Final Technical Report

PI name: **Qimin Yan**

Project title: **Synthesis of motif and symmetry for accelerated learning, discovery, and design of electronic structures for energy conversion applications**

Institution: **Department of Physics, Temple University**

Local: **Philadelphia, PA 19122**

Award number: **DE-SC0020310**

Award duration: **09/01/2019 – 08/31/2022**

Overview of the project:

The overall goal of the projects is to develop a framework to incorporate structure motifs and crystal/orbital symmetries into the data-driven materials discovery infrastructure. The PI proposed to develop structure-motif- and symmetry-based graph convolutional networks for effective learning and efficient predictions of electronic structures and related properties. Fundamental understanding of the roles of structure motif and symmetry will establish new hypothesis and design rules, which will be combined with high-throughput computations based on density functional theory to discover novel light absorbers, transparent conductors, as well as 2D light emitting materials and heterojunctions for optoelectronics.

Scientific achievement:

Along the proposed research directions, we have made the following scientific achievements: (1) the incorporation of structure motif information in graph-based machine learning architecture; (2) incorporation of symmetry-based constraints in machine learning by contrastive representation learning; (3) applications of graph neural network to other important material problems such as material Hamiltonian learning; (4) discovery and design of function materials and heterojunctions for energy applications; (5) other collaborative work in the field of magnetic materials for energy applications. Eight publications supported by the grant have been published in the award period (3 years), including first-author papers published in Science Advances and Physical Review Materials, a first-author paper that is under review by npj Computational Materials, and collaborative publications with experimental groups in high-profiles journals (such as PNAS and ACS Nano). Following are the detailed scientific achievements of published work.

1. Incorporation of structure motif in graph-based machine learning

Incorporation of physical principles in a machine learning architecture is a fundamental step toward the continued development of artificial intelligence for inorganic materials. In the work we published in 2021 in Science Advances [Sci. Adv. 7, eabf1754 (2021)], as inspired by the Pauling's rule, we proposed that structure motifs in inorganic crystals can serve as a central input to a

machine learning framework. We demonstrated that the presence of structure motifs and their connections in a large set of crystalline compounds can be converted into unique vector representations using an unsupervised learning algorithm. We developed an atom-motif dual graph network (AMDNet) which is more accurate than atom-based graph neural network models in predicting the electronic structures of metal oxides such as band gaps. The work illustrates the route toward fundamental design of graph neural network learning architecture for complex materials by incorporating beyond-atom physical principles such as structure motifs. Based on this work, we are in the progress of constructing two different graph structures (bipartite graphs and hypergraphs) that incorporate motif information in materials networks and general graph-based machine learning for inorganic materials.

2. Incorporation of symmetry-based constraints in electron density learning

Another achievement we've made is the application of contrastive learning for the incorporation of symmetry-based constraints in machine learning, especially for density functional design. In a data-driven paradigm, machine learning is the central component for developing accurate and universal exchange-correlation (XC) functionals in density functional theory (DFT). In a work that is under review in npj Computational Materials [arXiv:2205.15071 (2022), npj Comput. Mater. *under review* (2023), DOI: 10.48550/arXiv.2205.15071], we demonstrate that contrastive learning is a computationally efficient and flexible method to incorporate a symmetry-based physical constraint in ML-based density functional design. We propose a schematic approach to incorporate the uniform density scaling property of electron density for exchange energies by adopting contrastive representation learning during the pretraining task. The results demonstrate that incorporating exact constraints through contrastive learning can enhance the understanding of density-energy mapping using neural network models. This work represents a viable pathway toward the machine learning design of a universal density functional via representation learning. Also, the contrastive learning framework can be applied to a large set of material science problems that are related to symmetry invariance.

3. Applications of graph neural network to other important material problems

Representing the interactions among atomic orbitals in any material, a material Hamiltonian provides all the essential elements that control the structure-property correlations in inorganic compounds. Effective learning of material Hamiltonian by developing machine learning methodologies therefore offers a transformative approach to accelerate the discovery and design of energy materials. With this motivation, in a collaborative work with Haibin Ling from Stony Brook [Neural. Comput. Applic. 34, 4625 (2022)], we present and compare several different graph convolution networks based on Hamiltonian matrix that are able to predict the band gap for inorganic materials. The models are developed to incorporate two different features: the information of each orbital itself and the interaction between each other. The results show that our model can achieve a promising prediction accuracy. It is one of the first works in the field that directly constructs graph neural networks for material property predictions with material Hamiltonian matrix as input.

In 2021, we published a review article in Computational Materials Science [Comput. Mater. Sci. 195, 110332 (2021)]. The article focuses on the recent development of graph-based deep learning frameworks and their applications for both molecules and solid-state material systems. The history of the development of graph-based representations for molecules and crystals was introduced. Current challenges and future perspectives on this emerging field at the crossroad of material science, physics, chemistry, and computer science was given, with an emphasize on how multiple tiers of material information can be organized and combined in a graph neural network setup.

4. Discovery of functional materials and heterojunctions for energy applications based on structure motif and symmetry

Discovery and design of two-dimensional (2D) materials with suitable band gaps and high carrier mobility are of vital importance for the photonics, optoelectronics, and high-speed electronics. In a work published in 2021 [Phys. Rev. Mater. 5, 014005 (2021)], based on high-throughput computations using density functional theory, we introduce a family of monolayer isostructural semiconducting tellurides $MNTe_4$, with M = {Ti, Zr, Hf} and N = {Si, Ge}. These compounds have been identified to possess direct band gaps from 1.0 to 1.31 eV, which are well suited for photonics and optoelectronics applications. Ultrahigh carrier mobility is predicted for this family

of 2D compounds, which host great promise for potential applications in high-speed electronic and optoelectronic devices. Detailed analysis of electronic structures reveals the origins of the promising properties of this unique class of 2D telluride materials.

