2023-2024 Grand Challenge Award Final Report

Awardee: **Feliciano Giustino, Professor, Physics**

Research Award Title: **Quantum design of materials for energy-efficient information and communication technology**

Research Summary

If current trends continue, information and communication technology (ICT) could account for 20% of global greenhouse gas emissions by 2040. This increase is driven by the rapid growth in data science, cloud computing, and artificial intelligence. For example, training the GPT-3 language model consumed about one gigawatt-hour of energy, resulting in over 500 tons of carbon dioxide emissions. To align with climate goals, a significant reduction in ICT power consumption is necessary.

Transistors are fundamental to ICT. Over recent decades, their integration density has increased dramatically, leading to the development of chips with trillions of transistors. For instance, Micron's V-NAND chip contains five trillion MOSFETs. To achieve such density while avoiding performance issues, transistors use the FinFET architecture, where the channel is an ultra-thin fin surrounded by the gate electrode. Upcoming technologies, such as Intel's RibbonFET and TSMC's GAAFET, aim to further reduce channel thickness. However, below 3 nm, the mobility of silicon channels—which quantifies the velocity at which electrons travel in the channel under an applied voltage, and is essential for high-speed operation—decreases sharply due to interface scattering and quantum effects, impacting performance and thermal management.

Two-dimensional (2D) materials, which are only a few atoms thick, offer a potential solution for further transistor miniaturization. Their atomically sharp surfaces and scalability make them especially attractive. However, most 2D materials have relatively low carrier mobility (10-100 cm^2/Vs at room temperature), which is insufficient to outperform current silicon technologies. To be viable, 2D materials need to achieve a tenfold increase in mobility, around 1000 cm^2/Vs.

Within this project we conducted quantum simulations to predict the electric transport properties of many candidate 2D materials. Quantum simulations allow us to design and test materials atom-by-atom, predicting their properties with high precision before physical fabrication. We used the *ab initio* Boltzmann transport equation (aiBTE), a quantum adaptation of the classical Boltzmann equation, to compute carrier mobility. This method requires high-performance computing resources, such as those provided by the Texas Advanced Computing Center (TACC), and is implemented in the EPW code, a leading software for calculating carrier mobilities from first principles which is developed by us and our collaborators (https://epw-code.org). Recent improvements to EPW enabled us to perform the most sophisticated and accurate mobility calculations ever reported.

We screened the 2D Materials Cloud database, which includes over 5,000 layered compounds to identify promising candidates. Through a rigorous hierarchical downselection process (Figure 1), we found several materials worthy of further analysis with the aiBTE. This screening required a total of 18 million hours on the Frontera supercomputer at TACC, the world's largest academic supercomputer.

Our computational screening identified several promising high-mobility 2D materials. Among them, monolayer tungsten disulfide (WS_2)—a semiconductor consisting of one atomic layer of molybdenum sandwiched between two atomic layers of sulfur—emerged as particularly exceptional. In our initial screening, WS_2 was the only material with a predicted hole mobility exceeding 1,300 cm^2/Vs at room temperature (Figure 1). Despite this, experimental measurements show a mobility of less than 100 cm^2/Vs, raising the question on why our calculations—which are very close to experiments for most materials—were so far from the measured values.

To answer this question, we conducted an in-depth analysis of carrier scattering in WS_2. Our findings indicated that the high theoretical mobility is due to strong spin-orbit coupling and weak ionic polarization. Spin-orbit coupling splits the doubly-degenerate bands at the K valleys, preventing intravalley interband scattering and lowering the Γ valley to restrict K-Γ scattering. Additionally, spin-orbit coupling leads to valley polarization, which blocks K-K' intervalley scattering. This behavior is unique among 2D materials; a similar effect occurs in the related compound WSe_2 but is less pronounced due to weaker spin-orbit coupling. Moreover, WS_2 exhibits the weakest ionic dielectric screening among polar compounds, resulting in minimal Fröhlich scattering and exceptionally long hole relaxation times.

These insights explained WS_2's high theoretical mobility, but did not resolve the discrepancy with experimental values. To further investigate, we included additional mechanisms not typically accounted for in the aiBTE due to their computational complexity. We incorporated quantum many-body quasiparticle corrections, examined carrier concentration effects, and accounted for quadrupole-type electron-phonon scattering and point-defect and grain-boundary scattering. These effects decreased the estimated mobility to approximately 200 cm^2/Vs, aligning more closely with experimental results. Therefore, we concluded that WS_2 is intrinsically a ultra-high mobility semiconductor, but the presence of defects and interfaces prevent it from reaching its fullest potential.

Our work suggests that, in the quest for high-performance 2D materials for ICT, instead of searching for exotic new materials, we should rather focus on improving fabrication processes and interface engineering of a well-known compound. We are currently collaborating with an experimental group to verify our predictions and realize high-mobility 2D transistors baded on WS_2.

A manuscript resulting from this project is in press at npj Computational Materials. Calculations were performed by Dr. Viet-Anh Ha, a research associate in my team who specializes in *ab initio* calculations of carrier transport. In addition to the Moncrief Grand Challenge Award which supported me during this period, this work was sponsored by the Seminconductor Research Corporation via the JUMP 2.0 Center SUPREME.

Beyond the above research on the design of high-performance 2D materials for next-generation ICT, this Moncrief Grand Challenge award allowed me to fully engage with several ongoing projects in my group. This activity has led to a number of publications, most notably in high-profile journals such as Physical Review Letters, Proceedings of the National Academy of Sciences, and Nature.

