Simulation of Excitation by Sunlight in Mixed Quantum-Classical Dynamics

Mario Barbatti

Aix-Marseille University, Marseille, France

In this talk, I will discuss a method I have recently proposed to simulate nonadiabatic dynamics initiated by thermal light, including solar radiation, in the frame of mixed quantum-classical (MQC) methods, like surface hopping. The method is based on the Chenu–Brumer approach, which treats the thermal radiation as an ensemble of coherent pulses. It is composed of three steps: (1) sampling initial conditions from a broad blackbody spectrum, (2) dynamics propagation using conventional methods, and (3) ensemble averaging considering the field and realization time of the pulses. The application of MQC dynamics with pulse ensembles (MQC-PE) to a model system of nucleic acid photophysics showed the emergence of a steady excited-state population. In another test case, modeling retinal photophysics, MQC-PE predicted that although the photoisomerization occurs within 200 fs, it may take tens of microseconds of continuous solar irradiation to photoactivate a single retinal. Such emergent long timescales may impact our understanding of biological and technological phenomena occurring under solar radiation.