

Measurements of Correlated Insulator Gaps in a Transition Metal Dichalcogenide Moiré Superlattice

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Abstract: Moiré superlattices of transitional metal dichalcogenides exhibit strong electron-electron interaction that has led to experimental observations of Mott insulators and generalized Wigner crystals. In this letter, we report direct measurements of the thermodynamic gaps of these correlated insulating states in a dual-gate WS_2/WSe_2 moiré bilayer. We employ the microwave impedance microscopy to probe the electronic features in both the graphene top gate and the moiré bilayer, from which we extract the doping dependence of the chemical potential of the moiré bilayer and the energy gaps for various correlated insulating states utilizing the Landau quantization of graphene. These energy gaps vary across different locations on the samples but are relatively insensitive to the application of an external electric field or magnetic field to the WS_2/WSe_2 moiré bilayer.

Keywords: *moiré superlattice, WS_2/WSe_2 , correlated insulating state, microwave impedance microscopy, energy gap measurement*

Moiré superlattices formed by two-dimensional (2D) transition metal dichalcogenide (TMD) layers have recently emerged as a promising platform to study the effects of strong correlation^{1–4}. A plethora of novel electronic states has been discovered, including Mott insulators^{5–8}, generalized Wigner crystal insulators at fractional fillings^{5,9–18}, and dipolar excitonic insulators^{19–23}. The strong electron correlation in these systems arises due to the formation of electronic flat bands in which the kinetic energy is substantially reduced and yields to the Coulomb interaction. Therefore, strong Coulomb repulsion can effectively localize carriers in the periodic moiré superlattice, leading to various charge-order states. Such correlated states can exhibit relatively high transition temperatures reflecting the strong correlation strength. For example, the Mott insulator state in the moiré superlattice of WS₂/WSe₂ survives above 180 K¹¹. Although the temperature dependence study gives a rough estimate of the bandgap, however, it does not directly lead to the quantitative measurement of energy gaps of these insulating states.

To gain deeper insights into the understanding and the engineering of correlated states, it is beneficial to characterize the correlation strength in a quantitative way by measuring the energy gaps of the various correlated insulating states in TMD moiré systems^{15,17,24–29}. Gap measurements in 2D correlated systems are in general challenging because the formation of the correlated states requires doping the system at appropriate carrier densities; hence, single particle spectroscopy techniques such as scanning tunneling spectroscopy and angle-resolved photoemission cannot be directly applied^{30–32}. A promising approach is to probe the chemical potential of 2D devices utilizing a sensing layer^{33–40}. In this work, we report a direct measurement of thermodynamic gaps of the correlated insulating states in the WS₂/WSe₂ moiré superlattice by sensing the chemical potential of the moiré bilayer as a function of carrier doping. The local probing nature of our scanning probe based technique further allows us to examine the spatial variation of the energy gaps in a moiré device. We find the energy gap at the Mott insulator state at one hole per moiré unit cell in the range of 40–80 meV and the gaps at fractional fillings of $-1/3$ and $-2/3$ in the range of 5–10 meV.

The schematic of the device is shown in Fig. 1a. (See Section 1 in Supporting Information for device fabrication details). A TMD heterobilayer is encapsulated by thin hBN flakes to form a dual-gate device geometry with monolayer graphene (MLG) as the top gate and thin (~ 10 nm) graphite as the bottom gate. We perform microwave impedance microscopy (MIM) measurements by parking a sharp metal tip over the device^{41–43}. A small microwave excitation (with a power of 1–10 μ W and frequency of ~ 10 GHz) is applied to the tip via an impedance-matching network. Oscillating electric fields at microwave frequency are generated near the tip apex and screened by the sample underneath. Changes in the sample's local electrical properties, including dielectric constant and conductivity, will induce changes in tip-sample admittance (the inverse of impedance), whose in-phase and out-of-phase components are acquired as the MIM-Re and MIM-Im signals, respectively, by analyzing the reflected microwave signal. With its