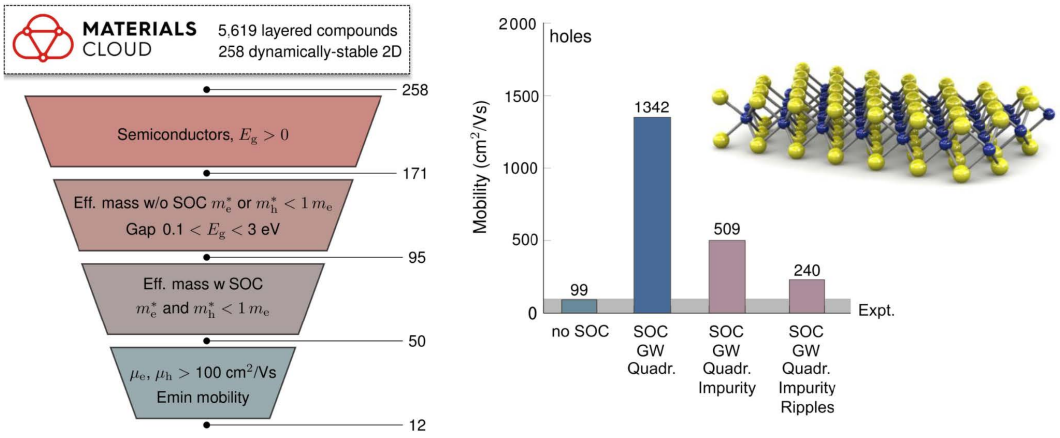

Figure 1: Left: High-throughput screening of carrier mobilities of 2D materials from the 2D Materials Cloud Database. Right: Computed hole mobility of monolayer WS_2, including various scattering mechanisms, and ball-stick model of WS_2.

Presentations

1. Invited Talk, "Polarons and self-trapped excitons in halide perovskites." European Materials Research Society (E-MRS) Fall Meeting, Warsaw, Poland, September 2023.

2. Invited Talk, "Polarons in 2D materials using the EPW code." Workshop "Quantum Espresso : Applications for Material Sciences and Chemistry," University of Evora, Portugal, November 2023.

3. Departmental Colloquium, "First-principles calculations of carrier transport in semiconductors." Materials Science and Engineering Department, University of Texas at Dallas, January 2024.

4. Invited Talk, "Ab initio self-trapped excitons without supercells." Frontiers in Condensed Matter and Materials Physics: A Scientific Symposium in Honor of the 75th Birthday of Steven G. Louie, University of California at Berkeley, March 2024.

5. "Ab initio calculations of carrier transport in semiconductors," American Physical Society March Meeting, Minneapolis, USA, March 2024.

6. Invited Talk, "One hundred years of polarons." Centre for the Physics of Materials, McGill University, Montréal, Québec, March 2024.

7. Invited Talk, "Polaron surprises & EPWpy abstractions," CECAM Psi-k Research Conference "Electronic-Structure Simulations for Large-Scale Facilities: Opportunities, Challenges, and Roadmaps," Ecole Polytechnique Fédérale de Lausanne, May 2024.

8. Invited Talk, "Polarons & Exciton Polarons." CECAM Flagship Workshop "Frontiers in many-body excited-state dynamics from first principles," Ecole Polytechnique Fédérale de Lausanne, July 2024.

9. "Quantum design of next-generation electronic and energy materials," 2024 MATS Symposium and Workshop: "Innovations for a Changing Environment," University of California at San Diego, July 2024.

Publications

1. Y. Li, F. Zhang, V.-A. Ha, C. Dong, Y.-C. Lin, Q. Gao, B. Kousa, H. Kim, E. Khalaf, J. A. Robinson, F. Giustino, C.-K. Shih "Tuning commensurability in twisted van der Waals bilayers," *Nature* 625, 494 (2024).

2. Z. Dai, C. Lian, J. Lafuente-Bartolomé, and F. Giustino, "Excitonic polarons and self-trapped excitons from first-principles exciton-phonon couplings," *Phys. Rev. Lett.* 132, 036902 (2024).

3. Z. Dai, C. Lian, J. Lafuente-Bartolomé, and F. Giustino, "Theory of excitonic polarons: From models to first-principles calculations," *Phys. Rev. B* 109, 045202 (2024).

4. A. Wang, K. Bushick, N. Pant, W. Lee, X. Zhang, J. Leveillee, F. Giustino, S. Poncé, E. Kioupakis, "Electron mobility of SnO2 from first principles," *Appl. Phys. Lett.* 124, 172103 (2024).

5. S. Tiwari, E. Kioupakis, J. Menendez, and F. Giustino, "Unified theory of optical absorption and luminescence including both direct and phonon-assisted processes," *Phys. Rev. B* 109, 195127 (2024).

6. J. Lafuente-Bartolomé, C. Lian, and F. Giustino, "Topological polarons in halide perosvkites," *Proc. Natl. Acad. Sci. U.S.A.* 121, e2318151121 (2024).

7. B. Cucco, J. Leveillee, V.-A. Ha, J. Even, M. Kepenekian, F. Giustino, and G. Volonakis, "Intrinsic limits of charge carrier mobilities in layered halide perovskites," *PRX Energy* 3, 023012 (2024).

8. V.-A. Ha and F. Giustino, "High-throughput screening of 2D materials identifies p-type monolayer WS_2 as potential ultra-high mobility semiconductor," *npj Comput. Mater.* (2024), in press.

Awards and Recognitions

1. 2023 Clarivate Highly Cited Researcher