Characterizing Insulating States in TaS_2 with Atomic Adsorbates

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Atomic Adsorbates as a Characterization Tool

- Can add states (filled or empty) to the valance band (VB) or conduction band (CB)
- Donate electrons to the CB → n-type doping
- Accept electrons from the VB → p-type doping
- Can disrupt electron-electron interactions, breaking Mott insulator state
- Probing electronic response to atomic adsorbates can give further info on material properties

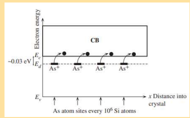


Figure 1: n-type doping schematic [6]

Tantalum Disulfide (TaS_2) – An Exciting Puzzle

- TaS_2 is a complex electronic system with many coupled interactions (spin-orbit coupling, layer hopping) and interesting electronic phases (charge-density wave states)
- When cooled, it goes through a series of phase transitions, eventually entering an insulating state [1]
- The type of insulating state is debated: conventional band insulator or Mott insulator
- Mott insulators: should conduct electricity according to band theory. Instead insulating due to strong electron-electron interactions
- If atomic adsorbates (adatoms) dope the sample, as indicated by band gap shift (but gap size unchanged) → band insulator
- If the band gap reduces significantly, the material behaves like a metal → Mott insulator

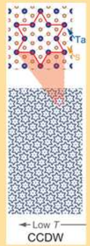
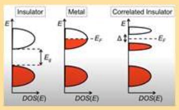


Figure 2: David star shaped charge density wave in cooled TaS_2 [5]



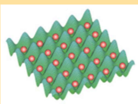


Figure 3: Density of states vs. energy for different material phases [4] (left)

Mott insulator state (right). Atoms localized at lattice sites because of interatomic interactions

Experimental Setup and Methods

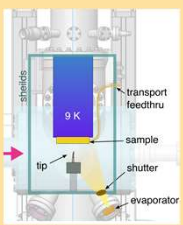


Figure 4: STM and evaporator setup (from RHK Technology)

Gold Evaporation

- Evaporator is set up to deposit gold on sample surface in a UHV chamber. Deposition for 30s at 1mA emission current
- 1000V is applied between ends of a tungsten filament, released electrons knock out gold atoms from nearby wire

Scanning Tunneling Microscopy/Spectroscopy

- An imaging technique used to study the electronic properties and topography of materials on the atomic scale.
- Works by scanning a metallic tip over a conductive sample
- Uses quantum tunneling of electrons across a nanometer-scale gap
- Exponentially depends on the vertical distance
- Sensitive to changes in local density of states

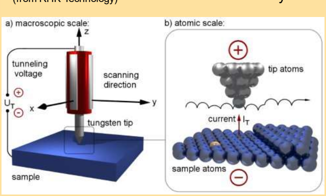


Figure 5: (a) Macroscopic model of an STM.
(b) Atomic scale model of an STM [3]

$$I \propto e^{-2z\sqrt{2m\phi}/\hbar} \int_0^{eV} \rho_s(\varepsilon)\, d\varepsilon$$

z = Tip-sample separation
ϕ = Sample work function
V = Bias Voltage
ρ_s = Sample DOS

Results

Finding Gold Adatoms on the Surface

- High resolution STM imaging to discern gold atoms on the sample surface
- Adsorption energy is found to be minimized at the center of David star charge density waves [2]

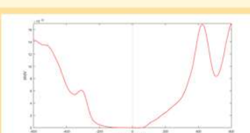


Figure 6: Typical density of states of bare TaS2 (corresponds to red dot in Figure 7, left)

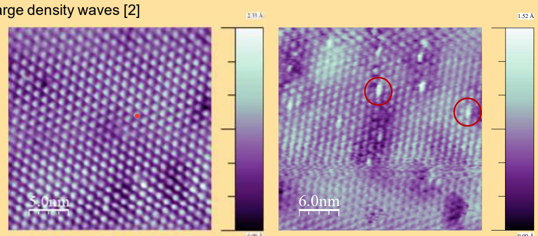


Figure 7: STM images of bare TaS_2 surface (left) and gold-speckled TaS_2 (left). Bright spots are gold atoms (examples circled in red)

Investigating the Gold and its Surroundings

- Performed dI/dV point-spectroscopy on the gold adatom, far away, and in proximity to the gold (in the dark areas) to probe electronic response of TaS_2
- **Far away:** Typical TaS_2 spectrum
- **On gold:** Small band gap, fitting for metals
- **Near the gold:** Tall peaks. Likely states that are being introduced by the gold. This hypothesis is currently being tested on standard graphite crystal with gold adatoms

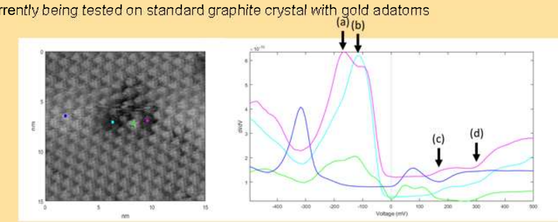


Figure 8: dI/dV point-spectroscopy of TaS_2 near gold adatom. Left: topography, right: waterfall plot. Data taken at each marked point

- Performed dI/dV spectroscopy along a line, starting far away from the gold, then closer, on the gold, and farther again
- Peaks that define the band gap shift to the right as we approach the gold and shift back as we move away from it. Gap size remains unchanged. Evidence the gold is doping the sample → insulating state being imaged is a band insulator

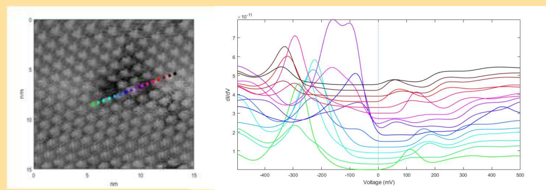


Figure 9: dI/dV line spectroscopy of TaS_2 near gold adatom. Left: topography, right: waterfall plot. Data taken at each point along the outlined path

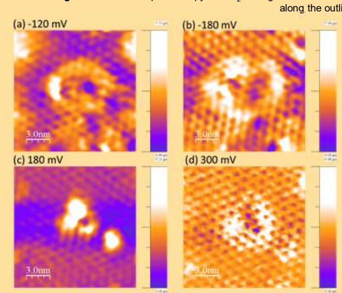


Figure 9: dI/dV maps of TaS_2 near gold adatom taken at different bias voltages. (a), (b), (c), (d) correspond to markers in Figure 8.

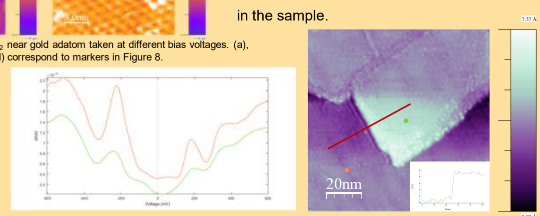
- To further investigate doping effects: took a map of electronic states at key peaks in the line spectra
- 180mV spectrum deprived of states

Stacking Order Dependence

- Literature suggest that stacking order of crystal layers determines the type of insulating state (odd # of layers → mott, even # of layers → band). Can test this by finding a step-edge in the sample.

Figure 10: Step edge in TaS_2 sample (right). Height profile (inset) shows single layer step, because TaS2 lattice constant in z-direction is 5.9Å

dI/dV point-spectroscopy taken on and off the step (left) demonstrate difference in electronic properties in different layers.



Future Work

- Explore the effect of stacking order on the type of insulating state and find Mott insulator surface
- Compare inherent doping vs. external doping

Acknowledgements

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References

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