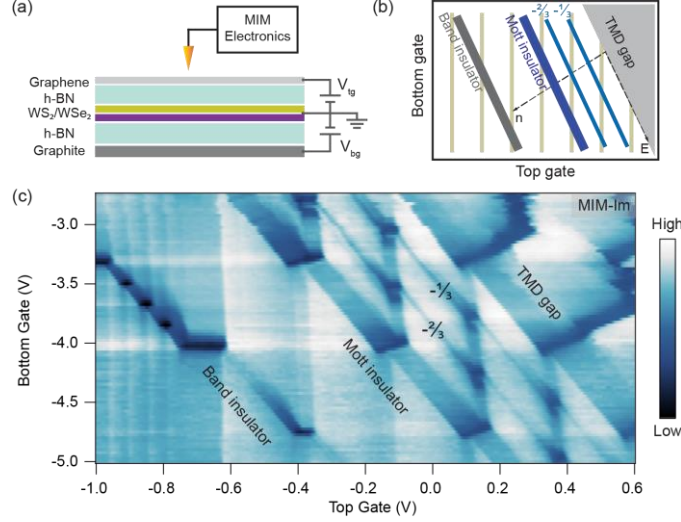


Figure 1. Probing correlated insulating states in a dual-gate WS_2/WSe_2 moiré device with microwave impedance microscopy. (a) Schematics of the measurement setup. (b) Expected features in a data map as a function of both top and bottom gate voltages. (c) Dual-gate map of MIM-Im in device D1 taken at 7 K and 9 T. The measurement spot corresponds to spot E in Fig. 3a.

sensitivity to local conductivity, MIM has been utilized to study the correlated insulating phases in semiconducting moiré superlattices by tuning the carrier density with a single bottom gate^{8,11}. In our dual-gate device, we find that when the MLG top gate is driven to the quantum Hall regime under a large out-of-plane magnetic field (above ~ 2 T), its bulk conductivity drops such that the microwave electric fields from the tip cannot be completely screened by the MLG top gate and are thus able to reach the TMD heterobilayer and probe its conductivity²⁰. The hBN thicknesses for the top and bottom gates are 5 and 16.3 nm, respectively, measured by atomic force microscopy and verified by the calibration of electron density via dual gate doping.

We record the MIM signals as a function of both top and bottom gate voltages to obtain a dual-gate map. A typical dual gate map of MIM-Im data measured at 9 T is presented in Fig. 1c. (See Section 7 of Supporting Information for more data at different magnetic fields). There are two main sets of features. The first set of features is the diagonal lines corresponding to insulating states in the TMD moiré bilayer. In a dual-gate structure, the carrier density of the moiré bilayer depends on both top and bottom gate voltages, which can be described by the following equation:

$$n_{\text{moiré}} = C_{tg}(V_{tg} + \frac{\mu_G - \mu_{\text{moiré}}}{e}) + C_{bg}(V_{bg} - \frac{\mu_{\text{moiré}}}{e}) \quad (1)$$

Here, C_{tg} (C_{bg}) is the geometric capacitance between the top gate (bottom gate) and the sample, $\mu_{\text{moiré}}$ and μ_G the chemical potentials of the moiré bilayer and MLG, respectively, e the electron charge, and V_{tg}

(V_{bg}) the voltage applied on the top (bottom) gate. The chemical potential change in the thin graphite bottom gate is minimal and has been neglected due to its large density of states. We note that Eq. (1) ignores a possible threshold voltage that can be present at the electrical contacts to ground the TMD device, but this term will be a constant in the linear regime and should not affect the analysis of chemical potential below. Each of these diagonal lines corresponds to a constant carrier density in the TMD moiré bilayer, and their pattern matches the expected filling factors, ν , defined as the number of carriers per moiré unit cell, for the correlated insulating states in the WS_2/WSe_2 moiré superlattice. The $\nu = -1$ state corresponds to a Mott insulator, the $\nu = -1/3$ and $-2/3$ states correspond to generalized Wigner crystal states, and the $\nu = -2$ state corresponds to the band insulator state with the Fermi level between the first and second moiré minibands in the WSe_2 layer.

The second set of features are a series of lines that appear at approximately constant top gate voltages which correspond to LL gaps in the MLG top gate²⁰. The carrier density in the MLG depends on the potential difference between the MLG and the TMD heterobilayer, which can be described by Equation (2)

$$n_G = C_{tg} \left(\frac{\mu_{\text{moire}} - \mu_G}{e} - V_{tg} \right) \quad (2)$$

