



SEMINAIRE ISMO

Alexander SCHUBERT

*Institute of Physical and Theoretical Chemistry,
University of Würzburg, Germany*

Ultrafast exciton self-trapping upon geometry deformation in perylene-based molecular aggregates

Perylene-based organic semi-conductors are intensively investigated since they represent promising materials for thin film electronic devices such as organic solar cells. These investigations have led to tremendous progress but various shortcomings still block the way towards higher efficiencies and/or simpler device architectures. One source for the lack of efficiency regarding excitation energy transport are exciton self-trapping processes. Theoretical approaches which simulate these self-trapping effects with atomistic resolution will considerably contribute to a better understanding of the photophysics of perylene-based dyes.

We introduce such a combined theoretical and experimental approach and use it to reveal the multi-step self-trapping mechanism in perylene tetracarboxylic acid bisimide (PBI) H-aggregates. Femtosecond time-resolved experiments demonstrate that the photo-excited state of PBI in solution decays non-radiatively on a timescale of 215 fs. High-level electronic structure calculations on dimers point towards the importance of an excited state intermolecular geometry distortion along a reaction coordinate which induces energy shifts and non-adiabatic couplings between various electronic states. Time-dependent wave-packet calculations incorporating a simple dissipation mechanism indicate that the fast energy quenching results from a doorway state with a charge-transfer character which is only transiently populated. Therefore, morphologies in which this channel is effectively suppressed could represent an important step on the road to a rational design of improved materials.

Mardi 12 février 2013 à 11h

Bât. 351 – 2^{ème} étage

Université Paris-Sud 91405 ORSAY Cedex