HL 35: Focus Session: Transient multi-wave mixing on excitonic resonances

Coherent nonlinear optical spectroscopy and, in particular, transient multi-wave mixing processes provide valuable information for basic research in material science and applications in photonics and quantum technology. It allows one to obtain detailed information about the energy structure and dynamic evolution of quantum systems including excited states which is not possible via linear optical spectroscopy and gives direct access to higher-order correlations among the elementary optical excitations. In addition, multi-wave mixing processes can be used as a powerful tool to coherently control the electronic states of the system and to generate non-classical states of light, which is appealing for the implementation in quantum information devices. Importantly for these studies is that excitons possess high oscillator strength which leads to a significant increase of higher-order mixing under resonant excitation with ultrashort optical pulses. This session will focus on multi-wave mixing processes as a unique tool to uncover the fascinating physics of excitons and many-body correlations in emerging materials such as transition metal dichalcogenides and perovskites and establish novel approaches for the coherent control of excitonic quantum systems with classical and quantum light.

Organized by I.A. Akimov and T. Meier

Time: Thursday 9:30–13:30 Location: POT 361

Invited Talk HL 35.1 Thu 9:30 POT 361

Quantum Dynamics of Polarons in Doped Semiconductor Monolayers — •Xiaoqin Elaine Li[1] and Di Huang[2] — [1]Physics Department, University of Texas at Austin, Austin, TX, U.S.A. — [2]Physic Department, TongJi University, Shanghai, China

When mobile impurities are introduced and coupled to a Fermi sea, new quasiparticles known as Fermi polarons are formed. We study Fermi polarons in two dimensional systems, where many questions and debates regarding their nature persist. The model systems we investigate are doped MoSe2 and WSe2 monolayers. In MoSe2, we find the observed attractive and repulsive polaron energy splitting and the quantum dynamics of attractive polarons agree with the predictions of a simple theory. As the doping density increases, the quantum dephasing of the attractive polarons remains constant, indicative of stable quasiparticles, while the repulsive polaron dephasing rate increases nearly quadratically. In WSe2, two distinct species of attractive polarons exist, singlet and triplet polarons. The singlet (triplet) dynamics are mediated by the Fermi seas in the same (opposite) valley. A long-lived valley polarization component is found and likely related to a reservoir of dark states.

Invited Talk HL 35.2 Thu 10:00 POT 361

Impact of phonons on time-resolved optical signals from excitons — •Doris E. Reiter — Condensed Matter Theory, TU Dortmund, 44221 Dortmund

A major difference between optical manipulation of atoms and excitons in semiconductors is the interaction of the latter with the vibrational modes of the solid, i.e., the phonons. In particular in nanostructures like quantum dots, the interaction with acoustic phonons is non-monotonous as function of energy, yielding interesting phenomena like phonon sidebands, reappearance of Rabi rotations or phonon-assisted state preparation. When a quantum dot is optically driven exhibiting Rabi oscillations of the exciton, the pure dephasing-type electron-phonon interaction results in polaron formation accompanied by the emission of phonon wave packets. While for the occupation, the phonon influence leads to an exponential damping, optical multi-wave mixing experiments are more sensitive to the quantum dot polarization. Here, the phonons cannot be described by a simple damping, but their interaction can be understood as a relaxation into the lower dressed state. In this talk, we use a simple model of phonons [1] to discuss their impact on time-resolved optical signals in semiconductor quantum dots. We consider the impact of phonon on continuous wave excitation in a pump-probe configuration [1] and on optical photon echo signals, where the model is in excellent agreement with experiments [2]. Ref: [1] Ann. Phys. 533, 2100086 (2021) [2] PRB 106, 205408 (2022).

Invited Talk HL 35.3 Thu 10:30 POT 361

Hot-Exciton Quantum Dynamics in Zero-Dimensional Structures — •Alfred Leitenstorfer — Department of Physics and Center for Applied Photonics, University of Konstanz, Germany

Electronic quantum processes in intrinsic dimensions of time and space are investigated with femtosecond transient transmission spectroscopy of individual II-VI quantum dots. With our experiments, we are aiming at single-photon amplification for the ultimate control of the quantum statistics of ultrashort light pulses [1]. In these systems, the elementary dynamics of charges and spins is dominated by fundamental aspects such as Coulomb correlations and the Pauli principle [2]. Harnessing a pump-probe microscope optimized for single-electron sensitivity at cryogenic temperatures and high magnetic fields [3], we find extremely asymmetric relaxation characteristics in valence and conduction bands where e.g. femtosecond quantum kinetics of hole-phonon coupling is compatible with persistent spin coherence of hot excitons [4].