These LL features are approximately equally spaced with a carrier density of $8.7 \times 10^{11} \text{ cm}^{-2}$ for graphene at the measurement magnetic field of 9 T. An illustration of these two sets of features is plotted in Fig. 1b. Based on Eq. (2), when the MLG is kept in a specific LL gap, both n_G and μ_G are fixed, and we find a relation: $\Delta\mu_{\text{moire}} = e\Delta V_{tg}$. We thus can extract the V_{tg} value for a particular LL as a function of V_{bg} , which is then converted to $\Delta\mu_{\text{moire}}$ vs n_{moire} . Fig. 2(a) plots a high-resolution dual-gate MIM-Im map taken at 8 K and 6 T. To track the LLs, we take the derivative $d(\text{MIM-Re})/dV_{tg}$ (Fig. 2b) in which the positions of LLs are sharply defined (See Section 7 of Supporting Information for raw MIM-Re data). For each LL, we follow the procedure described above to extract the curves of $\Delta\mu_{\text{moire}}$ vs n_{moire} . Of particular interest are the crossings of LLs with the insulating states in the TMD moiré bilayer. We find that the LLs shift their positions when they cross the insulating states in the TMD moiré bilayer. This is because the chemical potential of the TMD moiré bilayer will change across an insulating gap, therefore, the top gate voltage must also change by the same amount to compensate for it so that the carrier density in MLG is maintained at the same value for each specific LL. This behavior of the graphene LL and the ability for MIM to track features in both MLG and the TMD moiré bilayer allow us to directly extract the energy gaps of the correlated insulating states. Fig. 2c plots the processed μ_{moire} vs n_{moire} curves extracted by tracking several LLs around the crossings as indicated by the dotted lines in Fig. 2(b) (see Section 15 of Supporting Information for data processing details). The multiple sets of data overlap nicely, demonstrating the validity

of our analysis in the linear regime of the dual-gate map. The abrupt jumps in these curves correspond to the energy gaps at various insulating states including the Mott insulating state at -1 , the generalized Wigner crystals at $-1/3$, and $-2/3$, as well as the band insulator gap at -2 . The values of these gaps are listed in Table 1 together with the transition temperatures determined in bottom-gate only devices. Their general trends match qualitatively. The ratio of $\Delta\mu/k_B T_C$ is well beyond 1 which is typical for strongly correlated states^{17,44–46}.

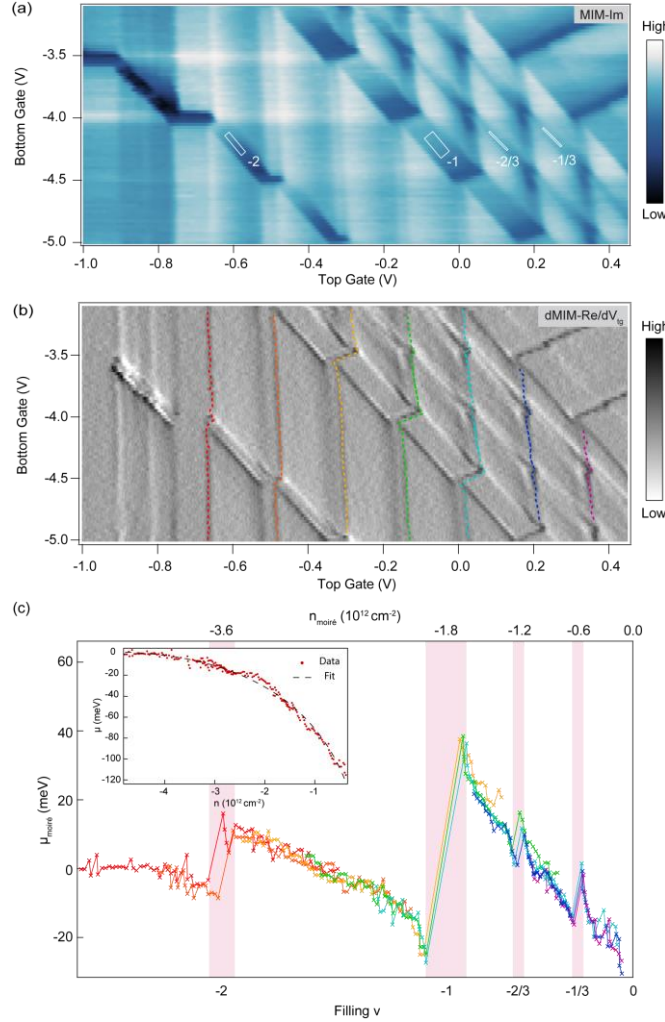


Figure 2. Extraction of chemical potential in TMD moiré bilayer. (a) $MIM-Im$ and (b) Derivative of $MIM-Re$, $d(MIM-Re)/dV_{tg}$, as a function of both top and back gate voltages, measured in device D1 at 8 K and 6 T. The measurement spot corresponds to spot D in Fig. 3a. Dash lines that track the LLs in the MLG are overlaid in (b). (c) Extracted chemical potentials of the TMD moiré bilayer plotted as a function of the filling factor. μ is referenced to 0 at the highest hole density. Line colors correspond to the dashed lines along different LLs as marked in (b). The inset shows the chemical potential after removing the gapped states. The dashed line presents a fit to the HF model.

