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Direct Observation of Structural Transitions in the Phase Change Material $Ge_2Sb_2Te_5$

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Schematic diagram of the FIB lift-off process:

We also prepared TEM specimens using a focused Ga+ ion beam (FIB, Helios NanoLab 600i, FEI) coupled with a lift-off technique. The schematic diagram of the FIB lift-off process and TEM images are shown in Figure S1

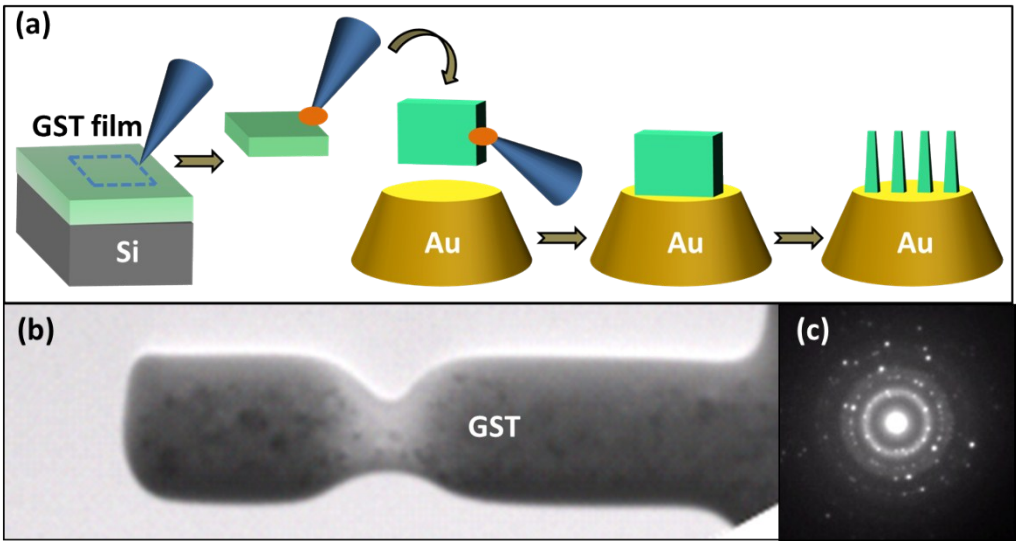

Fig. S1. (a) Schematic diagram of the FIB lift-off process. (b) TEM image of a polycrystalline status GST film. (c) The corresponding SAED patterns.

Crystal structure:

We investigated the crystal structure of the single-crystalline status GST nanofilm after the application of the DC voltage, and the results are shown in Figure 2. The SAED patterns correspond to face-centred cubic [001], [012] and [110] orientations, respectively. It is interesting to note from the corresponding SAED pattern that the polycrystalline status GST transformed to a single-crystalline status after applying an external voltage, however, the PCM still remained FCC structure. This transition is quite different from pervious observation of the structural change from FCC to HCP caused by heating.

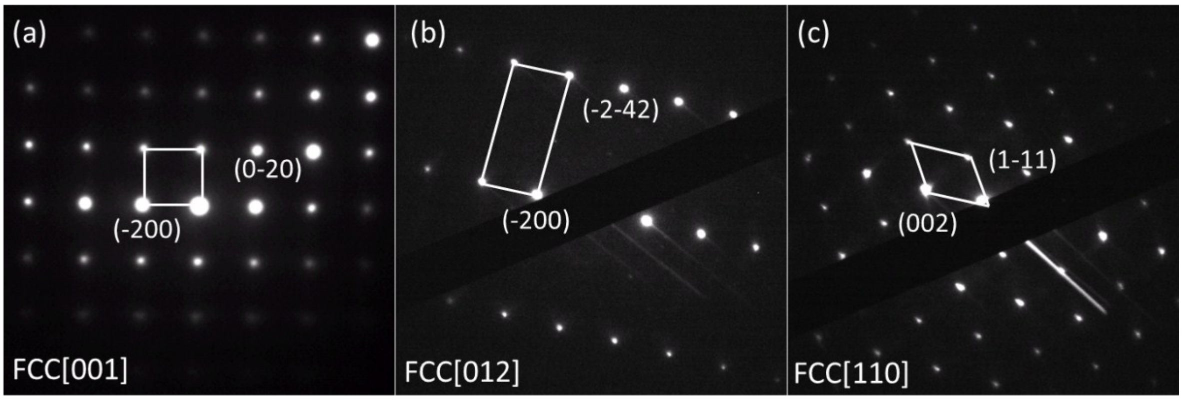

Fig. S2. (a-c) The corresponding SAED patterns along [001], [012] and [110] orientations respectively.

Large current induced breakdown of the PC material:

Fig. S3a shows a TEM image of a polycrystalline status GST film. No upper limit of the current was set. Then we increased the voltage gradually. When high amplitude voltage was applied, the GST specimen suffered from breakdown due to the joule heating effect induced by the large current.