[1] F. Sotier et al., Nature Phys. 5, 352 (2009)
[2] C. Hinz et al., Phys. Rev. B97, 045302 (2018)
[3] C. Traum et al., Rev. Sci. Instr. 90, 123003 (2019)
[4] P. Henzler et al., Phys. Rev. Lett. 126, 067402 (2021)

HL 35.4 Thu 11:00 POT 361

Multi-wave mixing applied to explore and control the coherent dynamics of ensembles of semiconductor quantum dots — •Hendrik Rose[1], Stefan Grisard[2], Artur V. Trifonov[2], Rilana Reichhardt[2], Doris E. Reiter[3], Matthias Reichelt[1], Christian Schneider[4,5], Martin Kamp[4], Sven Höfling[4], Manfred Bayer[2], Ilya A. Akimov[2], and Torsten Meier[1] — [1]Paderborn University, Department of Physics & Institute for Photonic Quantum Systems (PhoQS), 33098 Paderborn, Germany — [2]Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany — [3]Condensed Matter Theory, Technische Universität Dortmund, 44221 Dortmund, Germany — [4]Technische Physik, Universität Würzburg, 97074 Würzburg, Germany — [5]Institute of Physics, University of Oldenburg, 26129 Oldenburg, Germany

Multi-wave mixing provides various possibilities to investigate the ultrafast dynamics of semiconductors. Here, we focus on ensembles of quantum dots where the nonlinear dynamics leads to emission in the form of photon echoes. We show that photon echoes can be temporally controlled by the application of multi-wave mixing [1] and furthermore demonstrate that a damping mechanism based on the spatial shape of the applied laser pulses can be circumvented by pulse shaping [2].

[1] A. N. Kosarev, H. Rose, et al., Commun. Phys. **3**, 228 (2020).
[2] S. Grisard, H. Rose, et al., Phys. Rev. B **106**, 205408 (2022).

HL 35.5 Thu 11:15 POT 361

Four-wave mixing at excitonic resonances in the telecom spectral range — •Sebastian Klimmer[1], Artem Sinelnik[1,2], Muhammad Hussain[1], Isabelle Staude[1,2], Habib Rostami[3], and Giancarlo Soavi[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Germany — [2]Institute of Applied Physics, Friedrich Schiller University Jena, Germany — [3]Nordita, KTH Royal Institute of Technology and Stockholm University, Sweden

Over the last few decades, nonlinear optical (NLO) processes have become a pillar for novel photonic devices. In particular, four-wave mixing (FWM) effects can be used to generate quantum optical states of light, such as entangled photons. Transition metal dichalcogenides (TMDs) offer significant advantages compared to conventional nonlinear materials, as their strong light-matter interaction and ease of integration into existing photonic platforms[1] make them perfect for enhancing the performance of integrated NLO devices. In addition, their atomic thickness softens phase matching limits, providing a virtually unlimited bandwidth for FWM[2]. This could be exploited to entangle photons from various spectral regions, making them an ideal source for quantum imaging applications[3]. In this work, we study broadband and exciton enhanced FWM in the telecom spectral range in MoS_2, highlighting the capability of TMDs for integrated photonics and communication applications.

[1] He, J. *et al.*, Nano Lett. **21**, 7, 2709-2718 (2021)
[2] Trovatello, C. *et al.*, Nat. Photonics. **15**, 6-10 (2021)
[3] Gilaberte Basset, M. *et al.*, Laser Photonics Rev. **13**, 10 (2019)

30 min. break

Invited Talk HL 35.6 Thu 12:00 POT 361

Ultrafast dynamics and wave mixing at excitonic resonances in atomically thin semiconductors — •Andreas Knorr, Dominik Christiansen, Florian Katsch, Manuel Katzer, and Malte Selig — Technische Universität Berlin

Atomically thin semiconductors constitute an ideal playground for exciton physics in two dimensions. This involves optically accessible (bright) as well as spin- or momentum-forbidden (dark) excitonic states including intravalley and intervalley excitations. The nonlinear, coherent exciton and wave mixing dynamics induced by short light pulses result from the interplay of strong intrinsic exciton-exciton and exciton-phonon interactions and can be described in a many body Heisenberg equation of motion formalism. Here, we present applications of the theory to:

-exciton-exciton scattering and biexcitons in wave mixing,
-exciton wave function dynamics in time resolved ARPES,
-control of nonlinear excitonic Rabi-oscillations, and
-limits of the boson description of excitons.

