Abstract: Startup Allocation Request for Quantum-Mechanical Investigation of Structure-Property Relationships in Organic Emitters for Efficient Thermally Activated Delayed Fluorescence (TADF), Y1

Thermally Activated Delayed Fluorescence (TADF) is one of the most promising mechanisms to realize high efficiency Organic Light Emitting Diodes (OLEDs) without the use of heavy transition metals and offers flexibility to fine-tune the electronic and optical properties of purely organic molecules. In TADF, nonemissive triplet excitons are harvested through a designed molecular emitter that undergoes efficient thermally activated reverse intersystem crossing (RISC) to a singlet manifold, which is followed by a radiative relaxation to the ground state. The rate of the RISC depends on the singlet-triplet energy gap, deltaE(ST), which needs to be minimized for an efficient TADF process. To minimize deltaE(ST), organic molecules with electron donor (D) and acceptor (A) groups are utilized, such that, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) are localized on the donor and acceptor, respectively, giving spatially separated HOMOs and LUMOs, giving in a small deltaE(ST). While D-A molecules were the first examples of effective TADF materials, how this efficiency is achieved in devices is not understood. We will use multi-scale quantum-chemical methods to characterize the (opto)electronic properties of carbazole-based TADF emitters with known D-A groups. We will benchmark our calculations with these systems. We will develop: (1) a comprehensive description, at the molecular level, of the fundamental photophysical processes of TADF emitters; (2) a modelling protocol that can accurately describe the electronic structure of such emitters; (3) structure-property relationships; and provide theoretical guidelines for the design of new materials and/or selection of existing materials with well-defined properties leading to devices with improved performances.

The above mentioned quantum-chemical calculations will require a High Performance Computing (HPC) cluster. Currently, there is no such HPC cluster present at the home institution. Therefore, we propose to use XSEDE resources to perform the above-mentioned theoretical calculations. We will use Gaussian 16 to perform DFT calculations. Therefore, we request to access Bridges and Pylon where we can access Gaussian 16. We request: Bridges RM (50K SU) and Pylon (500 GB) to run test calculations and benchmarks leading to the development of a research proposal.