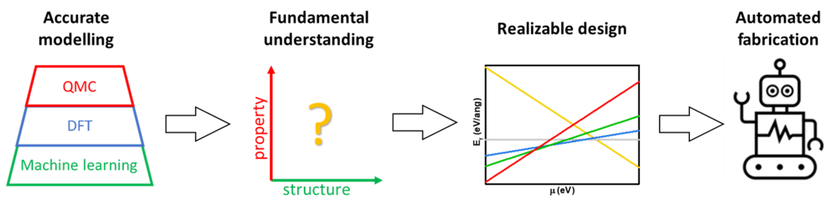
New Energy Solutions with Computational Materials Chemistry

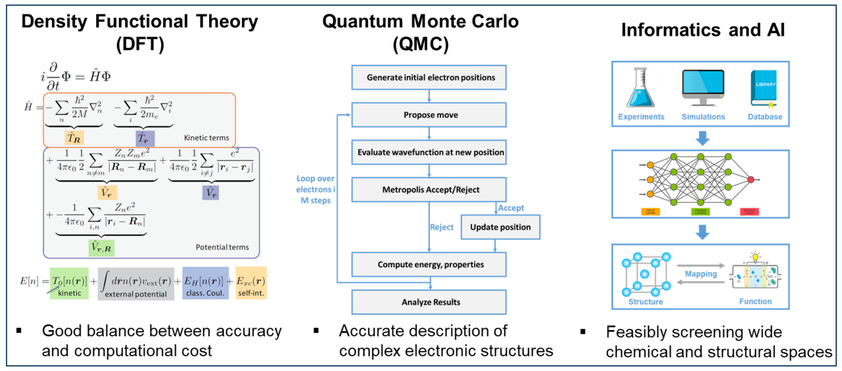
Seungjun Cha, Zhendian Zhang, Beenish Bahir, Courtney Brea, Rahul Somni, Ankit Bansal, Prajeet Oza, Pranav Khadilkar, Heejoon Jeon, Guoxiang (Emma) Hu*

School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA.

Synopsis: Efficient and sustainable energy harvesting and utilization is one of the prime scientific and engineering challenges of today. A key objective lies in finding high-performance and low-cost materials for these applications. However, better methods are needed for targeted materials design and precise fabrication than the current Edisonian approaches. Our group at Georgia Tech seeks to understand the mechanisms of energy harvesting, storage, and conversion at the atomic level and reveal structure-property relationships for knowledge and data-driven materials design. We mainly work on earth-abundant and environment friendly materials with a focus on 2D materials and perovskites. Notably, we take advantage of a host of computational tools at different levels of theory to fully capture the underlying physical phenomena governing the broad range of functional properties in the materials. Through close collaborations with experimental groups from both universities and national labs, our ultimate goal is to realize atomic precision and automation in materials design.

Quantum Mechanical Modeling and Materials Informatics

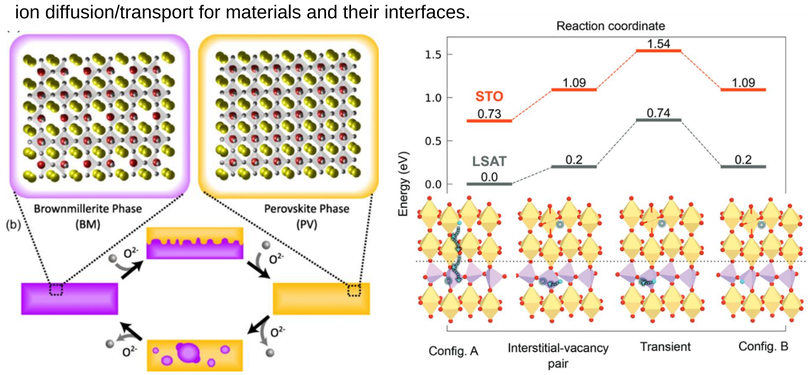




- Good balance between accuracy and computational cost
- Accurate description of complex electronic structures
- Feasibly screening wide chemical and structural spaces

