# Final Report: DE-SC0010308, University of Texas at Austin

**Project Title: Microscopy of Electrostatic Field Effect in Novel Quantum Materials**
**Principle Investigator: Keji Lai**
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## Project Accomplishments

The objective of this DOE Early Career project "Microscopy of Electrostatic Field Effect in Novel Quantum Materials" is to explore the nanoscale electronic properties in electrostatically modulated functional materials by microwave impedance microscopy (MIM). Field-effect transistors (FETs), where charges are electrostatically modulated at the surface of active materials, have been the backbone in modern semiconductor industry. The same FET structure has been widely used in the research of advanced quantum materials with remarkable success. Due to the difficulty to access the buried interface in FETs, however, little is known about the nanoscale uniformity in these structures, which may dominate the performance of devices. This DOE project is therefore of fundamental importance for the applications of complex quantum systems in basic energy science. In the following, we briefly summarize the scientific achievements under the five-year DOE support.

### 1. *Ferroelectric field effect on semiconductors*

The development of nonvolatile logic through direct coupling of spontaneous ferroelectric polarization with semiconductor charge carriers is nontrivial, with many issues ranging from the epitaxial ferroelectric growth, demonstration of ferroelectric switching, and measurable semiconductor modulation. Working with our collaborators, we demonstrated the c-axis oriented epitaxial $BaTiO_3$ (BTO) layer on Ge and the modulation of carrier densities at the ferroelectric-Ge interface. As seen in Fig. 1, both piezo-force microscopy (PFM) and MIM data show clear contrast between the region written by $V_{tip} = -4V$ and the as-grown area. The PFM contrast confirms that BTO polarization is switched by a large negative tip bias. More interestingly, the MIM contrast originates solely from the underlying Ge layer accompanied by the poling process. To our knowledge, it is the first time ferroelectric field effect is reported on semiconductors, which is not possible without the unique capability of the MIM. This work is published in *Nature Communications*[1].

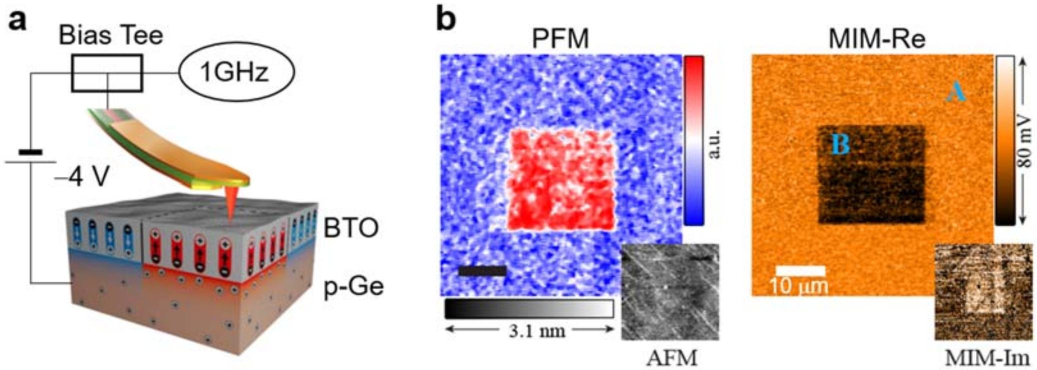


Figure 1. (a) Schematic diagram illustrating the measurement setup and configuration of samples. A negatively biased conductive scanning probe makes downward to upward switching in ferroelectric polarization of BTO layer, which modulates the hole density at the BTO/p-Ge interface. (b) PFM/AFM and (c) MIM images acquired after writing a 20 μm × 20 μm square by applying -4 V DC bias on the tip. The scale bars are 10 μm.

2. *Ion-gel gated electric double layer transistors*

Using ionic liquids or gels as the gate dielectrics, the electric double-layer transistors (EDLTs) developed in the past decade have demonstrated the ability to modulate the sheet carrier density at a level much higher than that in conventional MOSFETs. In most EDLT devices, however, the electrolyte-semiconductor interface is usually buried underneath a large droplet of ionic liquid and thus cannot be directly studied by surface-sensitive probes. As a result, a major challenge in EDLT research is to study the microscopic conductance fluctuations in the device. In this work, we first spin coat a 50nm-thick layer of ionic gel on the device that maintains the gate functionality, as schematically shown in Fig. 2a. The gel freezes below its glass transition temperature of ~ 200 K so that one can perform contact-mode imaging at low temperatures. Note that the MIM response is most sensitive to electrical inhomogeneity around the crossover region (Fig. 2b). In Fig. 2c, when the ZnO channel is either insulating ($G_{DS}$ = 0 μS) or conducting ($G_{DS}$ = 6.7 μS), only surface particles with lower signals than the background are seen. For an intermediate $G_{DS}$ = 1.3 μS, appreciable mesoscopic conductance fluctuation could be observed in the image. This is the first time electrical inhomogeneity is resolved in the EDLT configurations, which is particularly useful for the study of strongly correlated systems with nanoscale phase separation. This work is published in *Nano Letters*[2].

