Engineering Correlated Insulators in Moiré Twisted Homobilayer Transitional Metal Dichalcogenide

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Abstract

In this report, we demonstrated the fabrication of the Moiré twisted homobilayer (tHBL) $MoTe_2$ with twisting angle 3.57° and used optical and electrical methods to characterize the properties of the device. We found that at various filling factors ($\boldsymbol{\nu = -1, 1, 5/3}$), the doping dependence Reflectance Contrast (RC) are showing enhanced oscillator strength in exciton resonance. The correlated insulator are further confirmed using doping dependent Photoluminescence (PL), where the trion emission reduced due to formation of insulator at the doping level. In the future, we are going to demonstrate the magnetic response of the system to the applied magnetic field and circular dichroism.

Keywords: TMDc, Correlated Insulator

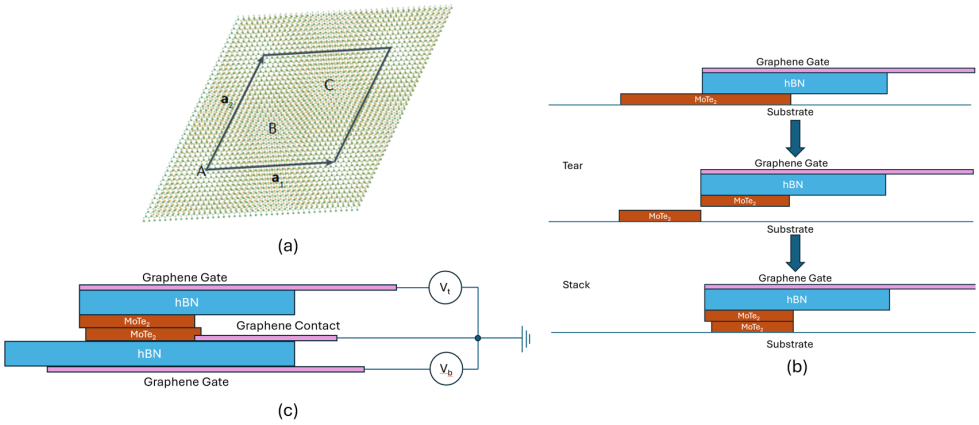

Fig. 1 **a.** Moire superlattices. **b.** Tear and stack procedures. **c.** Sample design diagram. **a.** is sourced from [1].

1 Introduction

Transition metal dichalcogenides (TMDcs) offer a versatile platform for the engineering and exploration of electronic and excitonic states in semiconductor materials, exhibiting direct bandgap transitions and light emission across the visible to near-infrared spectrum due to excitons. In their bulk form, TMDcs are maintained by Van der Waals forces which are foundational to 'Twistronics.' Twistronics involve the use of Van der Waals forces to stack and twist TMDc layers into moire devices where the original lattices are remodulated and realign into moire superlattices (Figure 1). The periodicity of this superlattice, denoted by a_M, is calculated using the formula $a_M = a_0/\sqrt{\theta_0^2 + \epsilon^2}$. The moiré lattice thereby introduces an unprecedented degree of freedom in tuning the band structure, electronic states, and excitonic states within the material.

From then on, Twistronics has emerged as a critical pathway for investigating correlated electronic states. Numerous studies have documented observations of correlated and many-body states in twisted moiré TMDc samples, demonstrating that exciton analysis is an effective method for determining electronic states [2, 3]. To analytically understand the dynamics behind the Twistronics, the Moire band hamiltonian is

$$H_0 = -\hbar^2\mathbf{k}^2/2m^\star + \Delta(\mathbf{r}) \tag{1}$$

In the flat band limit [1], the isolated moire band hamiltonian using tight banding model is

$$H_0 = \sum_{\mathbf{R},\mathbf{R}',\tau} t\left(\mathbf{R}' - \mathbf{R}\right) c^\dagger_{\mathbf{R}'\tau} c_{\mathbf{R}\tau}, \tag{2}$$

where t is the hopping parameter between the moire lattice. At minimal twist angles, like near-0° (R-stacked) and near-60° (H-stacked), the moiré pattern's large periodicity limits electron hopping and heightens Coulombic interactions, leading to diverse

