## Supporting Information

## Enhanced performance of phase change memory by grain size reduction

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**Figure S1.** BF TEM images and the raw integrated diffraction curves of electronic diffraction intensity extracted from the respective SAED patterns for (a)-(b) GST and (c)-(d) TaGST films.



Figure S2. In-situ XRD results for GST and TaGST films.



Figure S3. Raman spectra for amorphous and crystalline GST and TaGST films.



**Figure S4.** Evolution of selected area electron diffraction patterns and the corresponding real-time radial distribution function G(r) curves of (a)-(b) GST and (c)-(d) TaGST at *r* ranging from 2 to 100 Å.



**Figure S5.** Evolution of real-time radial distribution function G(r) curves at *r* ranging from 2 to 10 Å for as-deposited films.



**Figure S6.** (a)-(b) Coordination number distribution and (c)-(d) the bond angle distribution of Ge, Sb, Te and Ta for amorphous GST and TaGST.



**Figure S7.** (a) HRTEM image of TaGST film after 400 °C for 20 min. (b) Ta element is distributed in a complete grain of TaGST film after annealed at 400 °C for 20 min.



Figure S8. The R-V of the device based on TaGST.



**Figure S9.** XRR patterns for a) GST, b) TaGST films. The insets are the plot of  $\sin^2\theta$  versus  $(m + 0.5)^2$  films.



Figure S10. (a) The R-V of the sample 1. (b) Endurance of the sample 2.



**Figure S11.** The variation tendency of basic element contents for regions 1-4 of the sample 2.