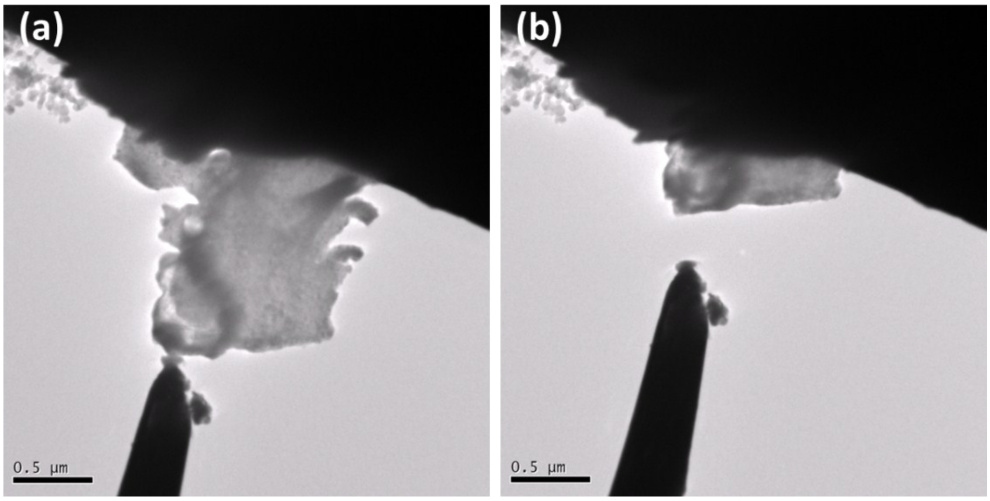

Fig. S3 Breakdown of the GST film induced by large current.(a) TEM image of a polycrystalline status GST film. (b) TEM image of a polycrystalline status GST film after applied a high voltage.

Electric pulse induced transition:

The polycrystalline status to single-crystalline status transition can also be realized by an electric pulse. The duration of the electric pulse used in Figure S4 is 30 ns and the amplitude is 3 V, indicating that this polycrystalline status to single-crystalline status transition is suitable for a multi-level operation.

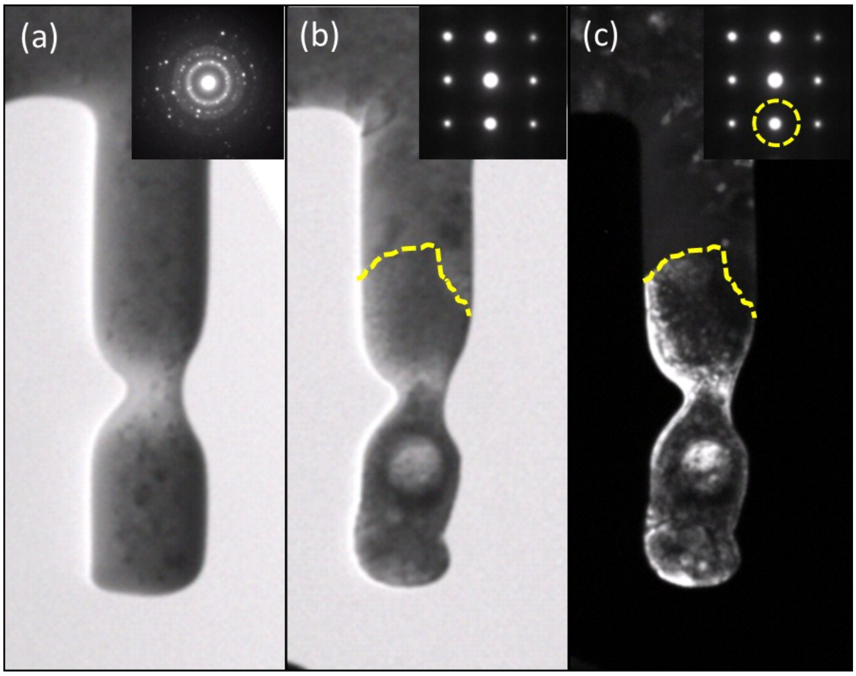

Fig. S4 (a) TEM image of a polycrystalline status GST film before applied an electric pulse. (b) TEM image of a single-crystalline status GST film after applied a electric pulse (30 ns duration of, 3 V amplitude). The insets are the corresponding SAED patterns. (c) Corresponding dark field images of the single-crystalline status GST film. The insets are the corresponding SAED patterns.