In a collaborative work with Deep Jariwala's group at University of Pennsylvania [ACS Nano *in press* (2023), DOI:10.1021/acsnano.2c12546], using first-principle calculations, time-resolved and circularly polarized luminescence measurements, we studied in detail the excitonic effects in 2D heterostructures based on transition metal dichalcogenides (TMDs) and 2D perovskites and demonstrated that Rashba spin-splitting induced by inversion symmetry breaking in 2D perovskites and strongly coupled spin-valley physics in monolayer TMDs render spin-valley-dependent optical selection rules to the interlayer excitons. The work expands the scope for studying spin-valley physics in heterostructures of disparate classes of 2D semiconductors.

5. Other collaborative works in the field of magnetic materials for energy applications

2D magnetic layered materials have revolutionized size dependent magnetism to manipulate spin-based devices for energy applications. However, it has been challenging to artificially create 2D magnetic materials from bulk crystal structures with a variety of material groups. In a collaborative work with Shenqiang Ren's group at SUNY Buffalo [ACS Nano 16, 13232 (2022], we present the dimensionality manipulation via cation exchange of a 3D Prussian blue analogue toward a 2D magnetic sheet with the magnetic ordering temperature rising from 12 to 330 K. We predicted the pressure dependent magnetic tunability of such 2D networks using first-principles calculations and demonstrated it using the phase transitions of the hydrogel. This previously unobserved phenomenon of dimensional manipulation of a bulk crystal structure provides a rational strategy to expand the diversity and chemical compositions of 2D molecular magnetic material libraries.

Magneto-ionics promise ultralow-field sensor technologies and the extent of real-time ion insertion/extraction of an electrode is the key state-of-charge (SOC) feature in batteries. In another collaborative work with Shenqiang Ren's group [PNAS 119, e2122866119 (2022)], we report lithiating magneto-ionic material to enable the precise SOC sensor monitoring in Li-ion battery using a molecular magnetic electrode. The magneto-ionic-based sensor shows a more than 2,000%

increase in accuracy and a more than 5,000% reduction in response time. Simulations provided a mechanistic understanding of its magneto-ionic control, on which the compound undergoes a large lattice expansion after Li insertion due to the change in charge distribution. The findings provided the pathway toward the real-time accurate SOC estimation using molecular magnetic electrode.

Publication list:

Following is a cumulative list of all publications resulting from the work for the grant period (09/01/2019 – 08/31/2022) (personnel supported by the grant are highlighted):

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1. S. Singh, **W. Gong**, C. Stevens, J. Hou, A. Mohite, J. Hendrickson, **Q. Yan**, D. Jariwala, "Valley Polarized Interlayer Excitons in 2D Chalcogenide-Halide Perovskite-Van der Waals Heterostructures", ACS Nano *in press* (2023), DOI:10.1021/acsnano.2c12546

2. **W. Gong**, T. Sun, H. Bai, P. Chu, A. Aryal, J. Yu, H. Ling, J. P. Perdew, **Q. Yan**, "Incorporation of density scaling constraint in density functional design via contrastive representation learning" arXiv:2205.15071 (2022) npj Comput. Mater. *under review* (2023), DOI: 10.48550/arXiv.2205.15071

3. Y. Huang, **W. Gong**, G. Zhang, Z. Li, H. Lin, **Q. Yan**, S. Ren, "Dimensional Transformation of Molecular Magnetic Materials" ACS Nano 16, 13232 (2022), DOI: 10.1021/acsnano.2c06912

4. Y. Hu, **W. Gong**, S. Wei, S. Khuje, Y. Huang, Z. Li, Y. C. Li, F. Yao, **Q. Yan**, S. Ren, "Lithiating Magneto-Ionics in Rechargeable Battery" PNAS 119, e2122866119 (2022), DOI: 10.1073/pnas.2122866119

5. H. Bai, P. Chu, **J. Y. Tsai**, N. Wilson, X. Qian, **Q. Yan**, H. Ling, "Graph Neural Network for Hamiltonian-Based Material Property Prediction" Neural. Comput. Applic. 34, 4625 (2022), DOI: 10.1007/s00521-021-06616-0

6. **H. R. Banjade**, S. Hauri, S. Zhang, F. Ricci, W. Gong, G. Hautier, S. Vucetic, **Q. Yan**, "Structure motif centric learning framework for inorganic crystalline systems", Sci. Adv. 7, eabf1754 (2021), DOI: 10.1126/sciadv.abf1754

7. **W. Gong**, **Q. Yan**, "Graph-based deep learning frameworks for molecules and solid-state materials",
Comput. Mater. Sci. 195, 110332 (2021), DOI: 10.1016/j.commatsci.2021.110332

8. **H. R. Banjade**, J. Pan, **Q. Yan**, "Monolayer 2D semiconducting tellurides for high-mobility electronics" Phys. Rev. Mater. 5, 014005 (2021), DOI: 10.1103/PhysRevMaterials.5.014005

Personnel supported by this grant:

Three graduate students (Huta Banjade, Jeng-Yuan Tsai, and Weiyi Gong) and one postdoc (Debajit Chakraborty) were fully or partially supported by this grant.

PhDs were granted to the two students supported by this grant: Huta Banjade (full support from 2019 to 2020) and Jeng-Yuan Tsai (partial support in 2022).

Another student supported by this grant, Weiyi Gong (full support from 2020 to 2022), will receive his PhD from Northeastern University in the coming year.

Owing to the work supported by this grant, the PI was selected as finalist for "2020 Rising Stars in Computational Materials Science Prize" and a review article was published in the special issue of Computational Materials Science in 2021.