NANYANG TECHNOLOGICAL UNIVERSITY SINGAPORE

School of Materials Science and Engineering

Seminar Topic:
Singlet Fission: Dynamics and Spectroscopy

Associate Professor Zhao Yang

Abstract

Intramolecular singlet fission (iSF) materials provide remarkable advantages in terms of tunable electronic structures, and quantum chemistry studies have indicated strong electronic coupling modulation by high-frequency phonon modes. We formulate a microscopic model of iSF with simultaneous diagonal and off-diagonal coupling to high-frequency modes. A nonperturbative treatment, the Dirac-Frenkel time-dependent variational approach is adopted using the multiple Davydov trial states. It is shown that both diagonal and off-diagonal coupling can aid efficient singlet fission (SF) if excitonic coupling is weak; and fission is only facilitated by diagonal coupling if excitonic coupling is strong.

In the presence of off-diagonal coupling, it is found that high-frequency modes create additional fission channels for rapid iSF. Separately, we investigate nonlinear spectra that monitor SF processes mediated by a conical intersection (CI) of two lowest excited electronic states for various optical dephasings. Simulated 2D spectra at different population times follow ultrafast population transfer through the CI and exhibit spectral features related to the tuning mode. The anticorrelated oscillations of cross peaks located at symmetric positions with respect to the main diagonal are clearly identified after the stimulated-emission contribution quenches. Simulated transient absorption signals show a fast decay of 1st excited singlet state and exhibit multiple peaks revealing the tuning mode

Biography

Dr Zhao Yang was selected from the best physics students to pursue PhD studies in the University of California at San Diego, mentored by Prof Katja Lindenberg, under the prestigious CUSPEA program sponsored by Nobel Laureate Tsung-Dao Lee. His Bachelor of Science was awarded by Zhejiang University. Following a brief stay in International Centre for Theoretical Physics (ICTP) in Trieste, Dr Zhao took up a postdoctoral appointment at Rochester Theory Center in University of Rochester, where he worked with Prof Shaul Mukamel in Chemistry and Prof Bob Knox in Physics. Prior to joining MSE in Jan 2007, he held positions in University of Hong Kong that include Research Assistant Professorship and Honorary Assistant Professorship.

Dr Zhao's research interests include ultrafast quantum dynamics in various materials and processes, light-matter interactions and nonlinear spectroscopy, light-harvesting in natural and artificial photosynthesis, quantum dissipation in many-body systems, and thermodynamics of small quantum systems. Dr Zhao was the lead-PI of a 10-million-SGD National Research Foundation project entitled "Toward Efficient Sunlight Harvesting."

Wednesday, 9 October 2019 || Time: 2:00 pm – 3:00 pm
Venue: MSE Meeting Room (N4.1-01-28)
Hosted by: Associate Professor Huang Yizhong

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