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What if we could electrically tune properties of strongly-correlated materials just like we can with semiconductors

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Our research funded by AFOSR under FA9550-16-1-0126 focused on fabrication of gate voltage-tunable superconducting weak links, using ionic liquid gating to modulate the very high carrier densities typically found in superconductors, too high to control with conventional gate electrodes. Three major thrusts were: 1) controlling superconductivity and tuning Josephson Junctions in diverse materials including high-Tc cuprates, and 2) fabrication of gate-tunable nanoscale ballistic junctions in SrTiO<sub>3</sub>, a superconductor that has much lower T<sub>c</sub> (<1K) but is highly responsive to electrostatic and ionic liquid gating, and can have large electronic mean free path.  
1) Electrolyte gating of diverse superconductors and junctions: In collaboration with Shane Cybart (at UC Riverside), we demonstrated gate tunable high-Tc Josephson junctions. Superconducting weak links in optimally doped YBCO were written with He<sup>+</sup> beam exposure. They showed high I<sub>c</sub>R<sub>n</sub> products up to 2.5 mV, indicative of high-quality barriers. The junction critical current was modulated in a reversible manner by the gate voltage applied across an ionic liquid, as shown in Fig. 1(a,b) [Stanwyck 2017].

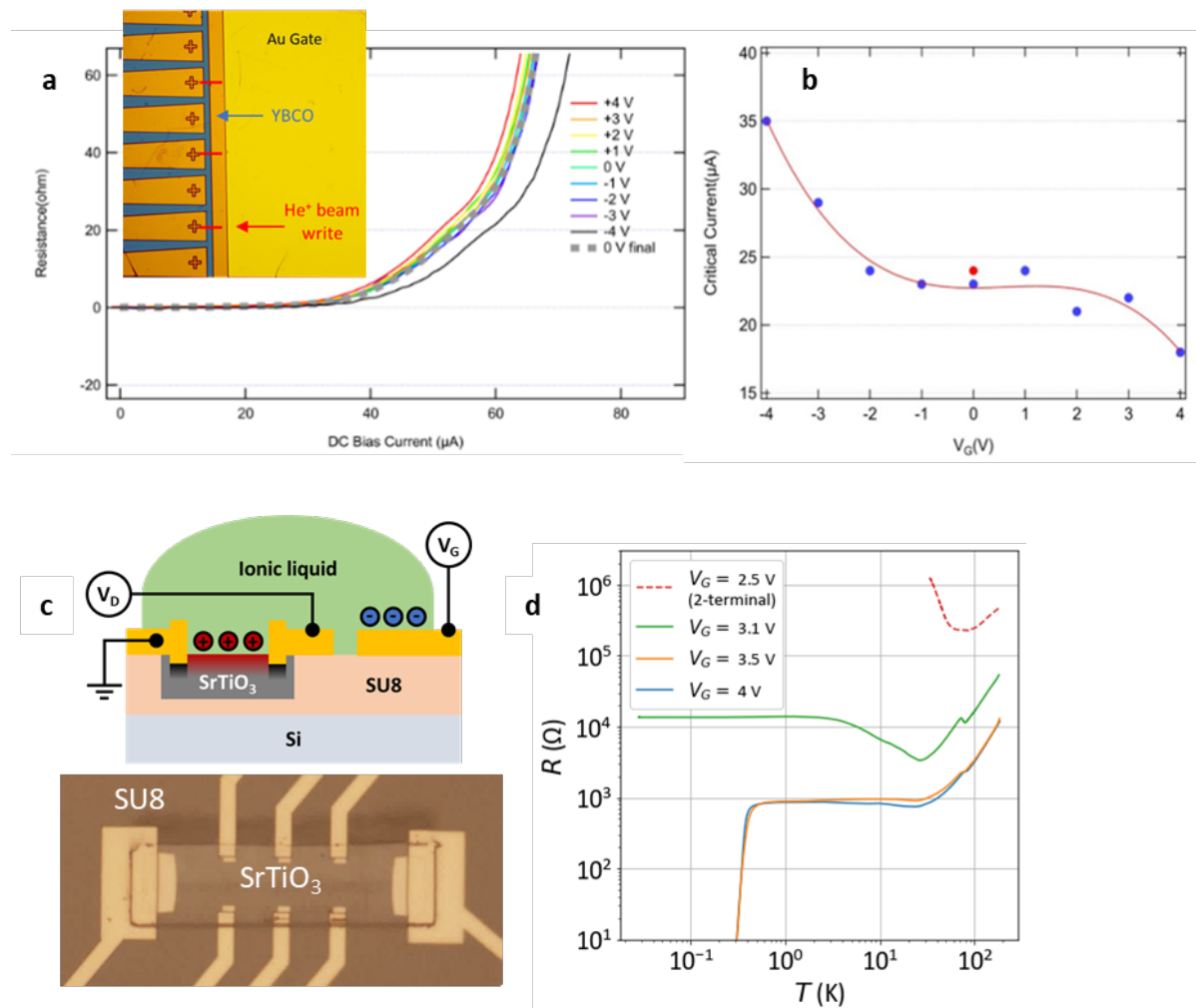
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Our research funded by AFOSR under FA9550-16-1-0126 focused on fabrication of gate voltage-tunable superconducting weak links, using ionic liquid gating to modulate the very high carrier densities typically found in superconductors, too high to control with conventional gate electrodes. Three major thrusts were: 1) controlling superconductivity and tuning Josephson Junctions in diverse materials including high- $T_c$  cuprates, and 2) fabrication of gate-tunable nanoscale ballistic junctions in  $\text{SrTiO}_3$ , a superconductor that has much lower  $T_c$  (<1K) but is highly responsive to electrostatic and ionic liquid gating, and can have large electronic mean free path.

