Fritz-Haber-Institut der Max-Planck-Gesellschaft

Physikalische Chemie — Direktor: Prof. Dr. Martin Wolf

MAX-PLANCK-GESELLSCHAFT

Department Seminar:

Monday, December 18, 2017, at **11:30** a.m.;
— all are invited to meet at around 11:10 for a chat and coffee —

Dr. Hélène Seiler Kambhampati Research Group,
Department of Chemistry
McGill University, Montréal.

Development of an "optical NMR" spectrometer to investigate electronic couplings in molecules and nanostructures

PC Seminar Room G2.06, Building G, Faradayweg 4 M. Wolf

Abstract:

Coherent Multi-Dimensional Spectroscopy (CMDS) is a powerful technique that is directly sensitive to couplings between quantum states. In the optical regime, the technique is well-suited to investigate interactions between the electronic degrees of freedom in systems such as biological light-harvesting complexes and nanostructures.

Following a general introduction on multi-dimensional spectroscopy, I will present an ultrafast optical two-dimensional spectrometer based on a hollow-core fiber for broadband visible continuumgeneration and two acousto-optic pulse shapers arranged in a Mach-Zehnder interferometer for the production of fully-coherent pulse trains. The setup can easily switch between a pump-probe geometry and a collinear geometry with polarization shaping capabilities. The methodological improvements presented here represent important enabling steps towards the longstanding goal of achieving an "Optical NMR", and extends the realm of all-optical multi-dimensional spectroscopy to spatially heterogeneous samples.

The methods developed are then applied on two classes of systems. The model system Nile Blue is used to validate the performance of the instrument. The spectrometer is also used to reveal new processes in colloidal semiconductor CdSe nanocrystals. One of the most fascinating aspects of semi-conductor nanocrystals is their ability to host multiple excitations per particle. When multiple excitons are created in the same nanocrystal, bound quasi-particles called multiexcitons form. In contrast to the single exciton, the structural and dynamics properties of multiexcitons remains, to this day, relatively poorly understood due to their complexity. In the last part of the seminar, I will discuss new insights gained on the early-time dynamics and structure of the ground state biexciton thanks to the optical CMDS method.