ν	-1/3	-2/3	-1	-2
$\Delta\mu$ (meV)	8 ± 4	6 ± 2	55 ± 4	18 ± 4
T_c (K)	35	35	160	60
$\Delta\mu/k_B T_c$	2.6 ± 1.3	2.0 ± 0.7	4.0 ± 0.3	3.5 ± 0.7

Table 1. Weighted average thermodynamic energy gaps $\Delta\mu$ of the correlated insulating states in the WS₂/WSe₂ moiré superlattice. The gaps are extracted from Figure 2c (see Section 5 and 11 of Supporting Information for details), compared with their transition temperatures T_c ¹¹ and quantitative ratios of $\Delta\mu/k_B T_c$.

Each crossing of a LL with an insulating state in the TMD bilayer will provide a measurement of the insulating gap. In the experimentally accessible gate range, the insulating states cross multiple LLs as seen in Fig. 1, and the extracted chemical potential profiles along these LLs are plotted together in Fig. 2c. These crossings correspond to different out-of-plane displacement electric fields on the TMD bilayer, and our measurement results do not show any appreciable changes in the gap size within the accessible range of displacement fields in our experiment. We note that our observation of weak displacement field dependence of the gap size differs from reports in other TMD moiré system^{15,25,27}, most likely due to the band alignment in different material combinations. We further performed measurements at magnetic fields from 3 T to 9 T and the gap values do not vary significantly in this field range. This magnetic field independence possibly arises because spins in the correlated states are already fully polarized in the range of applied magnetic fields, rendering the gap size insensitive to the magnetic field, consistent with a previous report⁶. More data and detailed analysis on the dependence of electric and magnetic fields are available in Section 8 of Supporting Information.

Away from the insulator gaps, we find that the chemical potential generally decreases with increasing carrier density, exhibiting negative compressibility, which is particularly pronounced right before and after the insulating gap (as shown in Fig. 2c). This behavior is not expected from a single-particle picture in which adding carriers to an energy band should only increase the chemical potential, but a negative compressibility can occur in systems with strong electron-electron interactions^{27,47,48}. This increasingly negative compressibility observed with decreasing hole density can be well explained through the Hartree-Fock (HF) model for a 2D electron gas (2DEG)^{27,49}:

$$\mu(n) = \frac{\pi\hbar^2}{m^*}n - \left(\frac{8}{\pi}\right)^{\frac{1}{2}}\left(\frac{e^2}{4\pi\epsilon}\right)\sqrt{n} = \frac{\pi\hbar^2}{m^*}n - A\sqrt{n},$$

where the first term represents the kinetic energy ($\propto n$) and the second term corresponds to the exchange energy ($\propto -\sqrt{n}$), which dominates in low carrier density. In strongly correlated systems where kinetic energy is suppressed, such an imbalance extends to higher carrier density, resulting in the observed negative compressibility in a broad density range. To quantify this behavior, we plot the chemical potential of the metallic phases as a function of n , excluding the abrupt jumps associated with the gap states (Fig. 2c (inset)). Fitting the data to the HF model yields a parameter $A = (2.06 \pm 0.04) \times 10^{-4} \text{ meV} \cdot \text{cm}$, consistent in magnitude with the HF prediction (see Section 15 of Supporting Information for detailed analysis).