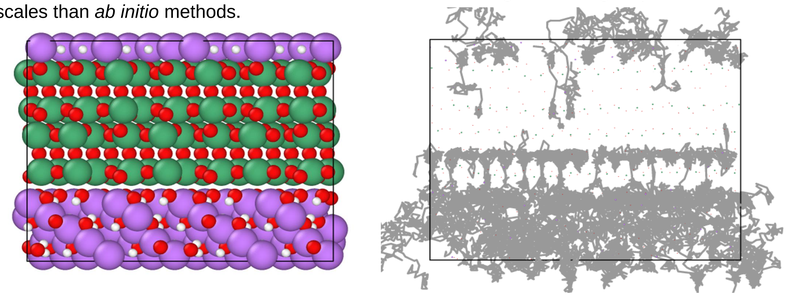
Energy Storage Materials

- We use **combined DFT and QMC** methods to predict the atomic structure, phase stability, and ion diffusion/transport for materials and their interfaces.



Adv. Mater., 35, 2305383, (2023). *Phys. Rev. Lett,* 129, 235701, (2022)

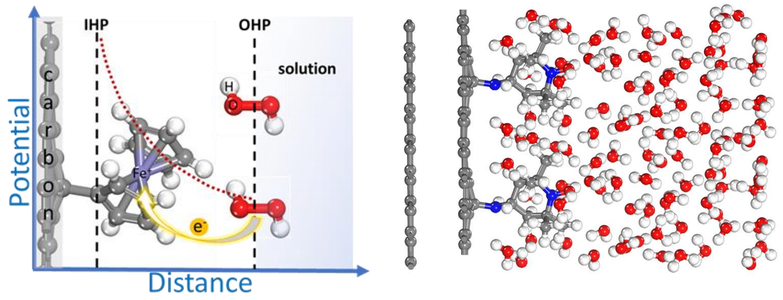
- We develop **machine learning force-fields (MLFFs)** using state-of-the-art graph neural network models to enable simulations at several orders of magnitudes larger spatial and time scales than *ab initio* methods.



Energy Conversion Materials

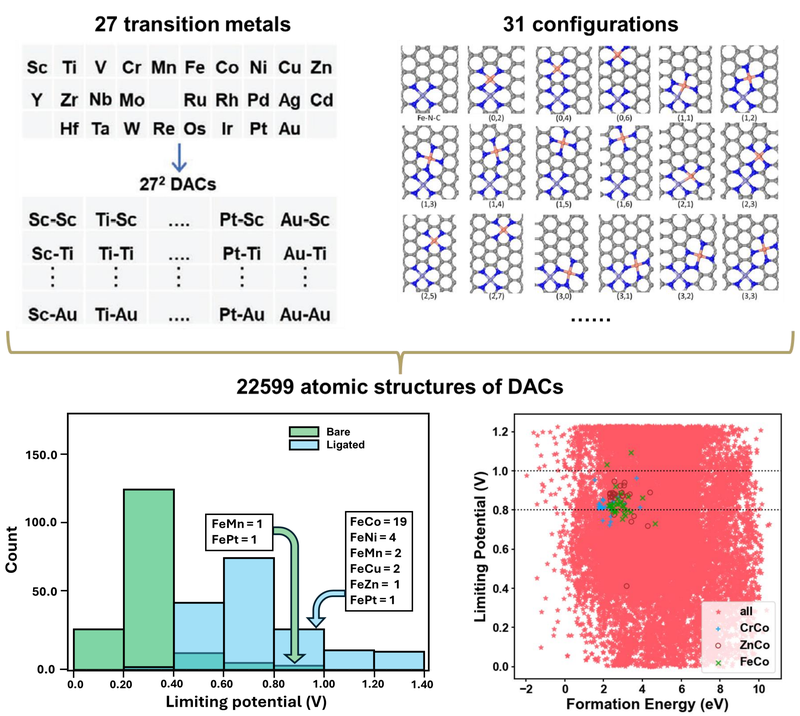
- We develop a concept of **voltage-driven molecular catalysis** for designing next-generation hybrid molecular/electrocatalysts for energy-related electrochemical processes.
- We perform **GCDFT calculations combined with hybrid solvation models,** and revealed that varying the applied voltage alters the potential drop and the chemical bonding, thereby increasing the reaction rate.

