

Electrochemistry in an MeV UED System: The Structural Dynamics of *In Situ* Lithium-intercalated WTe₂

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Layered transition metal dichalcogenides (TMDs) are a class of materials with remarkable electronic, optical, mechanical and chemical properties, holding promise for a wide range of applications such as nanoelectronics, sensing and catalysis. TMDs consist of covalently bound quasi-2D layers of transition metal atoms sandwiched between chalcogen atoms. These layers are weakly bound in the out-of-plane direction by van der Waals forces. Alkali metal intercalation into this van der Waals gap is an effective pathway for modulating the structure and properties of TMDs. For example, in MoS₂, lithium intercalation has been shown to cause a structural phase transformation accompanied by a semiconductor-to-metal transition ^[1].

In this work we explore the effects of lithium intercalation into the Weyl semimetal WTe₂, a TMD with a multitude of unique physical properties and with non-equilibrium phases that are accessible by optical excitation of a 0.24 THz shear mode on ultrafast timescales ^[2]. Using a combination of electrochemical, optical, X-ray diffraction and electron diffraction techniques, as well as density functional theory calculations, we confirm the formation of a novel TMD polytype in lithiated WTe₂, accompanied by symmetry breaking and large-scale lattice expansion.

We developed an ultra high vacuum-compatible all-solid electron-transparent electrochemical platform for the MeV ultrafast electron diffraction (UED) facility at SLAC National Accelerator Laboratory to investigate the ultrafast structural dynamics of WTe₂ *in situ* during controlled lithium intercalation (Fig. 1). We confirm electrochemical control over the structure of individual nanoscale WTe₂ flakes in the UED system using electron diffraction. We track the evolution of the ultrafast structural dynamics of a WTe₂ flake at different stages of the lithiation process in a pump probe regime, exciting the flake with 800 nm wavelength, ~100 fs light pulses and probing its structure using diffraction from ~3 MeV, ~100 fs electron pulses. We observe changes in the amplitude and damping time constant of the 0.24 THz shear mode in WTe₂ with lithium doping. In the lithium-induced novel WTe₂ polytype the 0.24 THz shear mode can no longer be excited with 800 nm light, instead we observe anisotropic in-plane coupling to laser-induced out-of-plane acoustic modes (Fig. 2).