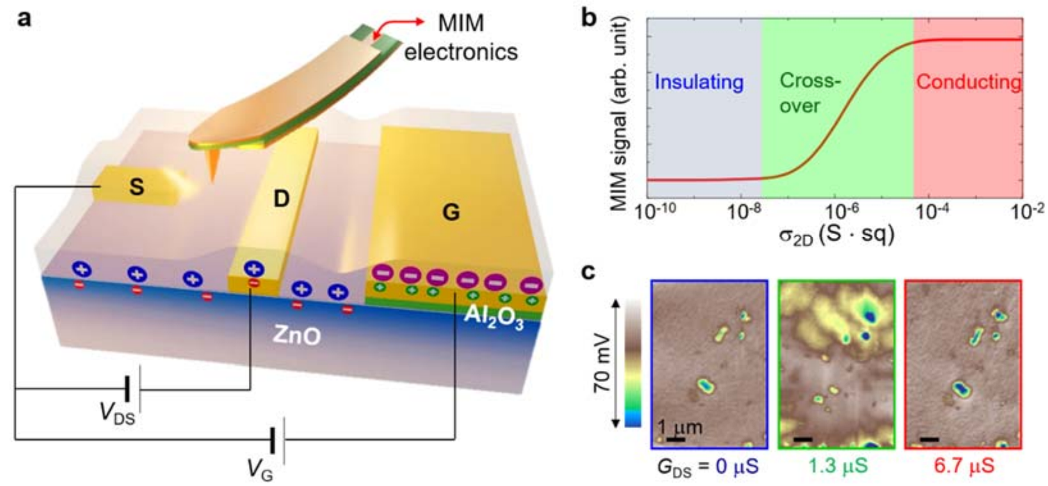


Figure 2. (a) Schematic diagram of the EDLT device and the MIM setup. (b) MIM response as a function of the 2D sheet conductance. (c) MIM images at three different $G_{DS}$'s after removing the background signals, showing the mesoscopic electrical inhomogeneity. All scale bars are 1 μm.

### 3. *Edge states and inhomogeneity in 2D materials*

The understanding of various types of disorders in atomically thin transition metal dichalcogenides (TMDs) is of fundamental importance for their applications in electronics and photonics. Because of the imperfections, electrons moving on these two-dimensional (2D) crystals experience a spatially non-uniform Coulomb environment, whose effect on the charge transport has not been microscopically studied. For layered van der Waals (vdW) materials, another unique feature takes place at the sample edges, where the broken crystalline symmetry and the presence of dangling bonds introduce additional electronic states to the bulk band structure. Spatially resolved conductance maps are therefore highly desired for the understanding of electrical inhomogeneity and edge channels in TMD FETs. A major effort of this DOE program is to investigate the conductance fluctuations in the mesoscopic length scale.

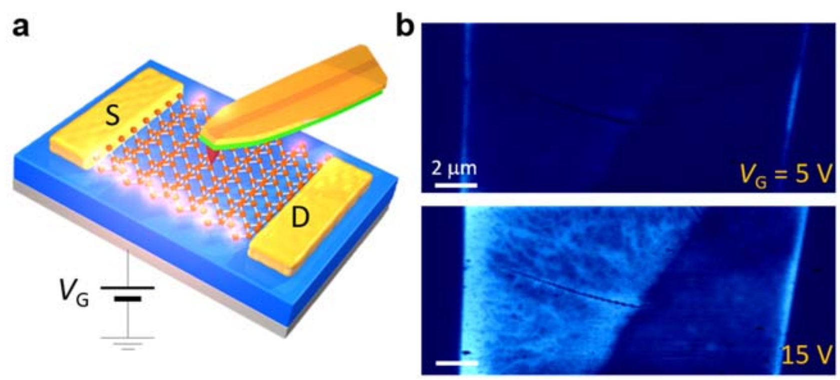


Figure 3. (Left) Schematic of the MIM setup and the back□gated $MoS_2$ FET. (Top right) MIM image showing the conductive channels at the two edges of the sample at a low gate bias $V_G$ = 5 V. (Bottom right) Thread-like defects when the left part of the device was turned on at $V_G$ = 15 V.

