SYMN 1: Transport and Spectroscopy in Molecular Nanostructures

Time: Wednesday 10:30–13:00 Location: HSZ 01

Invited Talk SYMN 1.1 Wed 10:30 HSZ 01

Exciton localization and dynamics in molecular aggregates — •Jasper Knoester — Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747AG Groningen, The Netherlands

Self-assembled aggregates of dye molecules are interesting nanoscale systems with a variety of morphologies and dimensionalities in which energy transport and optical properties are governed by Frenkel excitons. The latter are collective electronic excitations, the extent of which is determined by the interplay between intermolecular excitation transfer interactions within the aggregate and the interaction with other degrees of freedom (static disorder and vibrations). The interest in these systems results from their strong interaction with light, their often sharp and color-tunable absorption bands, and their fast energy transport. In this talk, I will address recent results on exciton localization and dynamics in linear and tubular molecular aggregates, and the associated spectral signatures. I will discuss the model of delocalized excitons weakly scattered on vibrations, which has allowed good comparison to experiments for a variety of properties and systems. I will also address surprising properties of linear exciton systems with diagonal disorder taken from distributions with divergent second moments (Levy disorder). Finally, I will go beyond the picture of weak exciton-vibration interaction by explicitly including vibronic effects, thus addressing the dynamics of exciton-polarons. Various spectroscopies, such as absorption, linear and circular dichroism, and two-dimensional correlation spectroscopy, will be discussed.

Invited Talk SYMN 1.2 Wed 11:00 HSZ 01

Spectroscopy and transport in carbon nanotubes and graphene nanoribbons for electronics and biological applications — •Oleg Prezhdo — University of Rochester, Department of Chemistry, Rochester, NY 14627, USA

Carbon nanotubes (CNTs) and graphene nanoribbons (GNRs) constitute a new class of materials with many potential applications. We will discuss the results of our time-domain ab initio studies of these materials aimed at designing biological detection tools and miniature electronics devices. First, we will focus on the spectroscopy of semiconducting CNTs and GNRs. These materials can replace traditional semiconductors, such as silicon, in electronics applications, allowing one to create smaller devices. In order to achieve this goal, the electrons and holes in semiconducting CNTs and GNRs should live for sufficiently long times. However, spectroscopic experiments show that the electron-hole pairs tend to recombine nonradiatively on a fairly short timescale. We will discuss the origin and mechanism of the nonradiative recombination and the role of structural and chemical defects in this process. Second, we will discuss the application of CNTs and GNRs for determining DNA sequence. In comparison to the current sequencing tools, CNTs and GNRs have a great potential for significantly faster and cheaper detection. DNA naturally wrap around CNTs, and DNA sequence can be read off the CNT by scanning tools. By creating pores in GNRs and pulling DNA through these pores, one can detect the modulation of the electrical current along GNRs and obtain the DNA sequence rapidly and with a single base resolution.

Invited Talk SYMN 1.3 Wed 11:30 HSZ 01

Multidimensional Optical Spectroscopy of Biological Complexes — •Shaul Mukamel — University of California, Irvine, Department of Chemistry, Irvine, CA 92697-2025, USA

The response of complex molecules to sequences of femtosecond optical pulses provides multidimensional snapshots of their structure and electronic and vibrational dynamics. Two-dimensional correlation plots of the signals show characteristic cross-peak patterns which carry information about structures, equilibrium binding fluctuations and relaxation processes.

2D signals of photosynthetic light harvesting complexes reveal couplings between chromosphores and signatures of quantum coherence. Quantum effects stemming from the entanglement of chromophores, and their spectroscopic signatures are predicted. Simulations of the photosynthetic reaction center of photosystem II based on the Lindbladt approach to quantum exciton dissipation clearly establish oscillatory energy transport at room temperature originating from interference of quantum pathways. Entangled photons provide new avenues for controlling and manipulating these signals.

Simulations will be presented for chiral infrared and ultraviolet probes of the structure and electrostatic fluctuations in amyloid fibrils. Time-domain experiments that employ sequences of attosecond x-ray pulses in order to probe electronic and nuclear dynamics in molecules are proposed. By creating multiple core holes at selected atoms and controlled times it is possible to study the dynamics and correlations of valence electrons as they respond to these perturbations.

Invited Talk SYMN 1.4 Wed 12:00 HSZ 01

Theory of light-harvesting in photosynthetic pigment-protein complexes — •Thomas Renger[1], Marcel Schmidt am Busch[1], M. El-Amine Madjet[2], and Frank Müh[1,2] — [1]Johannes Kepler University Linz, Institute of Theoretical Physics, Altenberger Str. 69, 4040 Linz, Austria — [2]Freie Universität Berlin, Institut für Chemie und Biochemie, Fabeckstr. 36a, 14195 Berlin, Germany

In photosynthesis, light energy absorbed in so called antenna or light-harvesting complexes is transferred via an exciton mechanism to a reaction center where it is used to drive electron transfer reactions. The quantum efficiency of the transfer is close to 100 percent, that is, all excitons created reach the reaction center. In order to bridge the gap between the crystal structures of these light-harvesting proteins and optical experiments probing their function, two essential problems need to be solved. On one hand, theories of optical spectra and excitation energy transfer have to be developed that take into account the pigment-pigment (excitonic) and the pigment-protein (exciton-vibrational) coupling on an equal footing. On the other hand, the parameters entering these theories need to be calculated from the structural data. I will give a summary of theories on dissipative quantum dynamics of photosynthetic excitons, of recent approaches to a structure-based calculation of the parameters of the theory and of applications on different light-harvesting and reaction center complexes revealing different strategies for efficient light-harvesting realized in different systems.

Invited Talk SYMN 1.5 Wed 12:30 HSZ 01

How do algae use quantum mechanics to harvest light for photosynthesis? — •Gregory Scholes — University of Toronto, Toronto, Canada

Light-harvesting in photosynthesis involves amplification and regulation of light capture by reaction centers using energy transfer from antenna proteins. Efficient energy transfer among specialized antenna proteins enables organisms to adapt to local solar spectra (e.g. at various depths in the ocean) and to function even under quite low light fluxes. The key biophysical processes at play have been discovered by investigating electronic energy transfer in a variety of photosynthetic proteins. A new twist on this old problem is suggested by recent research that provides evidence that these dynamics can involve quantum-coherence, even at ambient temperature conditions. Our experiments, using two-dimensional photon echo spectroscopy, have revealed that in light-harvesting antenna proteins isolated from a family of marine cryptophyte algae, quantum-coherence effectively 'wires' together the light-absorbing molecules in order to facilitate efficient long-range energy transfer [1]. In this talk I will describe the measurement, using 2D-photon echo spectroscopy, of quantum-coherence in energy transfer and I will report new insights into the mechanism underpinning the process.

[1] E. Collini, C. Y. Wong, K. E. Wilk, P. M. G. Curmi, P. Brumer, and G. D. Scholes, "Coherently wired light-harvesting in photosynthetic marine algae at ambient temperature", Nature 463, 644-648 (2010).