1) Electrolyte gating of diverse superconductors and junctions: In collaboration with Shane Cybart (at UC Riverside), we demonstrated gate tunable high- $T_c$  Josephson junctions. Superconducting weak links in optimally doped YBCO were written with  $\text{He}^+$  beam exposure. They showed high  $I_c R_n$  products up to 2.5 mV, indicative of high-quality barriers. The junction critical current was modulated in a reversible manner by the gate voltage applied across an ionic liquid, as shown in Fig. 1(a,b) [Stanwyck 2017].

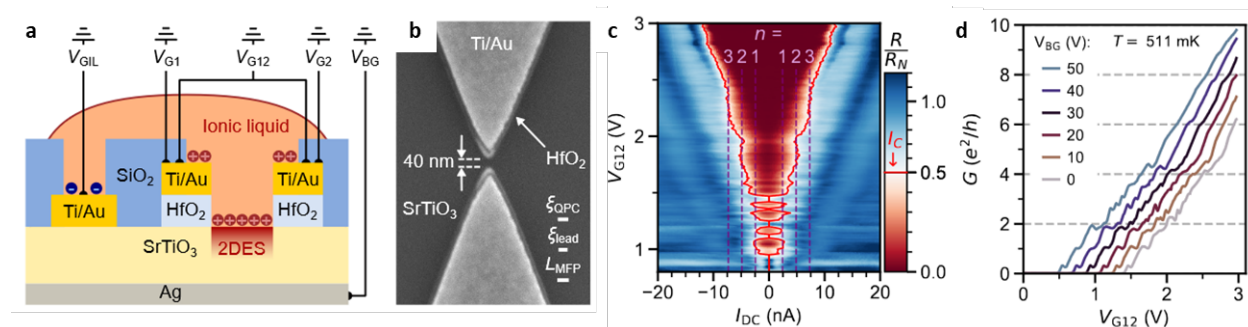


**Fig 1:** (a,b) high  $T_c$  Josephson junction direct written by  $\text{He}^+$  beam with a critical current at 1.6 K tunable by ionic liquid gate voltage (gate voltage must be adjusted at 250 K). (c) Lamella device defined by focused ion beam (dimensions: 54  $\mu\text{m}$  length, 15  $\mu\text{m}$  width, 3  $\mu\text{m}$  thickness), (d) ionic liquid gate control of the insulator-normal metal-superconductor transition.

In developing gate-tunable superconducting devices, a key bottleneck is the difficulty of obtaining new and exploratory materials in the form of high-quality thin films with smooth surfaces, or very large single crystals. In collaboration with Philip Moll (at EPFL), we developed a new fabrication route that removes some of these restrictions and can be implemented even on small sized crystals of exotic quantum materials. A manuscript is currently in preparation with a focus on the process itself. We demonstrated that a chosen pristine surface can be preserved during focused ion beam machining of a micron-sized lamella. Such a lamella can then be transferred onto the surface of a crosslinked SU8 resist for further fabrication of a superconducting device that is tunable with ionic liquid gating (Fig. 1(c,d)). Our test case was SrTiO<sub>3</sub> since we had established recipes for electrolyte gating it, but we expect this approach will be applicable to many materials.

2) Gated ballistic superconducting devices: Under funding by AFOSR, our group demonstrated that ionic liquid gated SrTiO<sub>3</sub> is one of the most promising platforms for fabrication of electrically-tunable ballistic quantum superconducting devices, joining a select group of such platforms: twisted bilayer or trilayer graphene, proximitized InAs, InSb, or HgTe, and other SrTiO<sub>3</sub>-based systems.

