located in a limit time, while long time exposure to electrons would cause the crystallization of the film.