

Supplementary Materials for

Hidden CDW states and insulator-to-metal transition after a pulsed femtosecond laser excitation in layered chalcogenide 1T-TaS_{2-x}Se_x

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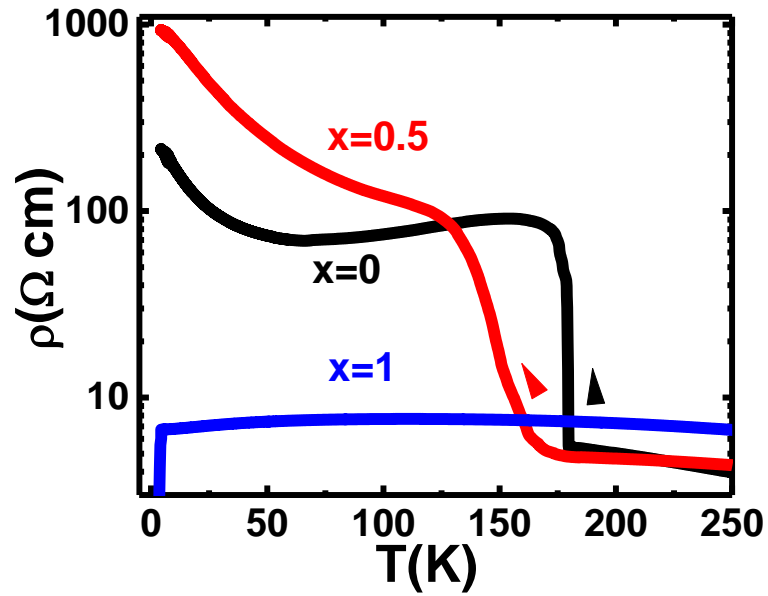


Fig. S1. Resistivity curves for the samples used in this study. Temperature dependence of resistivities for $1\text{T-TaS}_{2-x}\text{Se}_x$ single crystal with $x = 0, 0.5,$ and $1,$ indicating the transitions from NCCDW to CCDW by arrows.

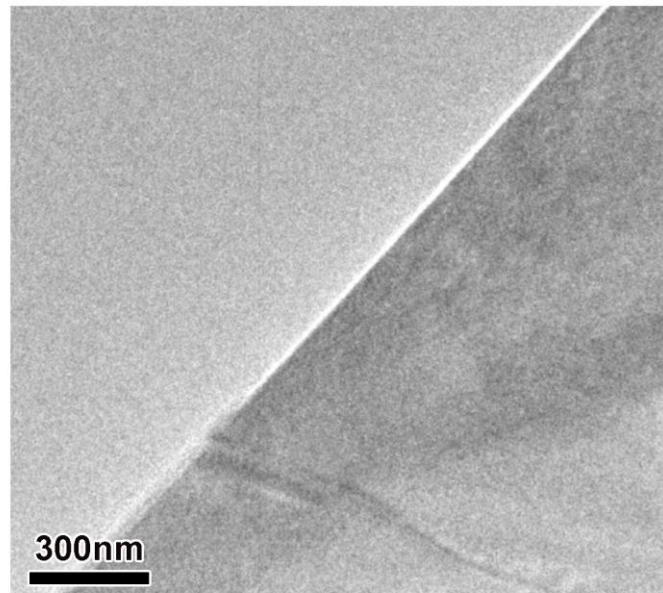


Fig. S2. TEM bright-field image of 1T-TaS_2 . Bright-field images obtained along $[001]$ zone-axis direction at 10 K for $x = 0$ sample. A remarkable structure defect indicates the observed area.

