Lithiation Induced Phases in 1T.-$MoTe_2$ Nanoflakes

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Abstract:

Molybdenum ditelluride (MoTe2) is a layered, two-dimensional (2D) crystal that naturally exists in three different structural phases. Each phase exhibits distinct optical and electronic properties. One way to induce new phases in 2D materials is through the reversible insertion of ions, atoms, or molecules into the gaps of crystalline materials, otherwise known as intercalation. Here we report electrochemical intercalation of lithium (Li) into the distorted octahedral or 1T'-phase of MoTe2 nanoflakes, leading to the discovery of two previously unreported lithiated phases [1].

Summary of Research:

We assembled coin-type cells with 1T'-MoTe2 powder to induce two, distinct and reversible phase transitions upon lithium intercalation. These phases are denoted as lightly lithiated phase I and heavily lithiated phase II (Figure 1). We also fabricated electrochemical cells on individual nanodevices using exfoliated crystals (Figure 2), which allowed for investigations into structure and electrochemical property changes in situ as we directly control intercalation of lithium ions into the 2D crystal. Structural differences between the 1T' and lithiated phase of MoTe2 were characterized by in situ Raman spectroscopy, and a change from the previously metallic to semiconducting phase upon lithiation was revealed through in situ transport measurements (Figure 3). In situ Hall measurements indicated bandgap opening in the lithiated phases, as evidenced by a significantly reduced Hall carrier density, and increasing resistance with decreasing temperature. Changes in structure for the heavily lithiated phase II was further analyzed using in situ angle-resolved Raman spectroscopy, in situ single-crystal x-ray diffraction (XRD), and in situ transition electron microscopy (TEM).

Conclusions and Future Steps:

Successful application of in situ experiments enabled detailed investigations of crystal structure and electronic properties of MoTe2 nanoflakes during electrochemical lithium intercalation. The discovery of new phases in initially metallic 1T'-MoTe2 highlights the effectiveness of electrochemical intercalation in adjusting phase stability and electron density in 2D materials.

References:

[1] S. Xu, K. Evans-Lutterodt, S. Li, N. L. Williams, B. Hou, J. J. Huang, M. G. Boebinger, S. Lee, M. Wang, A. Singer, P. Guo, D. Y. Qiu, and J. J. Cha, Lithiation Induced Phases in 1T'-MoTe2 Nanoflakes, DOI: 10.1021/acsnano.4c06330.

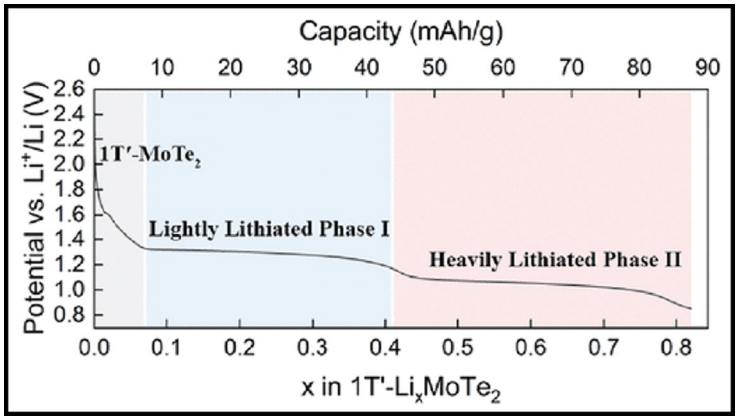

Figure 1: Galvanostatic discharging of 1T'-MoTe2 powder in a coin cell showing two distinct, intermediate phases upon 1T'-MoTe2 lithiation.

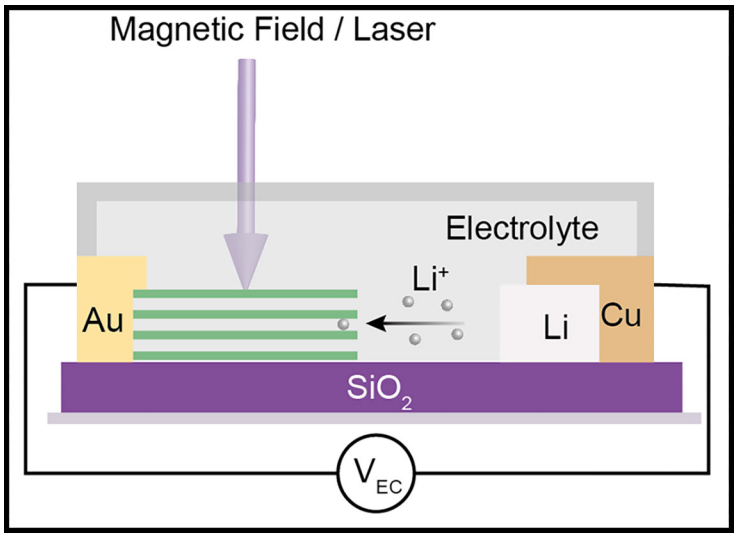

Figure 2: A schematic of a planar electrochemical intercalation cell capable of in situ experiments, such as for Raman spectroscopy, single-crystal XRD, and transport measurements.

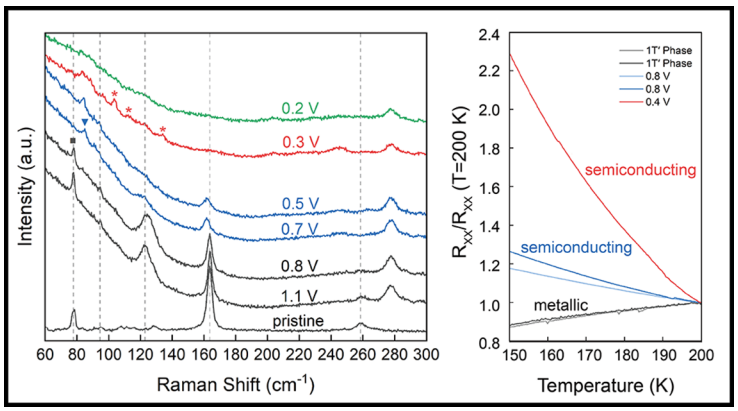

Figure 3: (Left) New structures of lithium intercalated 1T'-MoTe2, namely lightly lithiated phase I and heavily lithiated phase II. (Right) Initially metallic 1T'-MoTe2 is transformed into lithiated, semiconducting phases as confirmed by transport measurements, showing increasing resistance with decreasing temperature.