Interfacial Optical Sensing of Ferroelectricity in Freestanding Perovskite Oxide by Using Monolayer Transition Metal Dichalcogenides

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Primary CNF Tools Used: SC4500 Odd-Hour Evaporator, YES Asher, Anatech Resist Strip, GCA 6300 DSW 5X g-line Wafer Stepper, Heidelberg Mask Writer - DWL2000, Dicing Saw - DISCO, Wire Bonder

Abstract:

Two-dimensional (2D) transition metal dichalcogenides (TMDs) can be easily integrated with other functional materials such as ferroelectric materials due to their lack of dangling bonds. Also, owing to their atomic thickness, electronic and photoluminescence (PL) properties of 2D TMDs can be modulated by external perturbations, which opens new avenues for quantum sensing and optoelectronic devices. In this work, monolayer WSe_2 is integrated with a freestanding perovskite oxide $BaTiO_3$ (BTO) membrane. We observe that the relative density of charge carriers in WSe_2 changes as the polarization switches, and this gives rise to the PL intensity modulation. The relative emission intensity of neutral excitons (X^0) and trions (X^T) shows gate dependent hysteresis, which confirms that WSe_2 senses and optically reads out the ferroelectricity in BTO.

Summary of Research:

Perovskite oxides BTO have strong spontaneous polarization and moderate coercive field [1], but their epitaxial mother substrates often limit the range of device structures. Recent studies showed that perovskite oxides can be released from the substrate by using a sacrificial oxide layer [2-7], which allows integration with Si-based substrates. In our work, BTO is epitaxially grown on a $Sr_3Al_2O_6$ sacrificial layer and released in water. Isolated BTO is transferred onto a SiO_2/Si substrate (BTO growth and transfer done by Kevin Crust from the Hwang group at Stanford).

After this transfer, small BTO membranes with suitable size for device fabrication are picked up by using a polymer stamping method. Then, this freestanding BTO membrane is interfaced with WSe_2 to form a field effect device. In this device, the polarization in BTO can be switched *in-situ* by applying a gate voltage, and the polarization-induced PL modulation in monolayer WSe_2 is studied.

To study the ferroelectricity induced optical properties change in monolayer WSe_2, two voltage application modes are used: DC mode and pulse mode. In the DC mode, a continuous DC voltage is applied, and in the pulse mode, a DC mode voltage is applied for five seconds and then removed before taking spectra. In the DC mode, a negative voltage is applied to switch the polarization to an out-of-plane direction. When the polarization in BTO is in an out-of-plane direction, holes are induced in BTO near WSe_2/BTO.

In response to this polarization induced charges, the electron population increases in WSe_2, which facilitates X^0 formation in p-type WSe_2. As in Figure 1, X^0 emission intensity is enhanced when -6V was applied. When a positive voltage is applied, the polarization in BTO is flipped to an opposite direction, and electrons in BTO are accumulated near WSe_2/BTO. This results in hole accumulation in WSe_2 and leads to positively charged trion (X^+) emission intensity enhancement (Figure 1). When the voltage sweep direction is reversed, the PL modulation shows different behavior as in Figure 2. This asymmetry is the manifestation of the ferroelectricity of BTO. To further corroborate this, X^0 and X^+ emission peaks were resolved by using the Gaussian fitting method and their relative emission intensity (X^0/X^+) is plotted as a function of the gate voltage (Figure 3). X^0/X^+ shows a hysteresis loop, which confirms that the PL modulation in WSe_2 is induced by the polarization switching in BTO.

In the pulse mode, the hysteresis direction is changed (Figure 4). This is possibly due to the polarization decay after the voltage removal. Further study is needed to understand the mechanism of this hysteresis direction reversal in the pulse mode.