Direct transition from single-crystalline to poly-crystalline state

The transition from single-crystalline to poly-crystalline state can be achieved through special electric pulses. Figure S6 shows this process. Fig. S6a is a TEM image of a single-crystalline state GST film. The inset is the selected electron diffraction pattern corresponding to dashed circle region showing the single-crystalline character (none zone-axis due to single-tilted holder). Fig. S65b is a voltage scan from 0 V to 4 V with a trailing edge of 64 ms. 64 ms is a fixed ramp time of the Nanofactory STM-TEM holder[1], to get a smooth transfer from the bias voltage go back to the 0 V after the last measurement. The front end of the GST in the yellow dotted circle of Fig. S6c transformed to poly-crystalline state after the voltage scan, which can be confirmed by the corresponding dark field DFTEM image (Fig. S6d).

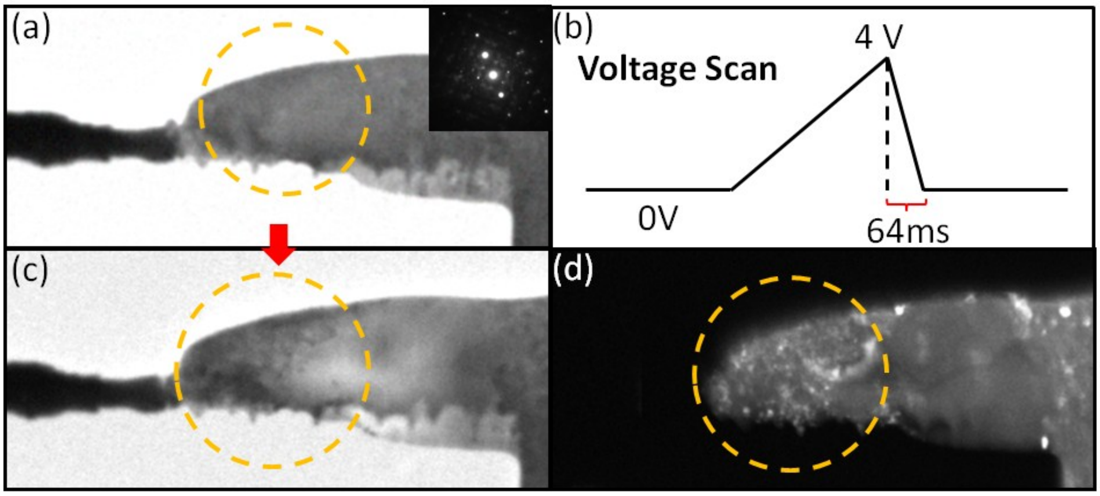

Fig. S5 Direct transition from single-crystalline to poly-crystalline state. (a) TEM image of a single-crystalline state GST film. (b). A voltage scan from 0 V to 4 V with a trailing edge of 64 ms. (c). Poly-crystalline state GST after the voltage scan. (d) The DFTEM image corresponding to (c).

The influence of the geometry of GST films:

We repeated the experiment using TEM specimens with different sizes. From which we found that when the TEM specimen is small, it is much easy to obtain single crystal across the entire GST film, while if the TEM specimen is large, the entire GST film tends to form several large elongated crystals. Fig. S5 is a TEM image of a large GST film after the poly-to-single crystal transition. It can be seen that the active region of the GST film is composed of 5 large elongated crystals.

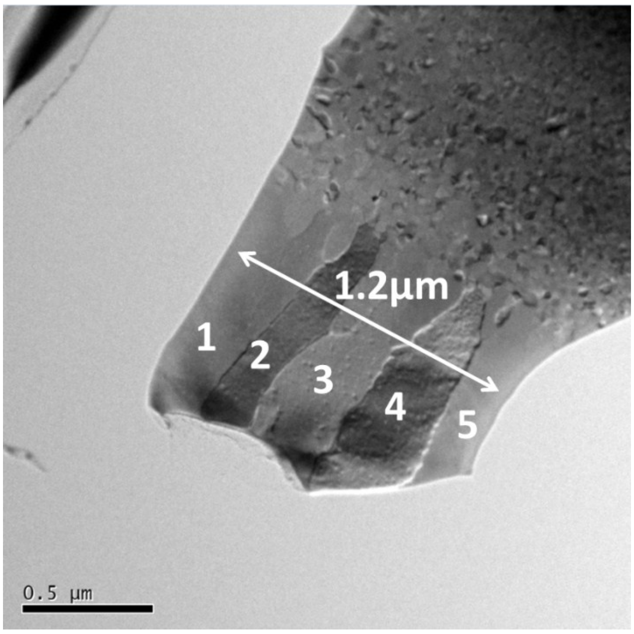

Fig. S6 TEM image of a large GST film after the poly-to-single crystal transition. It can be seen that the active region of the GST film is composed of 5 large elongated crystals.

Reference

1. K. Svensson, Y. Jompol, H. Olin and E. Olsson, *Rev. Sci. Inst.*, 2003,**74**, 4945.