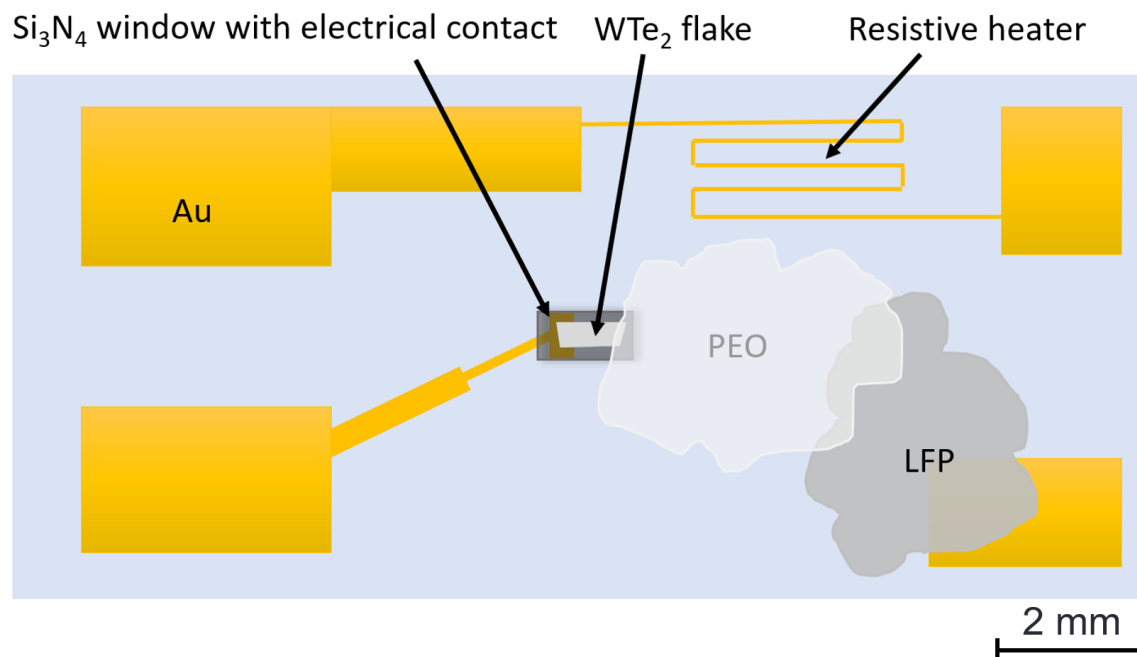


Figure 1. The solid-state electrochemical lithium intercalation platform. A single flake WTe_2 is stamped onto an electron-transparent Si_3N_4 membrane and contacted to both gold electrodes and Poly(ethylene oxide) (PEO) electrolyte. Lithium iron phosphate (LFP) is used as the lithium source.

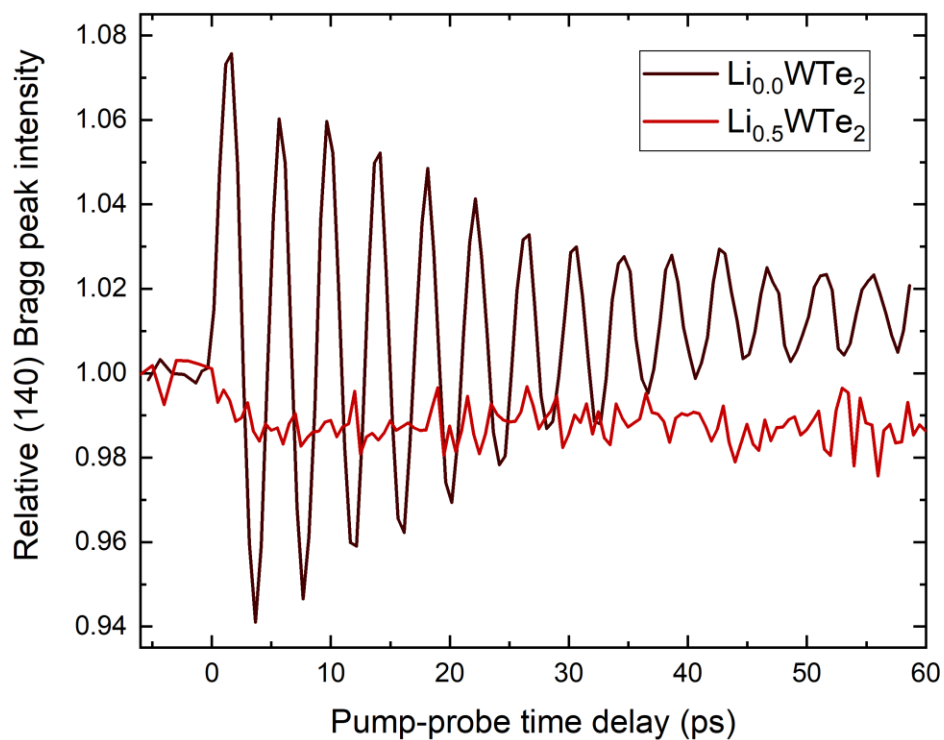


Figure 2. Changes of the ultrafast structural dynamics of WTe₂ after excitation with 800 nm wavelength light pulses. The intensity of the (140) Bragg reflection from MeV electron diffraction is shown as a function of pump-probe time delay.

References

- [1] L. Zhang *et al.*, Electrochemical Reaction Mechanism of the MoS₂ Electrode in a Lithium-Ion Cell Revealed by in Situ and Operando X-ray Absorption Spectroscopy. *Nano Lett.* 2018, 18, 2, 1466-1475
- [2] Sie, E.J., Nyby, C.M., Pemmaraju, C.D. *et al.* An ultrafast symmetry switch in a Weyl semimetal. *Nature* 565, 61–66 (2019).