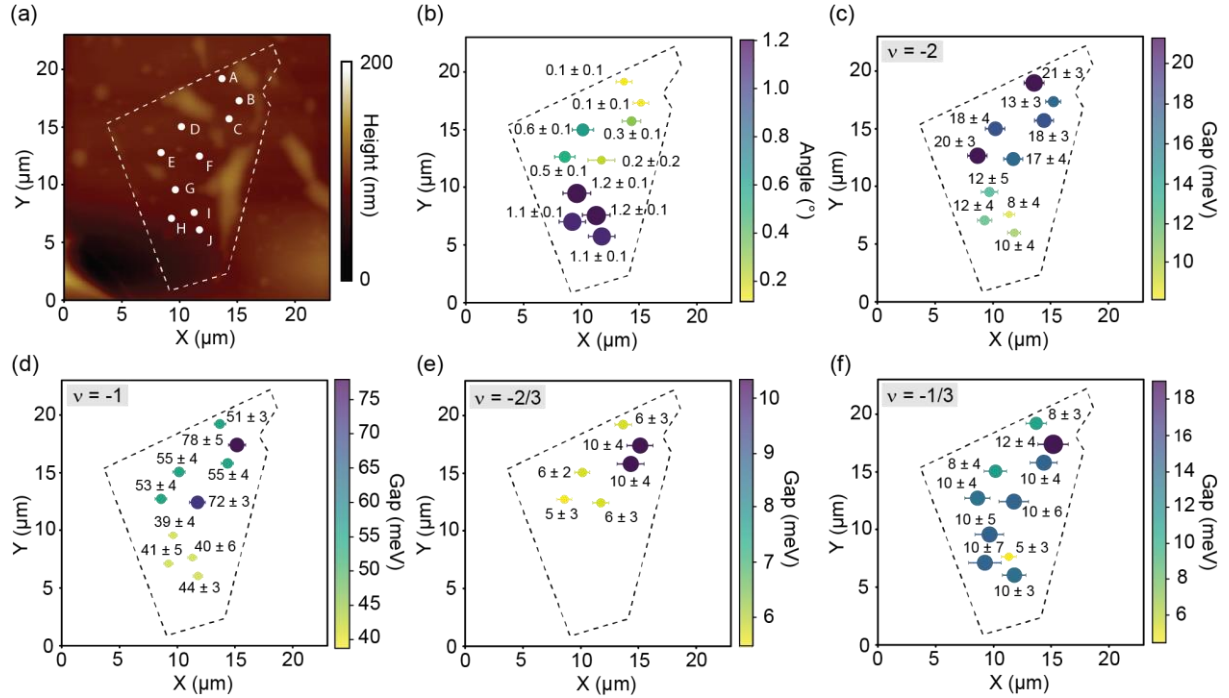


Figure 3. Spatial map of correlated insulating gaps. (a) AFM image of the WS₂/WSe₂ moiré superlattice device. The white dots indicate the selected spots for the gap measurement. The white dash line delineates the boundary of the dual-gate WS₂/WSe₂ moiré superlattice region. (b) Spatial map of the twist angle, derived from the gate voltage corresponding to the $\nu=-1$ and $\nu=-2$ states (see Section 6 of Supporting Information for details). (c)-(f) Spatial map of the average correlated insulating gaps at states (c) $\nu=-2$, (d) $\nu=-1$, (e) $\nu=-2/3$ and (f) $\nu=-1/3$. The twist angles and energy gaps are represented by both the color and the diameter of each spot. The length of the error bars indicates the uncertainties in the twist angles and energy gaps.

With the capability to extract gap values from single-point gate spectroscopy data, we further carry out measurements at various locations across the sample to examine the spatial variation of the correlated insulator gaps. Fig. 3a is an atomic force microscopy (AFM) image of device D1. Ten spots are selected across the sample region of approximately $15 \mu\text{m} \times 7 \mu\text{m}$, labeled as A through J in Fig. 3a. At each spot, the local twist angle can be determined by the moiré density calculated from the gate dependence data. The

