**Electronic Supplementary Information**

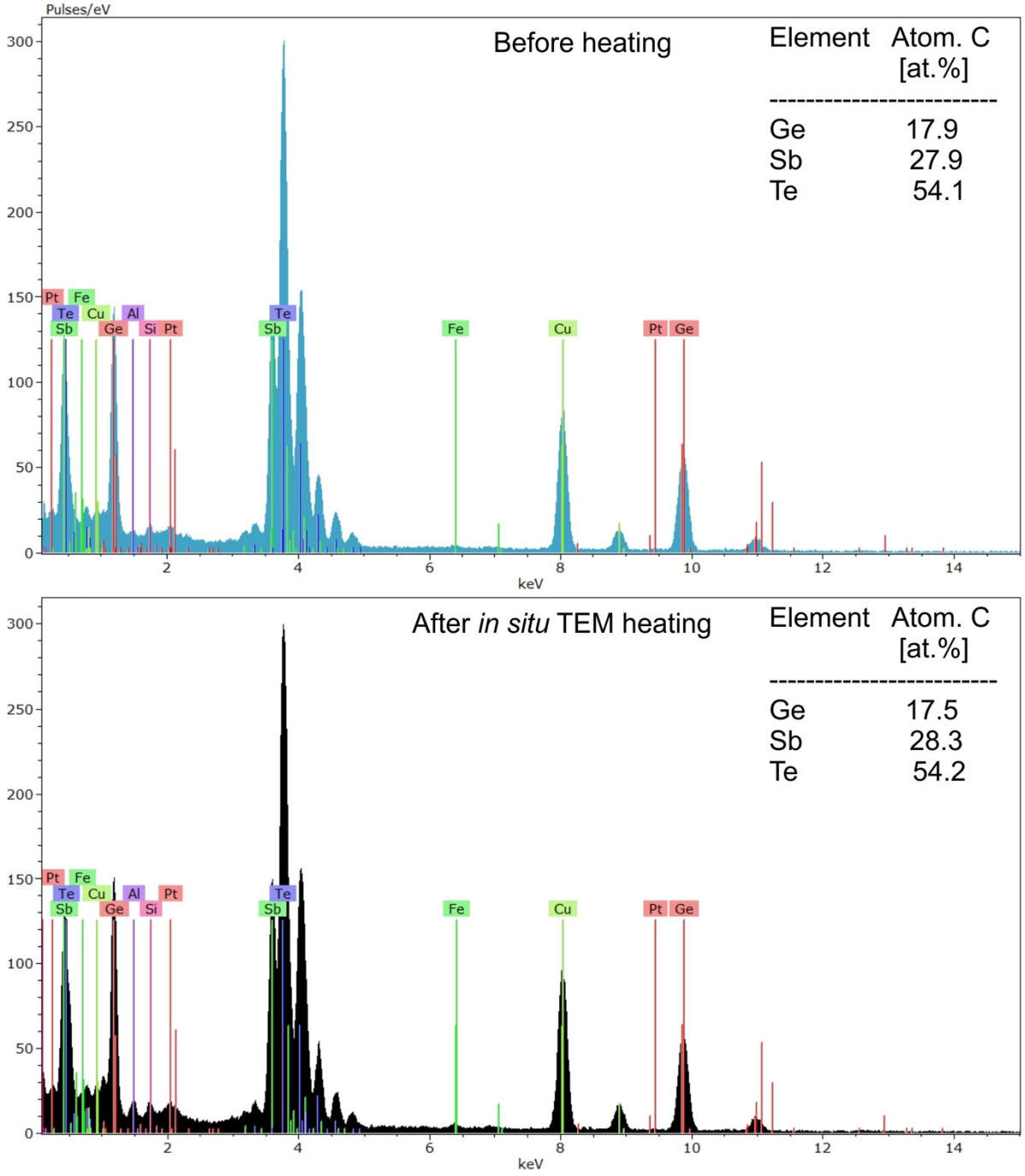


**Fig. S1.** EDX spectra with quantification acquired before and after in situ TEM heating. The Cu signal is due to TEM specimen support whereas Pt signal is caused by Pt protective layer deposited durin FIB lamella preparation. The Si, Al and Fe signals are stray signals coming from the detector design and TEM system.

