HL 1: Optical Properties I

Time: Monday 9:30–12:45 Location: POT/0006

HL 1.1 Mon 9:30 POT/0006

Critical-Point Analysis of Ta_3N_5 Light Absorbers from Room-Temperature Photo-Modulated Spectroscopy — •Matthias Quintern[1,2], Johannes Dittloff[1,2], Lukas Wolz[1], Gabriel Grötzner[1,2], Johanna Eichhorn[1], and Ian D. Sharp[1,2] — [1]Technical University of Munich, TUM School of Natural Sciences, Department of Physics, 85748 Garching, Germany — [2]Technical University of Munich, Walter Schottky Institute, 85748 Garching, Germany

Tantalum nitride (Ta_3N_5) thin films are promising photoanode materials for solar water splitting. However, the fundamental nature of the Ta_3N_5 bandgap remains under debate. To overcome the limitations of conventional Tauc-plot analysis, we built a new combined photoreflectance (PR) and phototransmittance (PT) setup. Our measurements indicate a direct fundamental bandgap of 2.1 eV, but also reveal higher-energy critical points. Furthermore, we find that the PR and PT spectra closely match temperature-corrected transient absorption spectra. The overall differences in the transmission spectra are minor, suggesting that the samples share a similar bulk electronic structure. At the same time, the more surface-sensitive PR signals exhibit greater variations and even additional sub-gap features in some samples. By performing measurements using different pump wavelengths and power densities, as well as employing a phase analysis, we assign individual features of PR spectra to a surface layer. This has implications for out-of-plane charge transport properties and interfacial energies, both of which are crucial parameters for photoelectrochemical performance.

HL 1.2 Mon 9:45 POT/0006

OptiMate3B: Three-body-information-based prediction of optical spectra for semiconductors and insulators — •Malte Grunert, Max Grossmann, and Erich Runge — Institute of Physics and Institute of Micro- and Nanotechnologies, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Optical spectra of semiconductors and insulators are essential both for many technologically important applications as well as for the understanding of materials. However, their computation via adequate ab initio techniques remains prohibitively expensive for either large-scale studies or many realistic systems. Recently however, machine learning techniques have emerged as promising alternative. We present OptiMate-3B, a line graph attention network capable of predicting the optical spectra of semiconductors and insulators under both the independent particle approximation and the random phase approximation - i.e. incorporating local-field effects - within milliseconds. OptiMate-3B incorporates three-body information to capture angular information in crystalline materials, further enhancing its predictive accuracy. Having been trained on tens of thousands of high-fidelity first-principles spectra, the model generalizes across a broad chemical and structural space. Its open and user-friendly interface facilitates direct integration into high-throughput materials workflows, providing an efficient surrogate for ab initio optical spectroscopy.

HL 1.3 Mon 10:00 POT/0006

Intensity-dependent excitonic second- and third-harmonic generation enhanced by static electric fields — Ruixin Zuo[1], •Matthias Reichelt[1], Cong Ngo[1], Xiaohong Song[2], Weifeng Yang[2,3], and Torsten Meier[1] — [1]Department of Physics and Center for Optoelectronics and Photonics Paderborn (CeOPP), Paderborn University, Warburger Straße 100, D-33098 Paderborn, Germany — [2]School of Physics and Optoelectronic Engineering, Hainan University, Haikou 570288, China — [3]Center for Theoretical Physics, Hainan University, Haikou 570288, China

The many-body Coulomb interaction plays a decisive role in the ultrafast photoinduced carrier dynamics governing semiconductor high-harmonic generation, see, e.g., [1,2]. Moreover, static electric fields were shown to surprisingly enhance excitonic second- and third-harmonic generation [3]. Here, we study this enhancement for material models corresponding to transition-metal dichalcogenides and study the dependencies on several parameters.

[1] A. Trautmann, R. Zuo, G. Wang, W.-R. Hannes, S. Yang, L. H. Thong, C. Ngo, J. T. Steiner, M. Ciappina, M. Reichelt, H. T. Duc, X. Song, W. Yang, and T. Meier, Proc. SPIE **11999**, 1199909 (2022).
[2] J. Hader, J. Neuhaus, J. V. Moloney, and S. W. Koch, Opt. Lett. **48**, 2094 (2023).
[3] R. Zuo, M. Reichelt, C. Ngo, X. Song, W. Yang, and T. Meier, arXiv:2511.05112.

HL 1.4 Mon 10:15 POT/0006

Thin Films of Size-Selected Cu_2ZnSnS_4 and Cu_2NiSnS_4 Nanocrystals: Structural and Optical Properties — •Oleksandra Ivakhno-Tsehelnyk[1], Serhiy Kondratenko[2], Volodymyr Dzhagan[3], and Dietrich R.T. Zahn[1] — [1]Semiconductor Physics & Research Center for Materials, Architectures and Integration of Nanomembranes (MAIN), Chemnitz University of Technology. — [2]Taras Shevchenko National University of Kyiv, 01601 Kyiv, Ukraine. — [3]Lashkaryov Institute of Semiconductor Physics, NAS of Ukraine, 03038 Kyiv, Ukraine.

An investigation was conducted into thin films of Cu_2NiSnS_4 (CNTS) and Cu_2ZnSnS_4 (CZTS) nanocrystals (NCs) in the context of their promising use for photovoltaics (PV). Colloidal NC ensembles were obtained by "green" colloidal synthesis and directly subjected to size-selective centrifugation, yielding fractions of different sizes. This approach provides a relatively non-toxic and scalable method of producing light-absorbing NC materials. The thin films were prepared by spin-coating the NC fractions onto various substrates. A range of analytical techniques were employed to investigate their properties, encompassing Raman spectroscopy, atomic force microscopy (AFM), X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and spectroscopic ellipsometry. These techniques enabled the investigation of the relationship between the crystal structure, composition, morphology, and optical properties with the size of CZTS and CNTS nanocrystals.

