Electronic Supplementary Information for

In-Situ High Temperature Atomic Level Dynamics of Large Inversion Domain Formations in Monolayer MoS₂

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Figure S1. Boundary structures of the inversion domain in Figure 2(f) in the main article. (a) The ADF-STEM image of the inversion domain, shown in false colour with atomic smoothness. (b, c) Magnified views of the boxed region in (a) with schematic atomic models of Mo (blue), 2S (yellow), and single S (pink) atoms overlaid, which indicate the domain boundaries of this inversion domain comprise of (b) 4|4E (edge sharing) grain boundary, (c) 4|4P (point sharing) grain boundary, respectively.



Figure S2. Some other examples of the inversion domains showing the presence of line defects nearby (marked by arrows), typically leaned on by the side of inversion domain and serve as an extending line of the domain GB.



Figure S3. Schematic illustration by atomic models showing the MoS_2 lattice shrinkage required as a precondition to form an inversion domain. (a) MoS_2 lattice only with 4|4E GB. The right side is pristine lattice, and the left part is the inverted lattice. (b) Adding another segment of pristine MoS_2 lattice as highlighted, to form another GB with the inverted lattice. (c) The formed structure with a 4|4E GB and a 4|4P GB separating inverted domain and the pristine lattice. It is found that in order to create this structure, there should be a lattice shrinkage in the pristine MoS_2 lattice, as marked by the white dashed rectangular in (c). For the typical inversion domain that is captured in our STEM experiment in Figure 2 in the main article, this lattice shrinkage is produced by the 2SVL, which is the extended line of the evolved 4|4E GB.



Figure S4. Parameters for using GPA analysis. (a) Two vectors selected. (b) Detailed parameters. (c,d) Referencing area chosen.



Figure S5. Supporting example showing the compressive stain fields of the line defects. (a) The ADF-STEM image of a MoS₂ monolayer consisting of three S vacancy defect lines. Scale bar: 2nm. (b, c) Stain components \mathcal{E}_{xx} (b) and \mathcal{E}_{yy} (c). The red arrows manifest the compressive strains caused by the line defects.



Figure S6. Stain fields of the lattices before (a-c) and after (d-f) the inversion domain formation.



Figure S7. The schematic crystal atomic models showing the formation of a large inversion domain starting from the MoS₂ lattice with a two-sulphur vacancy line (2SVL), corresponding to the change from Figure 2e to 2f in the main article. (a) Atomic model of monolayer MoS₂ lattice with a long 2SVL and the associated side view. (b) Atomic model of the formed inversion domain along that 2SVL, with two different types of grain boundaries (GB). The 4|4 edge (4|4E) sharing GB (with the model side view beneath) evolves from the 2SVL, and the 4|4 point (4|4P) sharing GB forms in another side of the domain. (c) The ADF-STEM image of the inversion domain corresponding to (b). (d) Image obtained by overlapping (a) and (b), showing how lattice changes when forming the inversion domain. The original lattice is displayed in a half-

transparent effect. For the lattice outside the inversion domain in the model, region (i) and (ii) are kept unchanged while the pristine region (iii) need to shift leftward for nearly half lattice, which actually in the experimental data is not a pristine lattice area and the required shift is accommodated by the compact line defects in the bottom side of the inversion domain in (c). (e) Expanded view of the red dashed box in (d), highlighting the atoms movement at a typical corner region intersected by 4|4E GB, 4|4P GB, and the 2SVL. The 4|4E GB evolves from the 2SVL structure in (a). The red arrows present the migration direction of Mo atoms in one side of the 2SVL, which shift half lattice, creating 4|4E GB and 4|4P GB simultaneously thus forming the inversion domain.



Figure S8. Another example illustrating the Mo atoms movement towards the defects area during the inversion domain formation, while S atoms almost stay at their positions. (a,c) and (b,d) are the ADF-STEM images before and after the inversion domain forms, respectively. (c) and (d) are the same frame as (a) and (b), separately, which are overlaid with schematic atoms marking their original positions, and the arrows in (d) indicate the displacement pathway of Mo atoms inside the inversion domain.



Figure S9. One corner structure of the inversion domain, which is also at the intersection area of two compact multi-S line vacancies. (b) is the large view of the boxed region in (a), overlaid with schematic atomic models consisting of Mo (blue) and S (yellow) atoms. It shows numbers of distorted lattices at this region. Also considering the tense strong field at the tip, it is expected that the lattice in this small area would be unstable, which indeed opens a hole in the following frame (Figure 5b in the main article) when continuing to be irradiated by electron beam.



Figure S10 ADF-STEM images of (a,b) original inversion domain and (c,d) expanded inversion domain, as supplementary to Figure 7 in the main article. (b) and (d) are the same images as (a) and (c), respectively, with overlaid schematic atomic models interpreting the boundary structure of the inversion domain.



Figure S11 Schematic illustration by atomic models showing the change when the MoS_2 lattice at one side of the 2SVL is inverted, forming a 4|4E GB adjoining 2SVL structure, corresponding to the structure change near the 2SVL during inversion domain expansion in Figure 7 (c-e) in the main article.