J. Am. Chem. Soc., 146, 28500, (2024). *J. Am. Chem. Soc.,* 145, 5786, (2023). *J. Am. Chem. Soc.*, 143, 17344, (2021).



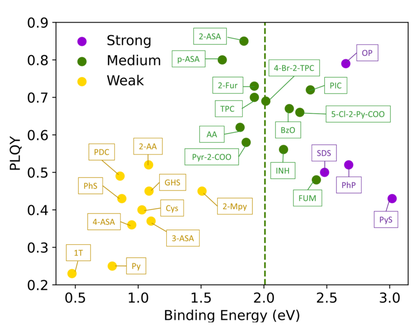
- In good agreement with experimental reports, our computational workflow predicts M1M2-N-C to be active for ORR, which **can not be captured by conventional computational models using bare structures.**
- Using **high throughput DFT combined with ML**, we efficiently evaluate over 22 K M1M2-N-C catalysts for ORR, and identify the top M1/M2 combinations.

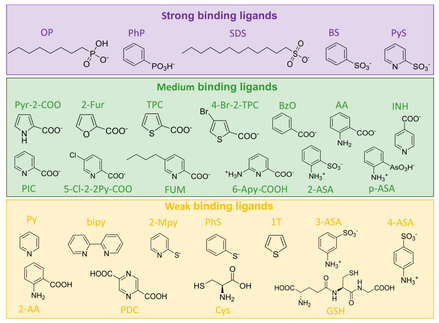
J. Am. Chem. Soc, under review. *ACS Catal.,* 13, 4992, (2023). *Nat. Energy,* 7, 281, (2022).



Energy Harvesting Materials

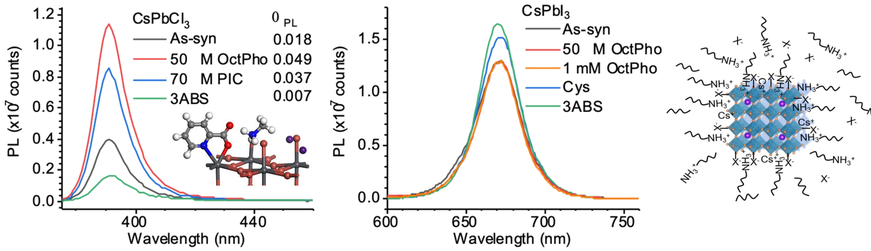
- We develop design principles for surface-passivating ligands of cesium lead halide perovskite nanocrystals in the **strongly quantum-confined regime**.





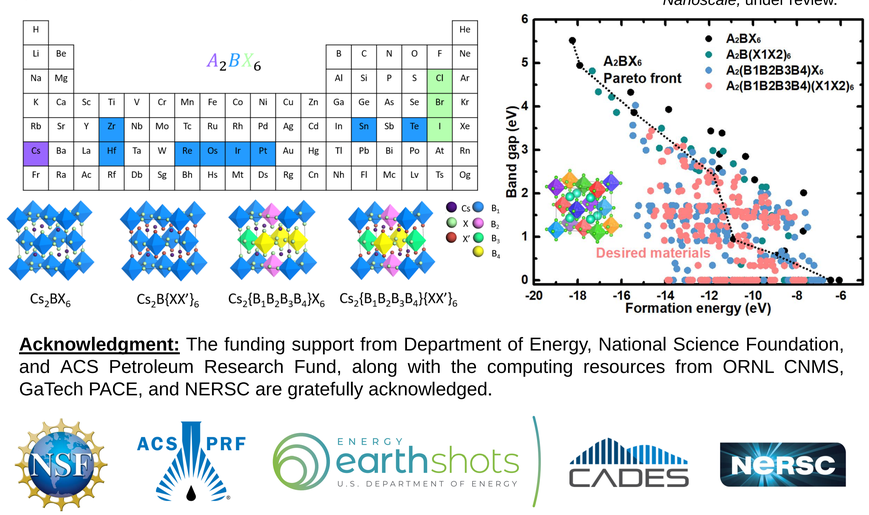
- We revealed **a volcano relationship** between the ligand binding energy and the experimental PLQY.
- Our preliminary calculations indicate that **conventional slab models fail to fully capture the organic/inorganic interface**. To address this, we leverage MLFF to conduct large-scale simulations of realistic nanocrystals.

J. Am. Chem. Soc, under review. *J. Phys. Chem. C*, 127, 1135, (2023). *J. Phys. Chem. C*, 125, 24521, (2021).



- We find medium/high-entropy vacancy-ordered double perovskites can **push the boundary of the stability and band gap pareto front**, exhibiting emerging properties which are not present in their pure counterparts.

Nanoscale, under review.



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