Invited Talk HL 35.7 Thu 12:30 POT 361

Spontaneous parametric down-conversion in semiconductor metasurfaces — •Maria Chekhova — Max-Planck Institute for the Science of Light, Erlangen, Germany — Friedrich-Alexander Universität Erlangen-Nürnberg

Spontaneous parametric down-conversion (SPDC) is the most efficient way to generate pairs of entangled photons for quantum photonics applications such as quantum communication, quantum imaging and sensing, and quantum metrology. Today, several groups are trying to implement SPDC on nanoscale quasi-2D platforms, such as subwavelength crystalline layers and metasurfaces. These ultrathin sources have numerous advantages. They are integrable, ultrafast, ultrabroadband and, most importantly, multifunctional. Metasurfaces, in addition, can enhance the rate of photon pair generation due to their *geometric* resonances.

In my talk I will show our recent results on the generation of photon pairs in metasurfaces made of gallium arsenide and gallium phosphide. The metasurfaces are structured to support bound states in the continuum (also known as Fano) resonances, with the quality factors Q reaching several hundred. The resonances enhance the spontaneous emission of pairs by a factor on the order of Q if the frequency of one of the photons coincides with the resonance. Due to the ultrasmall thickness of such sources, SPDC can be pumped slightly above their bandgap without considerable effect of absorption. The inevitably high level of photoluminescence can be overcome by time-resolved registration of photon pairs.

HL 35.8 Thu 13:00 POT 361

Time-resolved four-wave mixing spectroscopy of excitons in a $FA_{0.9}Cs_{0.1}PbI_{2.8}Br_{0.2}$ perovskite crystal — •Stefan Grisard[1], Artur V. Trifonov[1], Dmitry N. Dirin[2,3], Maksym V. Kovalenko[2,3], Ilya A. Akimov[1], Dmitri R. Yakovlev[1], and Manfred Bayer[1] — [1]Experimentelle Physik 2, Technische Universität Dortmund — [2]Institute of Inorganic Chemistry, Department of Chemistry and Applied Bioscience, ETH Zürich — [3]Laboratory for Thin Films and Photovoltaics, Empa-Swiss Federal Laboratories for Materials Science and Technology

Formamidinium lead triiodide ($FAPbI_3$) shows outstanding characteristics for future photovoltaic or light emitting devices, suffers however from poor phase stability. Additives such as cesium and bromine help to improve the phase stability, but possibly increase the localization of the charge carriers. In this work, we present time resolved four-wave mixing spectroscopy as a versatile tool to investigate the coherent optical properties of a $FA_{0.9}Cs_{0.1}PbI_{2.8}Br_{0.2}$ crystal. At low temperatures of 1.5K, the coherent optical response is represented by photon echoes due to the strong inhomogeneous broadening of exciton resonances. We find that the coherence time T_2 is surprisingly long (≈100ps), comparable with the lifetime T_1. The spectral dependence of T_2 and T_1 indicate the importance of carrier localization.

HL 35.9 Thu 13:15 POT 361

Multi-photon pump probe of magnetic-field-induced quantum beats in Cu_2O — •Nikita V. Siverin, Andreas Farenbruch, Dietmar Fröhlich, Dmitri R. Yakovlev und Manfred Bayer — TU Dortmund, Dortmund, Germany

Multi-photon processes such as second harmonic generation are suitable for the investigation of exciton symmetries by analyzing the linear polarization angles of the incoming and the outgoing light. We use difference frequency generation (DFG) with two-photon as the optical technique. The initial laser pulse excites an exciton population by a two-photon excitation process. Probe pulse stimulates a DFG photon. Delaying the second pulse in time enables us the measurement of exciton coherence times and quantum beats between several states. There is also an additional degree of freedom compared to the SHG technique in the polarization of the emission channel. We measure the coherence time of yellow series in Cu_2O. By applying a magnetic field up to 10T in Voigt geometry the 1S orthoexciton splits into three states denoted by the quantum number M = -1,0,1. We observe a beating between these three states. By a selection of the linear polarization setting in the emission channel one can deliberately choose to detect single-frequency beats between the M=+1 and M=-1, triple-frequency beats between all three states or only the M=0 state without beats. Beat frequencies scale with magnetic field strength.