Optoelectronic Modulation in 2D Mo_{1-x}W_xTe₂ Monolayers

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Alloys of transition metal dichalcogenides (TMDs), such as Mo_{1-x}W_xTe₂, exhibit a wide range of electronic properties (semiconducting, semimetal and metallic phases) with unique polymorphs that depend on atomic stacking and coordination. However, reversible tunability between such phases is challenging. This is largely due to a combination of differences in the free energy between TMD polymorphs and phase transition kinetic barriers. Phase modulation in MoTe₂ has, however, gained recent interest due to the low barrier for transition (~40 meV) between its semiconducting and semimetal phases when compared to other TMDs. Evidently, dynamic control between the thermodynamically favorable semiconducting 2H phase and the metastable semimetal 1T' phase in MoTe₂ is achieved with a variety of external stimuli, including strain, temperature and electrostatic doping. In the latter case, theory predicts that this phase transition occurs when the charge density exceeds ~10¹⁴ cm⁻², which has been experimentally verified in monolayer MoTe₂ using ionic gating. However, reversible switching between these phases with a solid-state electrostatic gate still remains elusive. To circumvent the charge density requirements, theory predicts that the barrier for phase transition can be reduced in MoTe₂ when alloyed with tungsten. Evidence of this was demonstrated by Zhang et al. (arXiv:1709.03835), where an all solid-state resistive random-access memory device fabricated with Mo_{1-x}W_xTe₂ showed reversible switching between high and low resistance states. In addition to changes in conductivity, another feature of the phase transition in MoTe₂ includes in-plane structural changes that should give rise to district optical anisotropic responses in monolayers which remains highly unexplored.

We investigate and compare changes in the optical response of $2H\text{-MoTe}_2$ and $2H\text{-Mo}_{1x}W_xTe_2$ alloys as they undergo phase transition between semiconductor-to-semimetal phases in an all vdW device structure using an hBN gate dielectric and graphene contacts. The optical response of the vdW stack are investigated using angle- and polarization-dependent reflection measurements as a function of gate voltage. Initial results showed changes in the optical response of the vdW stack between $(1.1-1.4\mu\text{m})$ with applied gate voltage in alloys with x=0.09. Furthermore, polarization and temperature-dependent Raman measurements are also performed to map out structural changes in $Mo_{1-x}W_xTe_2$ as a function of applied gate voltage. These results will provide new insight into the optical response of these materials to enable new avenues for application in low-voltage and ultrafast modulators and other nanophotonic devices.

Supplementary Information

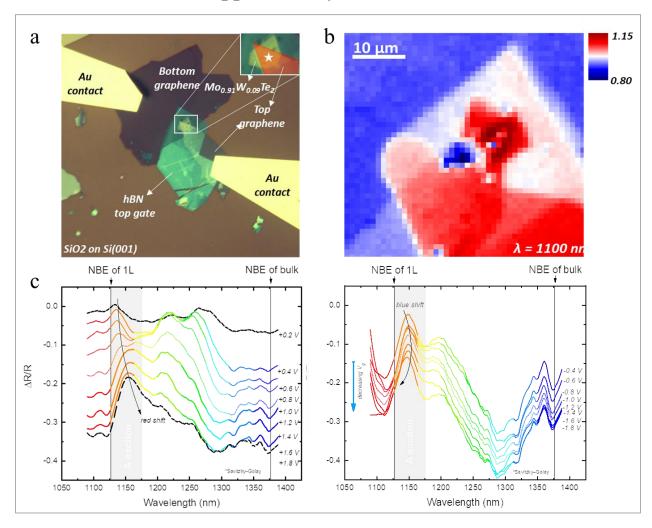


Figure 1: (a) Optical micrograph of device structure. (b) Reflection map of the device structure collected at 1100 nm. (c) Gate dependent reflection measurements of the A exciton of $2H-Mo_{1-x}W_xTe_2$ monolayers.