Figure 3 shows the schematic and experimental data on a multi-layer $MoS_2$ FET. Here a shielded cantilever probe carrying the 1 GHz excitation is scanning on the device with source-drain contacts and $SiO_2$/Si back gate. The sample is an exfoliated $MoS_2$ flake with two distinct regions, 4 layers on the left and 3 layers on the right. A number of interesting features are seen in the MIM images. First, as the back gate voltage $V_G$ increases to 5 V, the conductive states emerge at the sample edges before the bulk of the sample is populated by electrons. Secondly, at $V_G = 15$ V, the 2D sheet conductance of the 4-layer region is higher than that of the 3-layer counterpart, indicative of the higher mobility and lower band gap of the thicker flakes. Finally, strong conductance inhomogeneity, which is associated with the fluctuations of disorder potential in the material, is also observed in the image, providing a guideline for future improvement of the device performance. This work is published in *PNAS* [3] and selected as a DOE Research Highlight (https://science.energy.gov/bes/highlights/2017/bes-2017-02-c/).

4. *Sketched transistors at oxide interface*

Nanoscale control of the 2D electron gas at the $LaAlO_3$/$SrTiO_3$ (LAO/STO) interface by a conductive probe tip has triggered the development of a number of electronic devices. While the spatial distribution of the conductance is crucial for such devices, it is challenging to visualize the local electrical properties at the buried interface. Taking advantage of the capacitive coupling in MIM imaging, we demonstrate the writing and imaging of sketched nanostructures at the LAO/STO interface (Fig. 4a). Fig. 4b shows the multiple bands written by the same MIM tip with +5 V bias, which are not present in the topographic image. Quantitative analysis indicates that the MIM signals are in good agreement with the transport data. Our results pave the way to study emergent phenomena at oxide interfaces by probing nanoscale conductance distribution. This work is published in *Applied Physics Letters*[4].

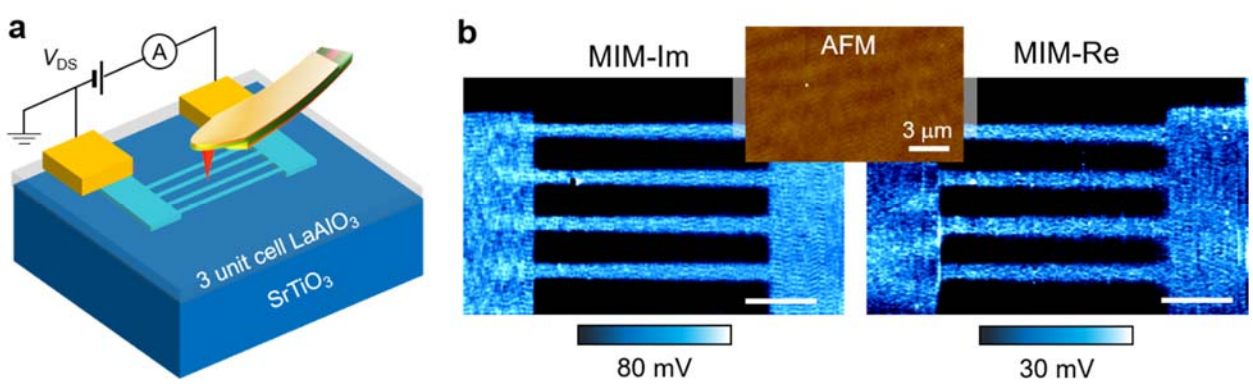

Figure 4. (a) Schematic of the experimental setup. Both the microwave excitation and the tip bias are applied to the tip through a bias-tee. The two-terminal conductance is monitored by a source-drain bias. (b) AFM and MIM images of four bands written by $V_{tip}$ = 5 V across two pads. The conductive features are clearly seen in the microwave image but absent in surface topography. All scale bars are 3 μm.

## 5. *Tuning-fork MIM on 2D heterostructures*

The vertical stacking of vdW materials introduces a new degree of freedom to the research of 2D systems. The interlayer coupling strongly influences the band structure of the heterostructures, resulting in novel properties that can be utilized for electronic and optoelectronic applications. In this work, we report the electrical mapping of a gated $MoS_2/WSe_2$ heterostructure transistor. To minimize perturbation to the sample, we implement a tapping-mode MIM based on the tuning-fork (TF) feedback, as shown in Fig. 5a. The MIM signal, modulated by the TF resonant frequency (~32 kHz), is demodulated by a lock-in amplifier to form the AC_MIM (published in *Review of Scientific Instruments*[5]). Fig. 5b shows that, on individual flakes, the MIM signals gradually increase (decrease) with increasing gate voltage on the $MoS_2$ ($WSe_2$) part of the device. Interestingly, in the region with significant source-drain current, electrons in the n-type $MoS_2$ and holes in the p-type $WSe_2$ segments are nearly balanced, whereas the heterostructure (HS) area is depleted of mobile charges. The result can be satisfactorily explained by the lateral band alignment. Since nanoscale conductivity is an essential property for electronic applications, our spatial mapping of the local conductance is of great importance for the rapid progress of vdW heterostructure research. This work is published in *Nano Letters*[6].

