Departmental Colloquium

Department of Physics, Kent State University
Kent, Ohio

Name: Hanning Chen, Ph.D.

Institute: Argonne-Northwestern Solar Energy Research Center
Department of Chemistry, Northwestern University
Evanston, Illinois 60208

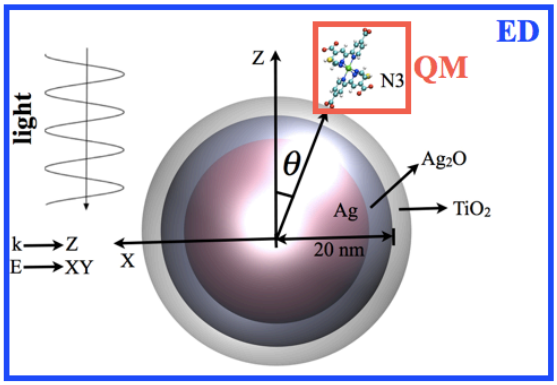
Title: Time-dependent Theories for Photo-induced Chemical Processes

Abstract:

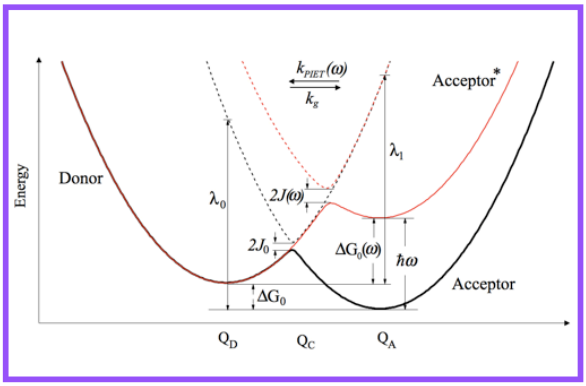
One of the most important and characteristic features of any physical object is the response of that object to light. To meet the emerging challenges of multi-scale modeling in multi-component novel optical devices such as dye-sensitized solar cells, single-molecule detectors and photocatalytic reactors, we have recently developed two time-dependent optical theories, namely the hybrid quantum mechanics/classical electrodynamics approach (QM/ED) and the constrained real-time time-dependent density functional theory (C-RT-TDDFT).

The QM/ED approach was introduced to effectively incorporate the scattered electric field of a plasmonic nanoparticle into the quantum mechanical optical property calculation for a nearby dye molecule. At no additional computational cost compared to the conventional classical electrodynamics and quantum mechanics calculations, this method provides a reliable way to couple the individual electronic transition of a dye molecule to the collective dielectric response of a nanoparticle.

The C-RT-TDDFT theory was formulated to accurately evaluate the effective photo-induced electronic coupling strength by tracking the time evolution of diabatic wavefunction overlap. In combination with the semi-classical Marcus theory, the rate of photo-induced electron transfer can be determined by treating the process as a coherent interference between the vertical excitation and the parallel transition. In addition, other important optical properties such as quantum yield and electron injection rate are also readily available.



hybrid quantum mechanics/classical electrodynamics (QM/ED)



constrained real-time time-dependent DFT (C-RT-TDDFT)