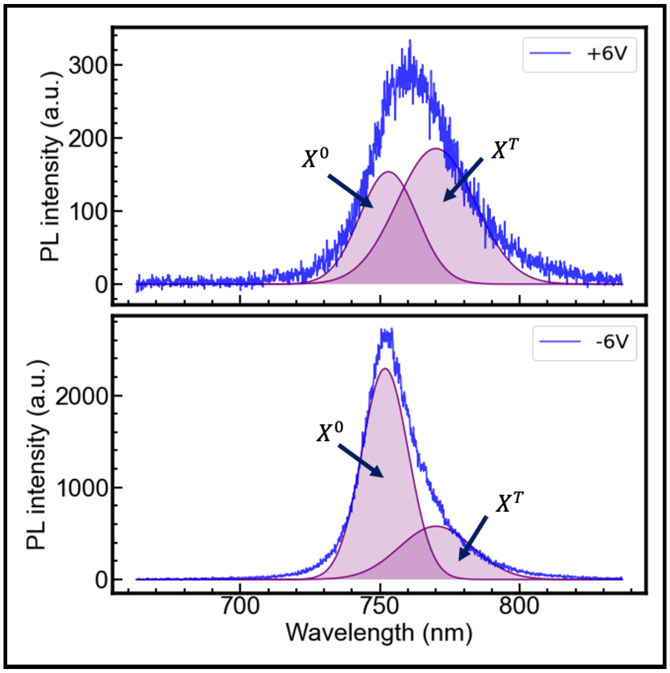


Figure 1: PL spectra at different gate voltage. X^0 and X^T emission is resolved by using the Gaussian fitting method. Spectrum at +6V (top) and spectrum at -6V (bottom).

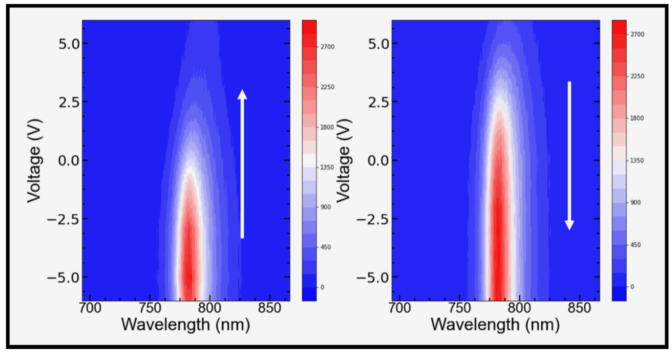
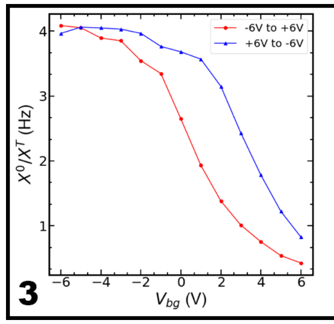


Figure 2: PL intensity modulation as a function of a gate voltage. PL intensity modulation shows different behavior when the voltage sweep direction is reversed.



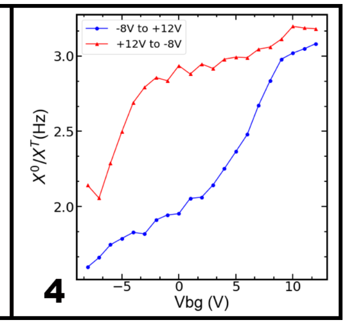


Figure 3, left: Relative ratio of X^0 and X^T emission (X^0/X^T) is plotted as a function of the DC gate voltage.

Figure 4, right: Relative ratio of X^0 and X^T emission (X^0/X^T) is plotted as a function of the pulsed gate voltage.

Conclusions and Future Steps:

In this work, we fabricate a freestanding BTO and monolayer WSe_2 based field effect device. The relative emission intensity of X^0 and X^T changes with the gate voltage and shows the hysteresis loop, which confirms that WSe_2 senses and optically reads out the polarization switching in BTO. As a next step, we will study the thickness dependent ferroelectricity in BTO by interfacing WSe_2 with BTO of various thickness. Also, BTO is known to have various phases at different temperatures [8]. We will explore the phase transitions in BTO by varying the temperature and study the optical and electronic property change in WSe_2 in response to the phase transition of BTO.

References:

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