Supporting Information

Revealing self-aligned γ -SnTe ultrathin nanosheets in

thermoelectric β-SnTe

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Figure S1. The single-phase polycrystalline β-SnTe sample. (a) A typical X-ray diffraction pattern of the Sn_{0.5}Te_{0.5} bulk sample. The scans were recorded in the range of $2\theta = 20-75^{\circ}$ with a step size of 0.02° . The X-ray diffraction pattern can be indexed as β-SnTe (*Fm*3*m*) with a rock-salt structure. The calculated lattice parameter of β-SnTe is a=b=c=0.63621(3) nm. (b) and (c) Back-scattered electron scanning electron microscopy image and the corresponding energy dispersive X-ray spectroscopy (EDS) elemental maps of a polished Sn_{0.5}Te_{0.5} sample. The EDS results are consistent with the X-ray studies, indicating that the single-phase polycrystalline β-SnTe sample was obtained. Additionally, there is no apparent element enrichment at the grain boundaries in the Sn_{0.5}Te_{0.5} sample.



Figure S2. (a) HAADF-STEM image and (b,c) the fast Fourier transform (FFT) patterns of β -SnTe sample containing a nanosheet, viewed along the [110] zone axis of β -SnTe. It is noted that the FFT-1 pattern is taken from the β -SnTe region containing the nanosheet, and the FFT-2 pattern from the perfect β -SnTe region. By comparing FFT-1 with FFT-2, the extra spots in the FFT-1 pattern can be discerned, as demonstrated by a yellow arrow in (b), resulting from the appearance of the γ -SnTe nanosheet in β -SnTe.



Figure S3. A schematic drawing of the β -SnTe region containing γ -SnTe nanosheet overlapped with the normal β -SnTe matrix, viewed along the (a) [100] and (b) [110] zone axis of β -SnTe. From the viewpoint of structural model, the overlapping of β -SnTe region containing γ -SnTe nanoscale precipitate (a₁ and b₁) and the normal β -SnTe matrix (a₂ and b₂) results in the superposed structure models (a₃ and b₃), which matched well with the experimental HAADF-STEM images (a₄ and b₄). In (b₁), two types of nanosheet/ β -SnTe interface can be distinguished. If the β -SnTe part containing γ -SnTe nanosheet (b₁) is in the majority of the HAADF-STEM sample along the viewing direction, these two types of nanosheet/ β -SnTe interface can also be discerned, as shown in the HAADF-STEM images (b₄). The interfaces are indicated by colored arrows in (b₁), (b₃), and (b₄).

Movie S1. The structure evolution of γ -SnTe ultrathin nanosheet to β -SnTe occurs under *in situ* electron-beam irradiation. The *in situ* electron-beam irradiation experiments were performed on a JEOL ARM200F microscope equipped with a probe aberration corrector, operated at 200 keV. Under the HAADF conditions, the video of the phase transition from γ -SnTe ultrathin nanosheet to β -SnTe was captured by recording the preview window during the experiments. In HAADF-STEM mode, the imaging conditions are a beam current of ~6.4 pA, a pixel dwell time of 37.8 μ s/pixel and a pixel size of ~0.037Å², which corresponds to a radiation dose of ~4.1×10⁶ e/nm². It is noted that the phase transition occurs in the course of the acquisition of time series consisting of 20 frames.