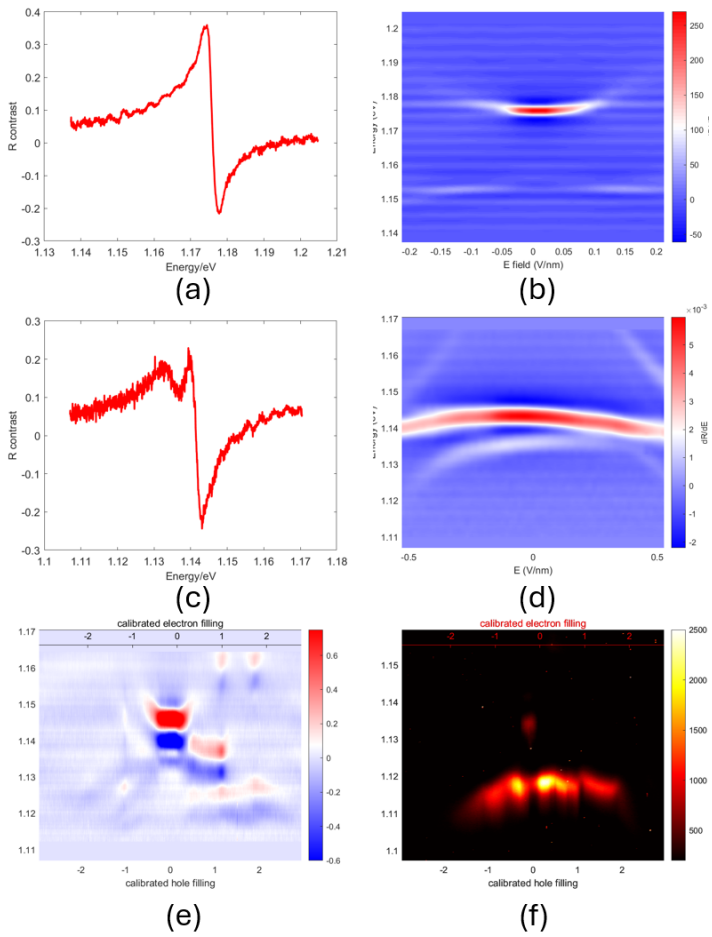

Fig. 2 **a.** This shows the RC of the monolayer exciton resonance at 1.175eV. **b.** This shows the doping dependence first order derivative of the RC. **c.** The RC of the HBL intralayer excitons. Those intralayer excitons are having resonance at 1.135eV and 1.139eV. **d.** This shows the electric field sweeping of the differential RC, which confirms anti-crossing of interlayer exciton with the two intralayer excitons. The red-shifting of the interlayer exciton from higher energy relates to the out-of-plane dipole of the interlayer excitons. **e.** The doping dependent second order derivative RC. Both top and bottom x-axes are calibrated electron and hole filling factors injected by the dual gate capacitors. Colorbar represents the resonance amplitude of the spectrum. The white part between red and blue is the central resonance of the peak. **f.** This is the doping dependent PL of the HBL using 635nm laser. The higher energy peaks at zero doping is the intralayer exciton emission. After applying gate voltage to supply doping, the intralayer excitons quickly coupled with fermi sea to form trion.

correlated states such as Mott insulating state, incompressible state, Fermi superfluid states, and spin liquids. Homobilayer TMDcs, created by stacking two identical monolayers, show promise for investigating these correlated states.[3, 4]. In the fabrication of homobilayer moiré devices, I employ the tear-and-stack technique (Figure 1). The process begins with a polymer-based stamp with a top gate graphene and a hexagonal Boron Nitride (hBN). Then, I tear a portion of the $MoTe_2$ monolayer under the hbn

and stack onto the remaining $MoTe_2$ monolayer with a twisting angle. To search for correlated states in $MoTe_2$ twisted homobilayer system, I add an electrode to ground the sample and provide doping (Figure 1).

2 Results

To probe the electronic and correlated states, I perform electrical and optical characterization to my $MoTe_2$ tHBL sample. The initial phase is to check device functionality through the monolayer $MoTe_2$ optical and electrical properties. As depicted in Figure 2, the reflection contrast spectrum of the monolayer $MoTe_2$ reveals a pronounced exciton resonance, dipping at 1.18 eV. The Loranzian peak in RC is characterized by a notably strong oscillator strength, indicating the quality of the transitions within the material.

With doping, the excitons couple with the Fermi sea, leading to the formation of composite quasiparticles known as polarons, as illustrated in Figure 2. At relatively high doping regiemes, polarons are prone to exhibit blue shifts. The proper characterization of the monolayer tells that my sample was functioning correctly so the data coming from the characterization in bilayer region is reliable.

To probe and demonstrate the electronic state in the tHBL, I apply the same characterization method plus out-of-plane electric field sweeping. Without doping, tHBL RC indicates two intralayer exciton absorption at 1.135eV and 1.139eV (See Figure 2 (c.)). The two intralyer excitons does not belong to specific layers in the HBL, but they are coming from the interlayer coupling between the top and bottom layer. After the interlayer coupling, the K-valley of both layer splits into K+ and K-, whose intralayer exciton transitions are red-shifted to 1.135eV and 1.139eV due to the alignment of conduction and valence bands [5, 6].

