Symposium Multidimensional coherent spectroscopy of functional nanostructures (SYCS)

jointly organised by
the Semiconductor Physics Division (HL),
the Chemical and Polymer Physics Division (CPP), and
the Surface Science Division (O)

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Overview of Invited Talks and Sessions

(Lecture hall Audimax 1)

Invited Talks

SYCS 1.1	Tue	10:00–10:30	Audimax 1	**Multidimensional coherent spectroscopy of perovskite nanocrystals** — •Steven Cundiff, Albert Liu, Diogo Almeida, Gabriel Nagamine, Lazaro Padilha
SYCS 1.2	Tue	10:30–11:00	Audimax 1	**Coherent multidimensional techniques for the characterization of nanomaterials** — •Elisabetta Collini
SYCS 1.3	Tue	11:00–11:30	Audimax 1	**Exciton Dynamics revealed by Multidimensional Coherent Spectroscopies applied to Light-Harvesting Systems** — •Thomas L.C. Jansen
SYCS 1.4	Tue	11:45–12:15	Audimax 1	**Revealing couplings with action-based 2D microscopy** — •Tobias Brixner
SYCS 1.5	Tue	12:15–12:45	Audimax 1	**Low-frequency phonons affect charge carrier dynamics in hybrid perovskites** — •Mischa Bonn

Sessions

SYCS 1.1–1.5	Tue	10:00–12:45	Audimax 1	**Symposium: Multidimensional coherent spectroscopy of functional nanostructures**

SYCS 1: Symposium: Multidimensional coherent spectroscopy of functional nanostructures

Multidimensional coherent spectroscopy has recently enabled fundamentally new insight into non-equilibrium quantum dynamics underlying elementary photophysical and photochemical processes, energy relaxation, charge transfer, and strong coupling phenomena in biological and artificial molecular systems, and semiconductors nanostructures. This knowledge has significant implications for the design of functional materials and for their application in optoelectronics. The aim of this Symposium is to bring together international leading experts to address the most recent developments and future research directions in multidimensional coherent spectroscopy techniques for the study of elementary light-driven processes in functional materials and nanostructures.

Organizers: Antonietta De Sio, Christoph Lienau (Universität Oldenburg)

Time: Tuesday 10:00–12:45 Location: Audimax 1

Invited Talk SYCS 1.1 Tue 10:00 Audimax 1
Multidimensional coherent spectroscopy of perovskite nanocrystals — •Steven Cundiff[1], Albert Liu[1], Diogo Almeida[1,2], Gabriel Nagamine[2], and Lazaro Padilha[2] — [1]University of Michigan, Ann Arbor, MI, USA — [2]University of Campinas, Campinas, SP, Brazil

Perovskite nanocrystals, including nanocubes and nanoplatelets, have attracted signficant attention recently due to their novel optoelectronic properties, in particular their high fluorescence quantum efficiency. However there are still debates about their properties, including electronic structure and optical linewidths. Measurements of these properties are hindered by the inhomogeneous broadening due to size dispersion.

We use optical multidimensional coherent spectroscopy at low temperatures to remove the effects of inhomogeneous broadening and elucidate the intrinsic properties of the nanocrystals. This include insight into the ordering of the bandedge states, dephasing rates of triple coherences and dependence of the dephasing excitonic dephasing on the number of layers in nanoplatelets.

Invited Talk SYCS 1.2 Tue 10:30 Audimax 1
Coherent multidimensional techniques for the characterization of nanomaterials — •Elisabetta Collini — Department of Chemical Sciences, University of Padova, via Marzolo 1, 35131 Padova, Italy

2D electronic spectroscopy (2DES) techniques have gained particular interest given their capability of following ultrafast processes in real-time. These techniques have been primarily applied to biological complexes but are now gaining ground to characterize transport processes in artificial nanomaterials and nanodevices. In this lecture, I will highlight the enormous potential of 2DES techniques to impact the field of nanosystems, quantum technologies, and quantum devices. The attention will be focused in particular on recent results obtained on semiconductor nanocrystals (*quantum dots*) in solid-state devices and metal-organic hybrid systems.

Invited Talk SYCS 1.3 Tue 11:00 Audimax 1
Exciton Dynamics revealed by Multidimensional Coherent Spectroscopies applied to Light-Harvesting Systems — •Thomas L.C. Jansen — University of Groningen, Zernike Institute for Advanced Materials, Groningen, The Netherlands

Natural light-harvesting systems as found in plants, algae, and especially bacteria are efficient in absorbing photons and transporting their energy to reaction centers, where the energy is converted to chemical energy. Time-resolved multidimensional coherent spectroscopies as Two-Dimensional Electronic Spectroscopy and Fluorescence-Detected Two-Dimensional Electronic Spectroscopy allow the detailed study of the mechanism and dynamics of the underlying light harvesting process. However, these spectra are often challenging to interpret. I will discuss how simulations can be used to distinguish between different processes as energy transport, exciton delocalization, electronic coherence, nuclear coherence, and exciton annihilation. Increasingly detailed models of light-harvesting systems allow increasingly refined understanding of the molecular scale dynamics directing the light-harvesting process. The understanding of what the spectroscopic methods reveal also pave the way for their application in other areas of research on functional nanostructures including organic, inorganic, and hybrid opto-electronic systems. This may aid the design of future artificial light-harvesting systems for photo-voltaic applications or help improving crop yield.

15 min. break.

Invited Talk SYCS 1.4 Tue 11:45 Audimax 1
Revealing couplings with action-based 2D microscopy — •Tobias Brixner — Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Coherent two-dimensional (2D) electronic spectroscopy provides frequency resolution both for the excitation and the probe step. We have developed "action-based" 2D micro-spectroscopy variants using either fluorescence detection (with additional 260 nm spatial resolution) or electron detection (3 nm spatial resolution). In several exemplary applications on nanostructured systems, it will be discussed how various types of quantum-mechanical couplings can be retrieved. First, we investigate a $MoSe_2$ monolayer and observe long-term quantum beating as a function of population time. We analyze the data with a Franck–Condon model and retrieve quantitatively the exciton–phonon coupling strength at room temperature, a quantity previously unknown for 2D materials. Further, we embed a WSe_2 van-der-Waals heterostructure into a microcavity and observe a rich multi-peak structure in 2D spectra which has not been captured in conventional photoluminescence. Simulations reveal hybridized exciton–photon–phonon states, and time-dependent beating signals indicate further fine structure. Lastly, we apply 2D spectroscopy within photoemission electron microscopy (PEEM) on a nanoslit resonator. We find that multiplasmon quantum excitations are required to understand photoemission, going beyond the commonly employed classical linear response model.

Invited Talk SYCS 1.5 Tue 12:15 Audimax 1
Low-frequency phonons affect charge carrier dynamics in hybrid perovskites — •Mischa Bonn — Max Planck Institute for Polymer Research

We study the effect of the gigahertz dielectric response of organic cations and the terahertz phonon response of the inorganic sublattice on the properties, in particular of charge carriers, of hybrid organic-inorganic perovskite materials. We show that electron-phonon coupling is efficient, as evidenced by the timescale of coupling of nascent charge carriers to low-frequency phonon modes occurring on the timescale of the oscillation time of the phonon. We reveal direct coupling between the phonon mode and the bandgap, explaining the anomalous increase of the bandgap of hybrid perovskites with increasing temperature. We also show that there is remarkably efficient coupling between the organic and inorganic sublattices in the perovskite, despite the lack of covalent bonds between the two sublattices.