Field of Interest and the Role of Computational Science

1. *In terms a general audience would understand, describe an important, outstanding scientific or engineering challenge in your field of interest where computational science can play an important role.*
2. *Describe the particular science or engineering problem that you would like to pursue in your research. What would be the impact on the field and/or on science, engineering and/or society in general if this challenge could be successfully addressed? (2250 characters)*

2D materials, such as transition metal dichalcogenides (TMDs), are atomically thin semiconductors with potential for flexible electronics and vertically stacked circuits. Their quantum properties, like their “valley” degree of freedom, offer a novel way to store information. However, a crucial difference from conventional bulk semiconductors is that 2D TMDs form strongly bound excitons— electron-hole pairs— when excited by light. Maximizing their potential thus requires understanding how their excitons can be manipulated for quantum applications.

Computation is crucial for studying excitons because they are challenging to directly image in experiments and they have complex interactions with each other and the environment. Further, the huge phase space of possible structures cannot be addressed experimentally (just 17 common semiconducting monolayers and 10 twist angles at each interface give 17 million 4-layer structures).

My research will focus on interlayer excitons, which are spatially separated electrons and holes in stacked TMD bilayers. Interlayer excitons are long-lived and maintain spin-valley polarization, so they can store quantum information. While basic exciton properties in popular TMDs like MoS2 and WS2 have been explored, their interactions, stabilities and tunability are less established. For instance, a critical open question is how to couple different excitons in a stable and scalable way, in order to transform their quantum information into correlated photon pairs to transport information in quantum networks.

Using first-principles simulations based on many-body perturbation theory, I will determine mechanisms of exciton-exciton coupling as a function of their spatial configuration (confined to one layer or not), spin quantum number, and external perturbations. Given the long-term goal of using interlayer excitons in real devices, I hope to predict their dynamical properties in realistic conditions, including finite temperature and encapsulation by passivating materials.

If successful, my research will progress our knowledge of fundamental exciton physics and could pave the way for a new generation of optoelectronic and quantum devices, such as spin- and valley-based quantum devices.

Use of Computational Science in your research

1. *What is the most complex calculation you have run on a high-performance machine as part of your research experience? Or if you haven't run a high-performance computing system, tell us about the most complex computational problem you have tackled.*
2. *Imagine if you were given access to resources 100 times more powerful than what you have access to. What would that enable you to do, and what do you perceive the mathematical and computer science challenges to be? (2250 characters)*

I used density functional theory to study lattice vibrations (phonons) in layered perovskites, which determines their thermal properties. This was a complex calculation because the system consisted of over 100 atoms in both inorganic and organic phases.

During convergence testing, I found that the parameters required for accurate answers (high plane wave energy cutoffs) scaled poorly with the number of nodes. Thus, I had to systematically try many parallelization schemes to maximize efficiency given constraints on computer resources. I also created approximations to reduce the computational cost, such as replacing asymmetrical molecules with ions to exploit crystal symmetries, which was previously overlooked. Ultimately, using a series of calculations on 280 cores, I produced the first phonon dispersion of layered hybrid perovskites, providing a microscopic view of their thermal properties.

More recently, I was trained on using the Frontera supercomputer, using up to 60 nodes to calculate optical properties of 2D materials. While machine learning (ML) has become a useful tool in many areas of computational materials science, ML is not straightforwardly applied to accelerate excited-state calculations. Given 100x more computing power, we can imagine generating, for the first time, sufficient data to train accurate ML models of excited-state potential energy surfaces with immediate relevance to photocatalysis and quantum computing research.

Despite these exciting possibilities, key barriers persist. Excited-state calculations typically scale quartically, limiting their application to small systems. We can instead use stochastic approaches for larger systems, which are costly now but scale quadratically or linearly. Additionally, excited state calculations routinely involve high-precision, distributed dense linear algebra, often incompatible with the latest hardware (e.g. single-precision GPUs and non-uniform memory access). Taking full advantage of expanding computational power would ideally require users to co-design hardware with computer scientists or, at least, to work closely with applied mathematicians to use algorithms where data layouts are compatible with memory access bandwidth and locality.

Program of Study

Describe how the courses listed in your planned program of study would help prepare you to address the challenges you have described in questions 1 and 2. Discuss your rationale for choosing these courses. (2250 characters)

Through my program of study, I hope to develop i) a rigorous understanding of the theoretical framework to describe complex excitations in 2D transition metal dichalcogenides and ii) proficiency with the computer science tools to implement simulations of these excitations.

My science and engineering courses build on my experience with ground-state electronic structure methods. In Quantum Theory of Electronic and Optical Excitations and Waves and Diffraction, I will dive into the mathematical formalisms for light-matter interactions and time-dependent phenomena. These topics will enable me to describe how excitons in 2D TMDs scatter, recombine, and thermalize. Finally, Electronic and Optical Properties of Solids emphasizes optoelectronic properties from a device perspective, contextualizing my research in realistic environments.

Given the ubiquity of linear algebra in quantum mechanics, I will take two advanced linear algebra courses to reinforce my understanding of the mathematical concepts. I will also develop algorithmic and software design skills relevant to dense matrix computations through courses on distributed algorithms, parallel computing, and Fortran software design. While code development will not be my primary focus, I hope to contribute to the scientific computing software used in my research. Further, by expanding my knowledge of high-performance computing, I will be able to better communicate physicists' needs to computer scientists who develop the underlying hardware and algorithms we use.

Beyond my research, my chosen courses connect me to other active areas of computational science. Statistics courses will deepen my understanding of adjacent methods for simulating excited states, such as quantum Monte Carlo and stochastic many-body perturbation theory. Given the proliferation of machine learning in materials science, I will also take Artificial Intelligence to understand the opportunities and challenges associated with developing machine learning models for excited state processes.

I believe the interdisciplinary program proposed will enable me to foster collaboration among experimentalists, theorists, and computer scientists and conduct innovative research on excitons in 2D materials.