angle varies from 0.1° to 1.2° (Fig. 3b), likely due to the presence of air bubbles as seen in the AFM image. We further extracted the weighted average energy gaps (see Section 11 of Supporting Information for details) of each spot for insulating states at $\nu=-2$, -1 , $-2/3$, and $-1/3$, and the results are plotted in Fig. 3c-f. The MIM data for all the spots are available in Section 10 of the Supporting Information. The six spots (A-F) in the upper half of the device exhibit larger energy gaps at $\nu=-1$ and -2 than the four spots (G-J) in the lower half. At $\nu=-1/3$, the energy gaps are similar (~ 10 meV) across all the spots. At $\nu=-2/3$, spots A-F show values between 5-10 meV, while spots G-J do not exhibit the $-2/3$ state at all in the MIM data. We also observe the $\nu=-4/3$ states in the MIM-Re data from spots A-C but cannot resolve the gap value from the chemical potential curve, which suggests the $-4/3$ gap is less than our measurement sensitivity of 2 meV. In general, we can conclude that the upper half of the device possesses a larger correlation strength than the lower half. We note that this difference also coincides with the difference in twist angles (0.1 - 0.6° in the upper half vs 1.1 - 1.2° in the lower half). We have also measured three spots in a second device D2 (See Section 12 of Supporting Information for data from D2). These spots have a similar twist angle of $\sim 0.6^\circ$. In general, these spots exhibit weaker features of correlated states: only $\nu=-1$ and -2 are well resolved while the $-4/3$ state is only visible in the MIM-Re channel. The weighted average gaps for $\nu=-1$ and -2 at these spots are in the ranges of 40-50 meV and 15-25 meV, respectively, which are comparable to the values for spots D-E in device D1 with twist angles around 0.6° (See Section 13 of Supporting Information for detailed analysis of the correlation between twist angle and energy gap sizes in both devices). The results from multiple spots of varying twist angles across two devices support that the correlation strength is weaker at larger twist angles (smaller moiré periodicity), although at this moment, we cannot rule out other factors such as differences in interface quality, local strain, etc, which could also play a role. Nonetheless, our results demonstrate the capability to resolve the local energy spectrum of the correlated states, representing an advantage over other global averaging measurements such as capacitance spectroscopy in the study of moiré physics in which local variations, especially in the twist angle, are common in the current generation of devices.

Finally, we would like to comment on the applicability of our methodology. The idea of using a sensing layer to probe the chemical potential of 2D devices has been demonstrated in earlier works^{24,25,29,33-40}, but an additional top gate is often needed to tune the sensing layer. Our technique utilizes a monolayer graphene as both the top gate and the sensing layer, thus only requiring a standard dual gate device structure, making it straightforward to study a wide range of 2D materials. To improve the measurement sensitivity, a thinner dielectric for the top gate sensing layer is preferred as it increases the geometric capacitance so that the effect of quantum capacitance is more pronounced.

In summary, our measurements provide quantitative results on the thermodynamic energy gaps of the correlated states in the archetypal WS_2/WSe_2 moiré superlattice, including both integer and fractional filling states. Spatial variations of the gap values reveal how the strength of electron correlation affects the different insulating states. Our methodology of using a specifically chosen top gate also as a sensing layer is fully compatible with fabrication of other van der Waals devices, and we believe this technique can be readily applied to other similar 2D systems.

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Supporting Information

Details about device fabrication, experimental setup of MIM, calculation of energy gaps, supplementary data, analysis on energy gap dependencies and discussion of negative compressibility.

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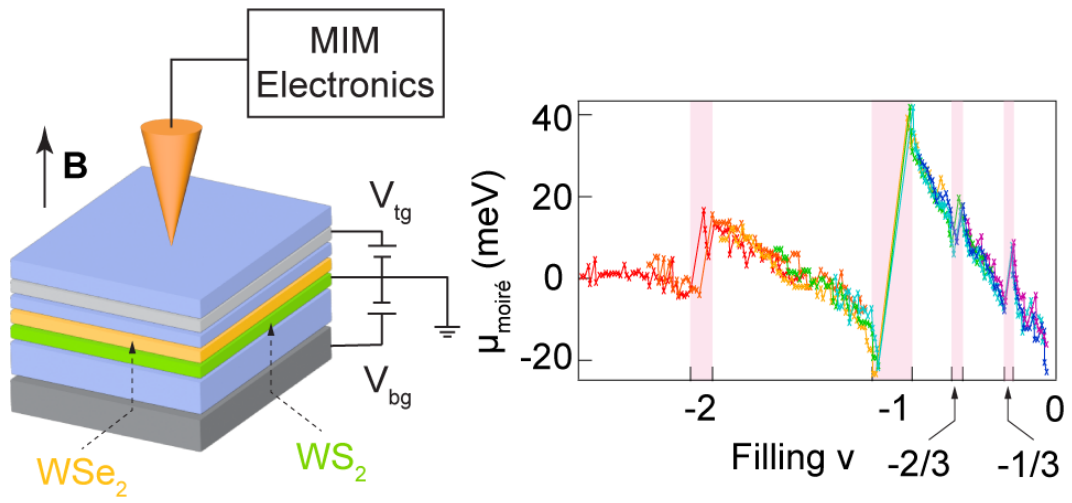
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