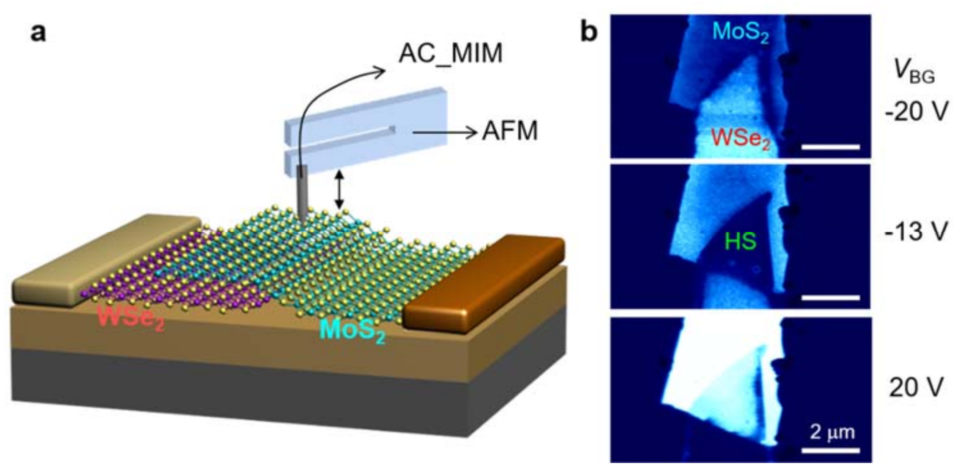


Figure 5. (a) Schematic of the experimental setup, showing the TF-based MIM and the heterostructure transistor. (b) AC_MIM-Im images at three back-gate voltages. Signals in the n-type $MoS_2$ (p-type $WSe_2$) region increase (decrease) with increasing gate bias, whereas the HS section is depleted of mobile charges in the intermediate $V_{BG}$. All scale bars are 2 μm.

In summary, using the microwave impedance microscopy, the PI's team has studied the spatial evolution of local conductivity in various FETs ranging from traditional metal-oxide-semiconductor FETs, oxide-interface sketch FETs, to ferroelectric FETs and electric double-layer transistors during this DOE project. The results are significant for understanding the nanoscale uniformity in technologically important materials towards energy-related applications.

**List of Papers**


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2. Yuan Ren, Hongtao Yuan, Xiaoyu Wu, Zhuoyu Chen, Yoshihiro Iwasa, Yi Cui, Harold Y. Hwang, and **Keji Lai**, "Direct Imaging of Nanoscale Conductance Evolution in Ion-Gel-Gated Oxide Transistors", *Nano Letters* **15**, 4730 (2015).
3. Di Wu, Xiao Li, Lan Luan, Xiaoyu Wu, Wei Li, Maruthi N Yogeesh, Rudresh Ghosh, Zhaodong Chu, Deji Akinwande, Qian Niu, **Keji Lai**, "Uncovering edge states and electrical inhomogeneity in $MoS_2$ field-effect transistors", *Proceedings of the National Academy of Sciences* **113**, 8583 (2016).
4. Zhanzhi Jiang, Xiaoyu Wu, Hyungwoo Lee, Jung-Woo Lee, Jianan Li, Guanglei Cheng, Chang-Beom Eom, Jeremy Levy, and **Keji Lai**, "Direct imaging of sketched conductive nanostructures at the $LaAlO_3$/$SrTiO_3$ interface", *Applied Physics Letters* **111**, 233104 (2017).
5. Xiaoyu Wu, Zhenqi Hao, Di Wu, Lu Zheng, Zhanzhi Jiang, Vishal Ganesan, Yayu Wang, and **Keji Lai**, "Quantitative measurements of nanoscale permittivity and conductivity using tuning-fork-based microwave impedance microscopy", *Review of Scientific Instruments* **89**, 043704 (2018).
6. Di Wu, Wei Li, Amritesh Rai, Xiaoyu Wu, Hema C. P. Movva, Maruthi N. Yogeesh, Zhaodong Chu, Sanjay K. Banerjee, Deji Akinwande, and **Keji Lai**, "Visualization of Local Conductance in $MoS_2$/$WSe_2$ Heterostructure Transistors", *Nano Letters* **19**, 1976 (2019).



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