In [Mikheev2020], we demonstrated nanopatterned split-gate devices that electrostatically define a quantum constriction between two large regions of superconducting SrTiO<sub>3</sub>. Near pinch-off by local dielectric gate voltage, the constriction behaves as a superconducting quantum point contact (SQPC): the normal state conductance ( $G_N$ ) and the critical supercurrent ( $I_c$ ) become discretized as the number of ballistic modes through the constriction is tuned by gate voltage (Fig. 2). The quanta of conductance and supercurrent have ideal, geometry-independent values:  $\delta G_N = 2e^2/h$  and  $\delta I_c = e\Delta/\hbar$  ( $\Delta$  is the superconducting gap).  $\delta I_c$  is however particularly sensitive to device non-idealities, such as finite junction length and imperfect normal-superconductor interface transparency. Our demonstrated  $\delta I_c$  was a quarter of  $e\Delta/\hbar$  (among the highest fractions reported to date) and overall we achieved arguably the best realization to date of an SQPC in terms of combining quantization definition and step size.



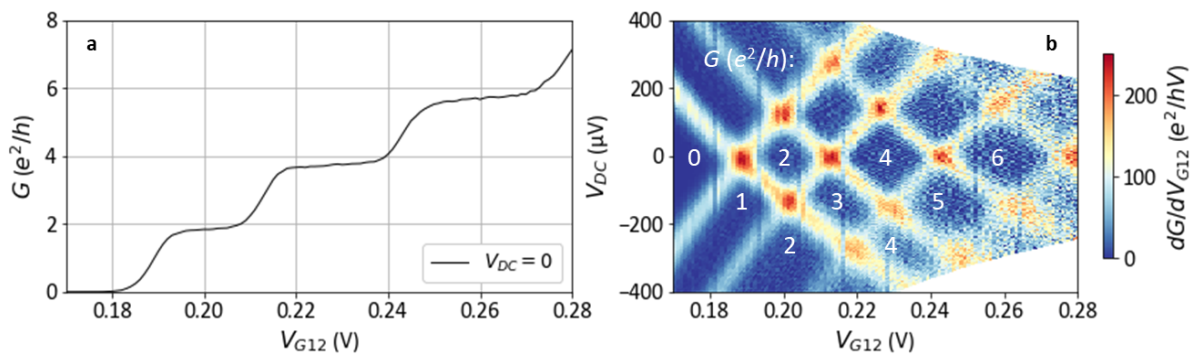
**Fig 2:** (a,b) Superconducting quantum point contact device in SrTiO<sub>3</sub>. (c) Quantized critical current at 30 mK. (d) Quasi-ballistic normal state conductance quantization above superconducting T<sub>c</sub>.

This remarkable demonstration experiment was enabled by intense technique development but had one substantial limitation: the relatively short 55 nm mean free path, on the order of junction size and in the typical range for SrTiO<sub>3</sub>-based 2D electron systems (2DESs), such as that formed at the LaAlO<sub>3</sub>/SrTiO<sub>3</sub> Interface. To try to improve device cleanliness, we started a collaboration with Bharat Jalan (at University of Minnesota), who grew homoepitaxial SrTiO<sub>3</sub> films by molecular beam epitaxy, with increased purity and quality in comparison to commercial single crystal SrTiO<sub>3</sub> substrates. After overcoming several hurdles in fabrication of such films into ionic liquid gated devices (tendency for

epitaxially strained SrTiO<sub>3</sub> to be stubbornly insulating, and formation of parasitic conductance paths), we were able to demonstrate a metallic 2DES for the first time in such films. But so far, the observed electron mobility is surprisingly low (mean free path below 25 nm). We attribute to increased confinement of 2DES near the surface in epitaxial films, a problem that can be addressed going forward with band engineering in the MBE growth process.

A parallel approach to enhancing mean free path is adding a barrier layer between the ionic liquid and the SrTiO<sub>3</sub> channel. Around the start of this grant, this approach was developed in our group using hexagonal boron nitride as a barrier. This increased the electron mobility of ionic liquid gated SrTiO<sub>3</sub> by an order of magnitude [Gallagher2015], and provided similar benefits for ionic liquid gated graphene [Petach2017]. Early in the program this effort also led to serendipitous discoveries involving hBN [Lee 2016, Yamoah 2017, Gallagher 2018]. However, exfoliated flakes of few layer hBN are very difficult to work with and are also insufficiently robust to be compatible with fabrication of devices with local dielectric gates.