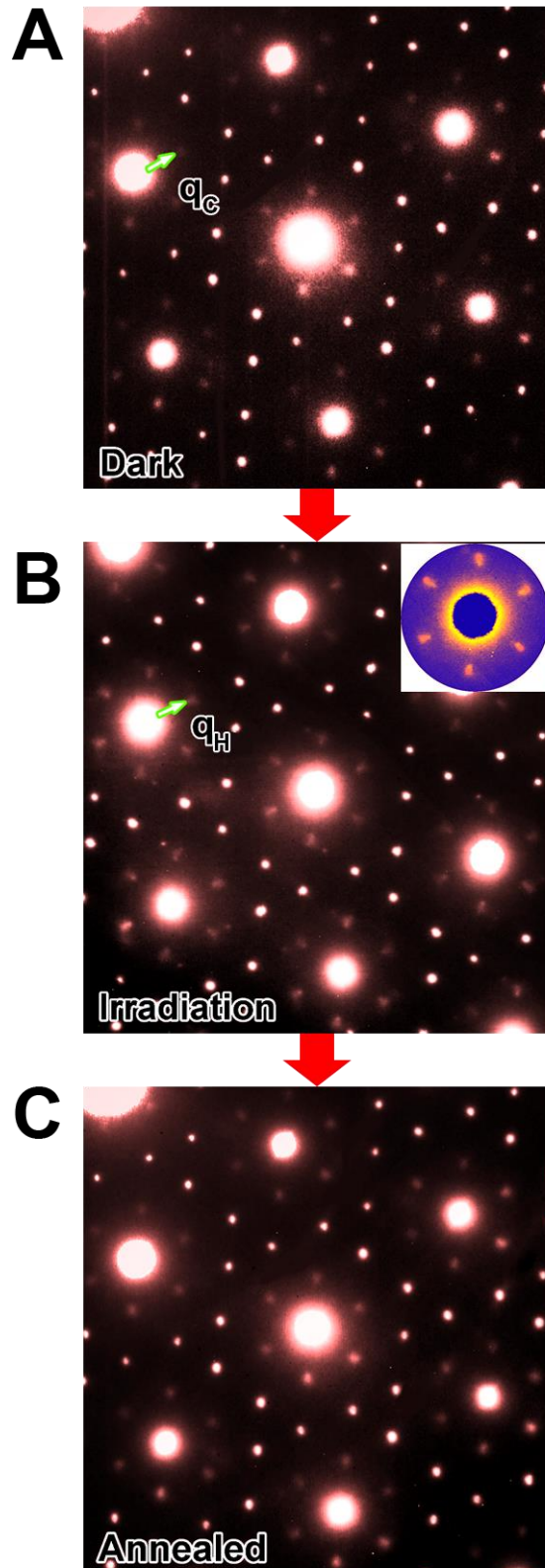


Fig. S3. In situ observation for the electron diffraction patterns of 1T-TaS_{1.5}Se_{0.5}. (Color online) Electron diffraction along the [001] zone-axis direction for $x = 0.5$ sample at 10 K under (A) dark (CCDW), (B) irradiation, and (C) after annealed, showing the space-anomaly H-CDW state microstructure in terms of clear satellite

spots splitting, the inset shows the enlarged diffraction spot. An annealing process is also able to drive the diffraction pattern of space-anomaly H-CDW state back to CCDW (Mott ground state), which follows the thermal erasure behavior of $x=0$ sample. Whereas, the situation remains unchanged when increasing the fluence up to 5 mJ/cm^2 instead of an obvious orientation anomaly in H-CDW state existing in the $x=0$ sample.

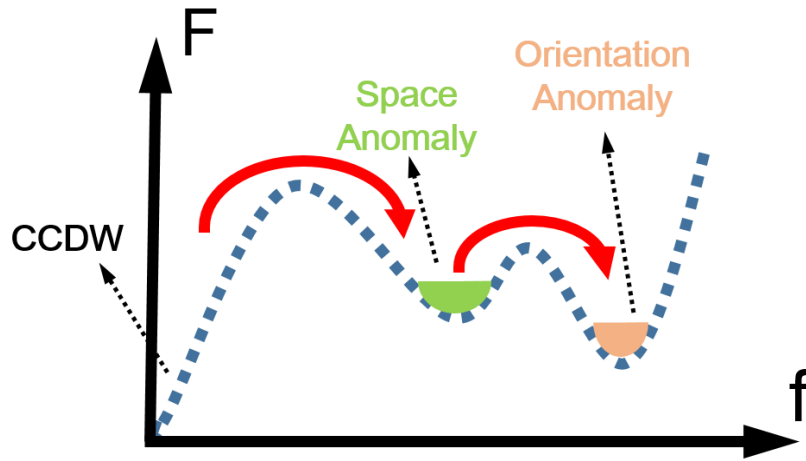


Fig. S4. CDW free energy schematic. (Color online) Schematic diagram of energy wells, illustrating the fluence-dependent switches of the space-anomaly H-CDW state and the orientation-anomaly H-CDW state.

