

Charge density waves (CDW) are an emergent periodic modulation of the electron density that permeates crystals with strong electron-lattice coupling. Tantalum sulfide ( $\text{TaS}_2$ ) and related materials host several CDW states that spontaneously break crystal symmetries, mediate metal-insulator transitions, and compete with superconductivity. The quantum states are promising candidates for novel devices, efficient ultrafast nonvolatile switching and suggest elusive chiral superconductivity.

Here, PARADIM's Electron Microscopy Facility enables users to explore a long-range order CDW state in engineered interdigitated polytypes of  $\text{TaS}_2$ . Originally dominated by a conductive nearly commensurate CDW the material transitions to an insulating twinned-commensurate CDW phase as polytype interleaving isolates and decouples monolayers that host commensurate 2D-CDWs. Combining structural and electronic investigations the users show that this new approach suppresses an intermediate non-commensurate phase and pushes the metal-insulator transition to well above room temperature.

To close the loop of their materials discovery the users developed a model to illustrate the pathway for accessing these coherent 2D quantum states through heat treatment of the host crystal, initializing the system to an incommensurate phase that chooses either of the commensurate structures when cooled down.

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