Under this AFOSR program, we collaborated with Eric Pop to grow and characterize high quality large-area monolayer hBN [Chen2020], but still found it challenging to transfer the hBN from the growth substrate while retaining desirable barrier qualities. We then developed HfO<sub>2</sub> deposited by atomic layer deposition into an alternative barrier layer that like hBN increased the mean free path by an order of magnitude into the 1-2 micron range. Unlike exfoliated hBN, HfO<sub>2</sub> is sufficiently robust and reproducible for fabrication of devices with nanopatterned gates. This enabled the recent demonstration of clean ballistic quantum point contact (QPC) behavior in SrTiO<sub>3</sub>/HfO<sub>2</sub>/ionic liquid structures. Fig. 3 shows successive opening of several subbands, as well as a classic “diamond” pattern in DC bias spectroscopy of such a device. The crispness and definition of higher lying quantum subbands is unprecedented for QPCs fabricated from oxide or other strongly-correlated materials and is only bested by the gold standard of GaAs-based high mobility heterostructures. This demonstration is very exciting in view of building SrTiO<sub>3</sub> into a new platform for studying mesoscopic and topological superconductivity. It occurred after the conclusion of this funding program, but AFOSR will still get appropriate credit.



**Fig 3:** Ballistic conductance quantization in a QPC from high mobility SrTiO<sub>3</sub>/HfO<sub>2</sub>/ionic liquid 2DES. (a) zero bias conductance in a 5 Tesla field. (b) DC bias spectroscopy of quantum subbands, shown as transconductance with split gate voltage.

**References (with one exception noted below, this is also the list of papers published and submitted):**

- [Chen 2020] Victoria Chen, Yong Cheol Shin, Evgeny Mikheev, Joel Martis, Ze Zhang, Sukti Chatterjee, Arun Majumdar, David Goldhaber-Gordon, Eric Pop, Application-Driven Synthesis and Characterization of Hexagonal Boron Nitride on Metal and Carbon Nanotube Substrates, arxiv:2011.14286 (2020).
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- [Gallagher2018] Patrick Gallagher, Yilei Li, Kenji Watanabe, Takashi Taniguchi, Tony F. Heinz, and David Goldhaber-Gordon, Optical Imaging and Spectroscopic Characterization of Self-Assembled Environmental Adsorbates on Graphene, *Nano Lett.* 18, 2603–2608 (2018).
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- [Mikheev 2020] E. Mikheev, I.T. Rosen, and D. Goldhaber-Gordon, Quantized critical supercurrent in SrTiO<sub>3</sub>-based quantum point contacts, arXiv:2010.00183 (2020).
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- [Stanwyck 2017] S. Stanwyck, Electrolyte gating of nanoscale complex oxide devices, PhD thesis, Stanford University (2017).
- [Yamoah 2017] Megan A. Yamoah, Wenmin Yang, Eric Pop, and David Goldhaber-Gordon, High-Velocity Saturation in Graphene Encapsulated by Hexagonal Boron Nitride, *ACS Nano* 11, 9914–9919 (2017).

**Students/postdocs supported**

- Evgeny Mikheev (Postdoc, still at Stanford. Received Nano and Quantum Science and Engineering Postdoctoral Fellowship.)
- Menyong Lee (Ph.D. 2016, Kavli postdoctoral fellow at Cornell, now Research Assistant Professor at Cornell). Support for research expenses, not salary.
- Patrick Gallagher (Ph.D. 2016, Heising-Simons postdoctoral fellow at Berkeley, then founded QIS startup). Support for research expenses, not salary.
- Sam Stanwyck (Ph.D. 2017, Senior Quantum Engineering Manager at Rigetti Computing, now Quantum R&D Physicist, Keysight)
- Trevor Petach (Ph.D. 2017, received Melvin P. Klein Scientific Development Award; Scientist, Exponent, now Data Science Consultant, Data CRT)
- Xiao Zhang (rotated in Goldhaber-Gordon group, now continuing with Ph.D.)
- Linsey Rodenbach (Ph.D. expected 2023)
- R. Joseph Finney (Ph.D. expected 2023)
- Megan Yamoah (High school student, graduated 2016, attended MIT, Goldwater Scholar, now Rhodes Scholar)
- Yu-Tse Lee (Undergrad, writing thesis that will acknowledge grant)

### **Awards**

Awards to supported students and postdocs are noted above.

Goldhaber-Gordon was recognized as a Fellow of the American Physical Society in 2018.

### **Inventions**

A patent disclosure was filed for a multichannel, feedback-enabled electronic measurement setup, which dramatically speeds measurements. We hope to partner with an established company to market this.

### **Significant collaborations**

These were mentioned in the project narrative, but to make sure they come through clearly:

Shane Cybart, UC Riverside (for electrical control of high-Tc Josephson Junctions)

Eric Pop, Stanford (for growth, characterization, and application of hexagonal boron nitride)

Philip Moll, MPI Dresden and EPFL Lausanne (FIB-based fabrication, preserving pristine surfaces)

Bharat Jalan, University of Minnesota (Gating of MBE-grown oxide films)