Ab initio approaches to nonequilibrium interactions in quantum matter

Prineha Narang

Harvard University, USA

prineha@seas.harvard.edu

Quantum systems host spectacular excited-state effects, but many of these phenomena remain challenging to control and, consequently, technologically under-explored. My research, therefore, focuses on how quantum systems behave, particularly away from equilibrium, and how we can harness these effects1. By creating predictive approaches to study dynamics, decoherence and photo-induced correlations in matter, our work could enable technologies that are inherently more powerful than their classical counterparts ranging from quantum information science, to ultra-high efficiency optoelectronic and energy conversion systems. In this talk, I will present a pedagogical introduction to theoretical and computational approaches to describe excited-states in quantum matter, and predicting emergent states created by strongly non-equilibrium external drives. Understanding the role of such nonequilibrium light-matter interactions in the regime of correlated electronic systems is of paramount importance to fields of study across condensed matter physics, quantum optics, and quantum chemistry. The simultaneous contribution of processes that occur on many time- and length-scales have remained elusive for state-of-the-art calculations and model Hamiltonian approaches alike, necessitating the development of new methods in theoretical and computational quantum chemistry 2–6. I will discuss our work at the intersection of ab initio cavity quantum-electrodynamics and electronic structure methods to treat electrons, photons and phonons on the same quantized footing, accessing new observables in strong light-matter coupling. Current approximations in the field almost exclusively focus on electronic excitations, neglecting electron-photon effects, for example, thereby limiting the applicability of conventional methods in the study of quantum chemical and polaritonic systems, which requires understanding the coupled dynamics of electronic spins, nuclei, phonons and photons. With our approach we can access correlated electron-photon and photon-phonon dynamics. Building on this, I will show selected examples of our approach in ab initio design of active defects in quantum materials leveraging the chemical degree-of-freedom7–9 towards selectively linking these active defects 10–12. Finally, I will present an outlook on driving quantum chemical systems far out-of-equilibrium to control the coupled electronic and vibrational degrees-of-freedom 13,14 and a pathway to link these with transport in materials15,16.

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