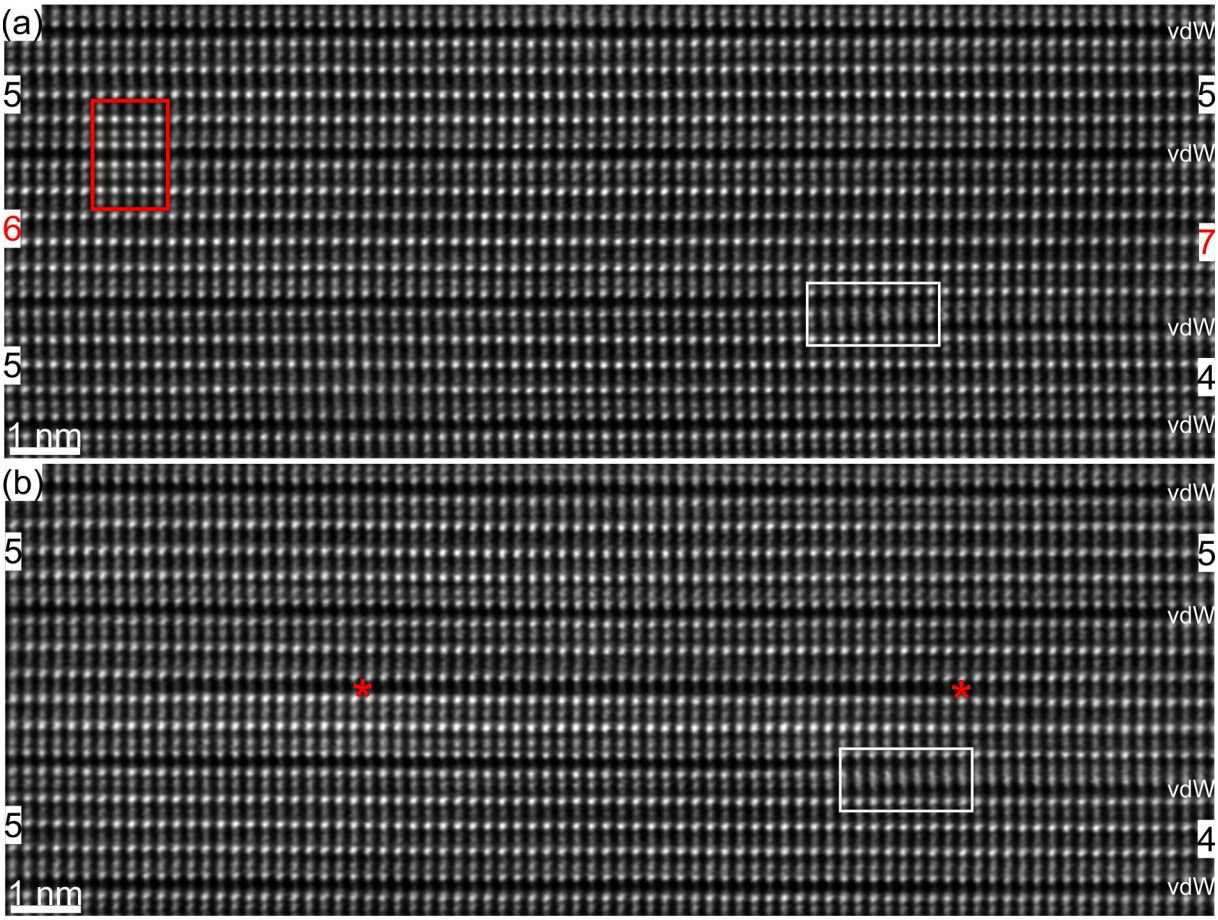


**Fig. S2.** (a) HAADF image shown in Fig. 2(c) of main manuscript. The rectangles mark the positions of pre-existing bilayer defect for visual guidance and area tracking, whereas the number in HAADF micrographs gives the quantity of Te layers per building unit. (d) Artificial writing of VL within of 6/7-fold Te layered unit by targeted electron beam exposure of cation layer inside of the unit as previously seen in Fig. 2a of the main manuscript. The formation of VL results in the relaxation of Te planes into the VL. The arrows in the image show the vdW gaps whereas the marks "*" point out the VLs. The insert in (a) shows simulated HAADF image. Due to vicinity of strong scatters Te to the vdW gaps, the electron probe intensity spreads into the gap. The intensity appears as weak streak lines between opposite Te atomic columns, supporting experimental HAADF images well.