The interlayer exciton also affects the energy of intralayer exciton because of the intra-interlayer exciton hybridization. Therefore, I applied out-of-plane electric field with zero doping in the sample. Figure 2 (d.) is the electric field sweeping, which exhibit the anti-crossing of the interlayer exciton with both intralayer exciton from higher energy, confirming the hybridizations.

To demonstrate the correlated insulator in the HBL system, I injected electrons and holes to the sample by applying the same bias to both top and bottom gate. Figure 2 (e.) illustrates the second order differential RC of the HBL system. The formation of the correlated insulator because of the Mott insulating states, generalized wigner crystal, or quantum hall effects [2, 7–9] will affect the formation of trionic states in the sample, which increases the oscillator strength of the excitonic transition in the RC spectrum. In Figure 2 (e.), there are multiple fillings which are exhibiting the local correlated insulating states with abrupt enhanced excitonic oscillator strength. Those states are corresponds to the filling facotr $\nu = -1, 1$, and 2.

To further confirm the insulating nature corresponds to those filling factors, I performed doping dependent PL to demonstrate the decrease in trion population. Figure 2 (f.) shows the doping dependent PL. With no electron or hole doping in the sample, there are two intralayer exciton peak emitting at 1.135eV and 1.139eV respectively, with a very weak trion emission. Then, after doping the sample, intralayer

excitons quickly couple with the electrons and holes injected to the sample and form trion. When reaching certain filling factors, the trion PL shows an abrupt change in the emission intensity with blue shifted dent. This is because the population reduction and the band renormalization due to the electronic environment change from the insulator. I matched the main dents of the PL spectrum with the RC spectrum, and they shows at the same filling factors where the RC spectrum resolves enhanced oscillator strength.

3 Conclusion

In this report, I have demonstrated the potential of twisted homobilayer $MoTe_2$ (tHBL) as a platform for investigating correlated electronic and excitonic states through the use of electrical and optical characterization methods. The combination of reflection contrast (RC) and photoluminescence (PL) techniques has proven effective in probing both the electronic structure and excitonic transitions within the twisted $MoTe_2$ bilayer system.

The RC data reveals distinct intralayer exciton absorption peaks, which shift in response to interlayer coupling. The hybridization of intra- and interlayer excitons is evidenced by the anti-crossing behavior of the interlayer exciton with intralayer excitons in the out-of-plane electric field sweeping.

Additionally, the doping-dependent PL and RC spectra demonstrate the presence of correlated insulating states at specific filling factors, marked by enhanced excitonic oscillator strength and a notable reduction in trion population. These observations align with the theoretical and experimental demonstration of Mott insulating states, Wigner crystals, and quantum Hall effects in the tHBL system, offering new insights into the role of moiré superlattices in engineering complex many-body states.

Overall, this work underscores the versatility of twisted TMDc homobilayers as a platform for exploring correlated phenomena, providing a deeper understanding of the interplay between electronic states and excitonic dynamics. In the future, we are planning to apply magnetic field to observe the magnetic response of different correlated insulator using circular dichroism. Also, the circular dichroism is able to probe the already reported time-reversal (TR) symmetry broken phase in the hole-doping side of the experiment, which is critical for the realization of spontaneous TR breaking polariton if we are able to strongly couple the excitons to the photonic bands using photonic crystal or fabry-perot cavities.

4 Methods

4.1 Fabrication of the Sample

To fabricate the 2D materials used in this report, we exfoliate the 2H-$MoTe_2$ from H.Q. Graphene using white tape to the 90nm SiO_2 in the nitrogen gas filled glovebox and use optial microscope to search for monolayer.

To transfer the material onto the prepatterned gold electrodes, we used PDMS-PPC-PC stamp to transfer the stacked sample altogether. Both gates of the sample are monolayer graphene.

4.2 Calibration of the Filling Factors

The gate voltages are provided by the Keithley 2634B under the control of LabView script. The filling factors are calculated using $\nu = n/n_0$, where n is the doped electron population, and n_0 is the Moiré lattice density per centermeter sqaured. The Moiré lattice period is estimated to be $a_M = a_0/\sqrt{\theta_0^2 + \epsilon^2}$, and $n_0 = 4/(\sqrt{3}(100a_M)^2)$. n is calculated from the parallel plate capacitor $n = C_{top}V_{top} + C_{bot}V_{bot}$, where $C = \epsilon_h BN\epsilon_0/d$ with d is the thickness of hBN determined using Atomic Force Microscopy

and $\epsilon_h BN = 4.2$. To determine the transport barrier of the sample, we used the equation $\delta V = \frac{E_G e^2 D_i}{eC}$ where E_G is the electronic band gap of the $MoTe_2$ and D_i is the impurity density of state, which is estimated to the order of $10^12 cm^{-2} eV^{-1}$. The filling factor of the gap is around 0.154, which is the difference between the zero electron and hole doping filling factors.

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