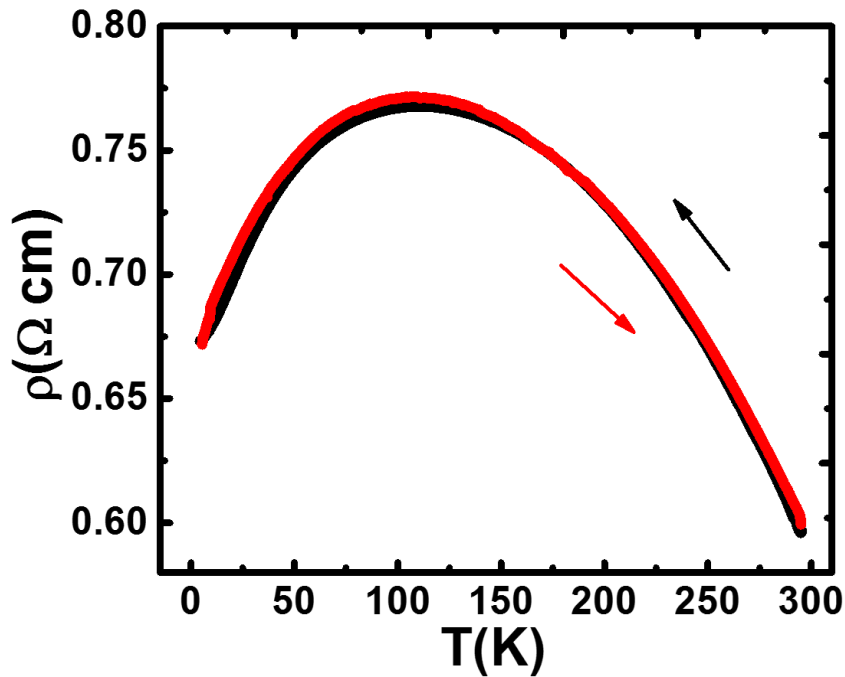


Fig. S5. Resistivity curve of 1T-TaS₂ with photoexcitation. Temperature dependence of the resistivity on temperature cycling 1T-TaS_{2-x}Se_x with $x = 1$. According to the electronic phase diagram (Fig. 1), there is no Mott state in $x=1$ sample. This sample performs superconductivity below ~ 3.6 K. We performed the ultrafast photoexcitation experiment in this sample at 4 K, 10 K and 20 K, but the resistivity has no change.

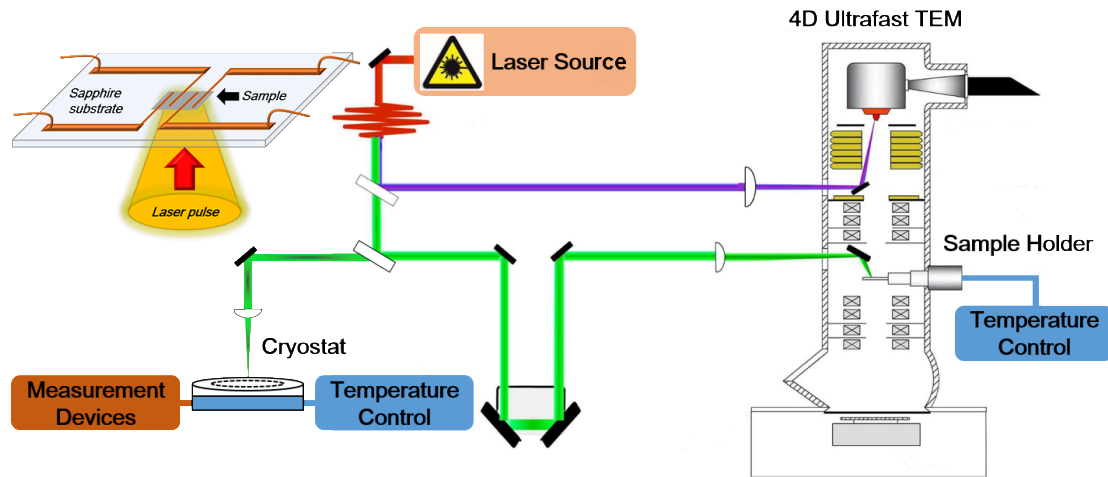


Fig. S6. Schematic diagram of the experimental setup. Schematic diagram of the experimental setup for in-situ photoexcitation measurements, as used inline with a 4D-ultrafast TEM (UTEM). The laser source can be shared for the measurements of physical properties and in-situ UTEM microstructure investigation. The inset displays the schematic of the experimental configuration of resistivity measurement; an exfoliated single sample on the sapphire substrate is used for resistivity measurements. Laser pulse illuminates the back of the sample.