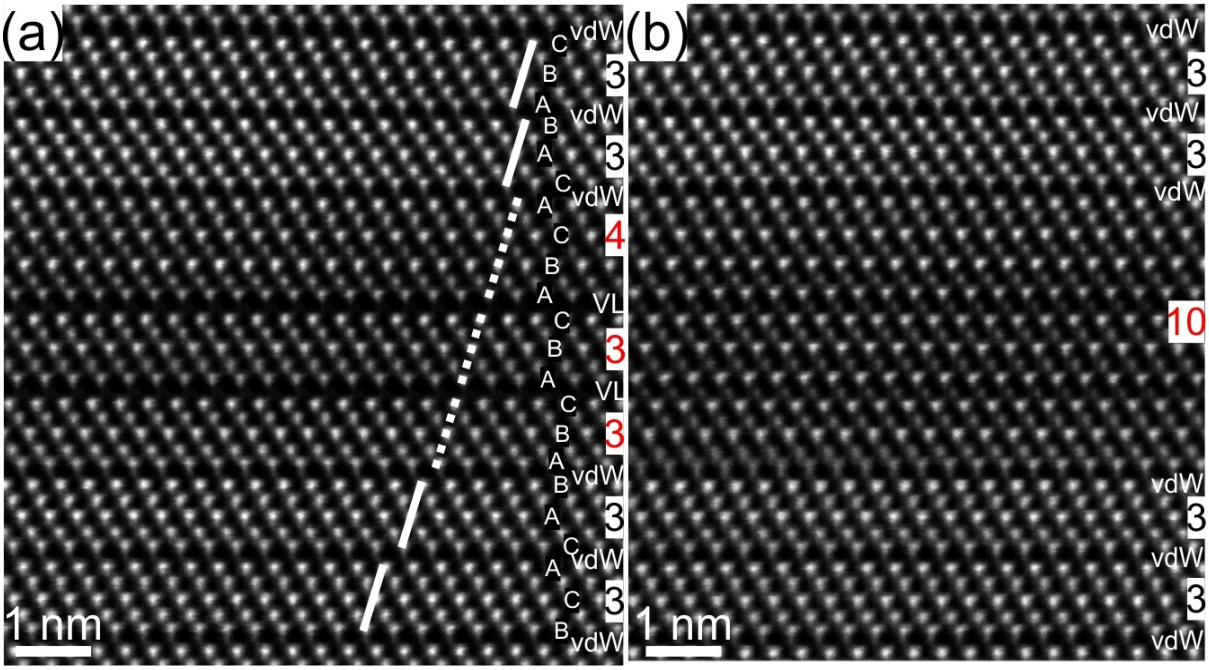


**Fig. S3** (a) and (b) HAADF-STEM images of TEM specimen *in situ* heated to a TEM holder temperature of 320 °C and after electron beam exposure, respectively. The images were acquired at a room temperature. The vdW shows the vdW gaps whereas the VL points out the vacancy layers. The number in HAADF micrographs gives the quantity of Te layers per building block. The reconfiguration of 3-fold and 4-fold Te layered structures separated by VLs into a single 10-fold Te layered unit has occurred after electron beam exposure. The stacking of Te planes is also shown. The stacking sequence of Te layers across the VLs is cubic whereas the stacking of Te layers across the vdW gaps is hexagonal.

