Tailoring excitonic and vibronic properties of 2D materials for energy applications

Ab initio computational methods such as density functional theory (DFT), time-dependent DFT, the GW approximation to the electronic self-energy, and the Bethe-Salpeter equation (BSE) can characterize the ground-state and electronic excitations in a materials specific way. However, electronic excitations typically induce, and are coupled to lattice vibrations. Treating such exciton-phonon coupling goes beyond present state-of-the-art computational methods, yet is essential for accurately describing the internal dynamics of solar cells and the efficacy of photo-catalytic processes including hydrogen production. Green's functions based methodological extensions of the standard BSE technique allow us to account for exciton-phonon coupling [1,2]. Computational results will be presented in the context of resonant inelastic x-ray scattering, which has emerged in recent years as a uniquely powerful probe for quantifying vibronic interactions throughout the Brillouin zone.

Due to weakened electronic screening, electron-phonon and exciton-phonon coupling plays a larger role in two-dimensional (2D) materials and contributes to some of their unique phenomena that have long fascinated physicists. I will introduce a few exciting scientific opportunities in the further development of 2D materials for energy applications, and then address some of the associated computational and materials challenges.

[1] Geondzhian & Gilmore, Physical Review B 98, 214305 (2018)
[2] Geondzhian et al., Physical Review Letters 125, 126401 (2020)