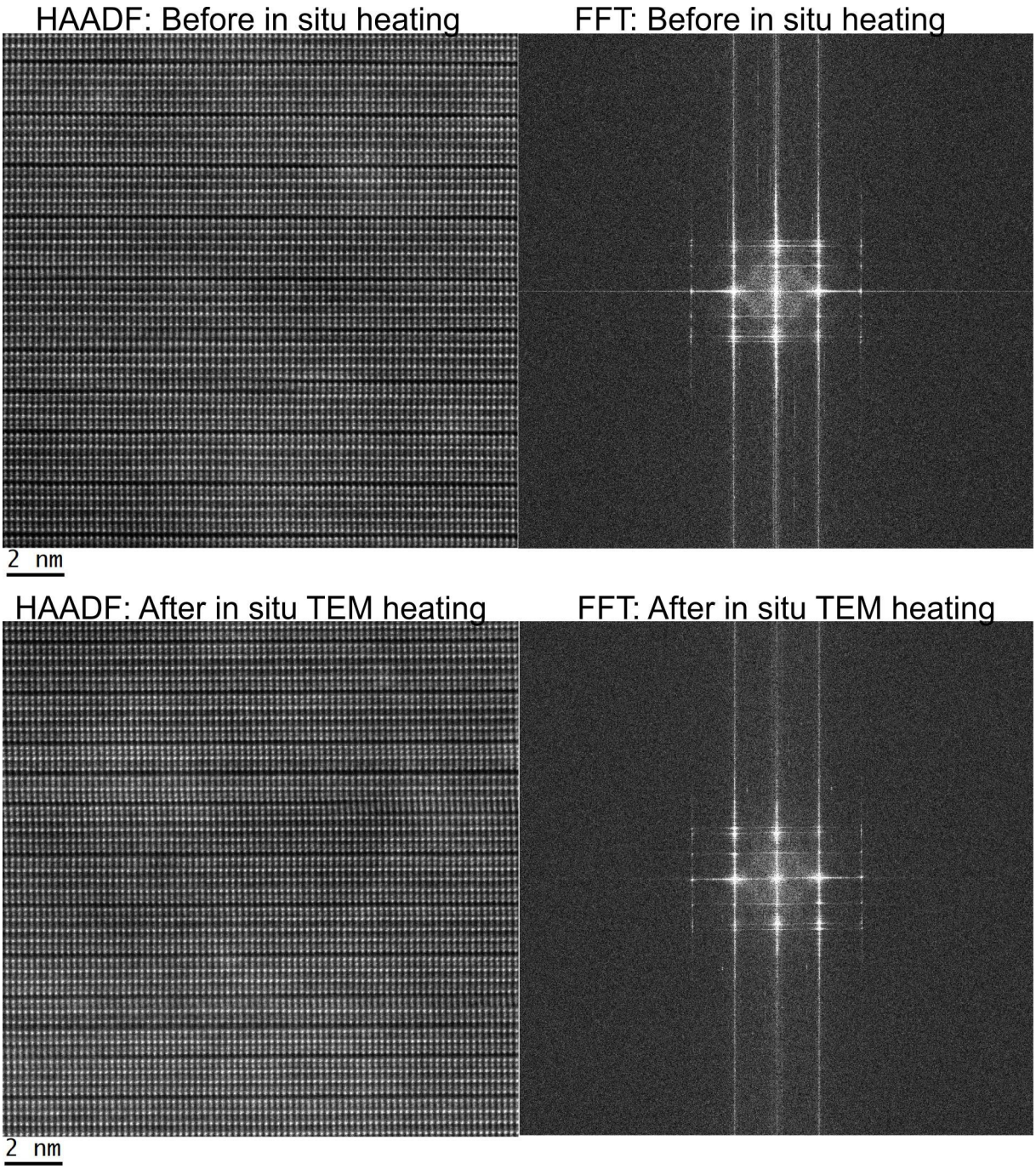


**Fig. S4.** HAADF-HRTSEM and FFT images before and after in situ TEM heating. The FFT images show a diffuse intensity, pointing out on a thin amorphous background layer. Qualitative evaluation suggests same background intensity for the both images. The FFTs are shown with the same image intensity settings.