HL 1.5 Mon 10:30 POT/0006

Simulations of the nonlinear optical dynamics of quantum dot ensembles in a microcavity — •Mohit Kumar[1,2], Hendrik Rose[2], and Torsten Meier[1,2] — [1]Department of Physics and Center for Optoelectronics and Photonics Paderborn (CeOPP), Paderborn University, Warburger Strasse 100, D-33098 Paderborn, Germany — [2]Institute for Photonic Quantum Systems (PhoQS), Paderborn University, Warburger Strasse 100, D-33098 Paderborn, Germany

Recently, it has been demonstrated that photon echoes emitted from ensembles of inhomogeneously-broadened quantum dots can be controlled in several ways by suitably designed light pulses [1,2]. The strong coupling of a single two-level system to the mode of a microcavity results in a complex nonlinear optical dynamics [3]. Here, we present numerical simulations of the coherent dynamics of quantum dots ensembles, described as two-level systems, inside a single-mode microcavity. The coupling to the field mode results in a complex dynamics which is studied numerically as function of the relevant system parameters and for different excitation conditions.

[1] A. N. Kosarev, H. Rose, S. V. Poltavtsev, M. Reichelt, C. Schneider, M. Kamp, S. Höfling, M. Bayer, T. Meier, and I. A. Akimov, Comm. Physics **3**, 228 (2020).
[2] S. Grisard, A. V. Trifonov, H. Rose, R. Reichhardt, M. Reichelt, C. Schneider, M. Kamp, S. Höfling, M. Bayer, T. Meier, and I. A. Akimov, ACS Photonics **10**, 3161 (2023).
[3] M. Koch, J. Shah, and T. Meier, Phys. Rev. B **57**, R2049(R) (1998).

HL 1.6 Mon 10:45 POT/0006

Optical Limiting Induced by Excited-State Absorption in Thiophene Oligomers — •Mustapha Driouech, Michele Guerrini, and Caterina Cocchi — Friedrich-Schiller Universität Jena, Institute for Condensed Matter Theory and Optics, 07743 Jena, Germany

Optical limiting (OL) is a nonlinear effect that reduces light transmission at high intensities, providing protection for sensitive optical devices and human eyes. Non-perturbative ab initio methods have proven to be powerful predictive tools for OL [1]. In this work, we investigate the optical absorption spectra of a set of thiophene oligomers under the influence of a broadband, instantaneous electric field [2]. At sufficiently high intensities, absorption peaks that are absent in the linear response emerge below the absorption onset in the linear regime. To elucidate the optical nonlinearities involved, we perform pump-probe

simulations and analyze the population dynamics, revealing that these emergent features originate from excited-state absorption in the near-infrared to visible region. Further analysis, taking into account both molecular length and applied laser intensity, demonstrates how these factors influence the nonlinear response. Our results show that this fully ab initio approach can accurately capture and unravel the fundamental processes driving optical nonlinearities in thiophene oligomers.

[1] C. Cocchi, et al., Phys. Rev. Lett. 112, 198303 (2014).
[2] M. Driouech, et al., J. Phys. Chem. Lett 16, 11867 (2025).

15 min. break

HL 1.7 Mon 11:15 POT/0006

Polarization-resolved Raman measurements on optically anisotropic crystals — •Susanne Moritz, Lukas Trefflich, Ron Hildebrandt, Marius Grundmann, and Chris Sturm — Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Germany

Raman spectroscopy provides a powerful, non-destructive way to probe lattice vibrations, crystal symmetry, and electron-phonon interactions. For years, optically anisotropic materials have been gaining importance in solid-state physics research. These materials are investigated by measuring the Raman intensity in two configurations, namely cross-polarized and parallel polarization. However, due to their low symmetry, this is not sufficient in order to extract the properties of the Raman tensor and extensive polarization-resolved Raman measurements are required for a precise determination, particularly of the Raman tensor [1,2,3]. Here, we present an efficient measurement scheme in order to determine the Raman tensor of optically anisotropic materials with high precision. Special care must be taken regarding the influence of the used polarizing optics, such as half-wave plates, as well as the impact of the non-idealities of all optical elements included in the setup, such as mirrors or beam splitters. For this purpose, we also show a procedure for calibrating these elements.

[1] C. Kranert et al., Phys. Rev. Lett. **116**, 127401 (2016)
[2] C. Kranert et al., Sci. Rep. **6**, 35964 (2016)
[3] R. Hildebrandt et al., Appl. Phys. Lett. **119**, 121109 (2021)

HL 1.8 Mon 11:30 POT/0006

Quadrupole Transitions in Molecules from Real-Time Time-Dependent Density Function Theory — •Anvar Khujakulov[1], Michele Michele Guerrini[1], Carlo Andrea Rozzi[1], and Caterina Cocchi[2] — [1]Institute for Condensed Matter Theory and Optics, 07743 Jena, Germany — [2]Istituto Nanoscienze - 41125 Modena, Italy

The breakdown of the dipole approximation in tightly focused or short-wavelength laser fields is driven by spatial electric field variations, leading to significant beyond-dipole effects including quadrupole transitions and a nonlinear optical response [1]. We extend the Yabana-Bertsch implementation of real-time time-dependent density functional theory to include quadrupole transitions in the absorption cross-sections and apply the developed approach to atoms and small molecules. Our results reveal absorption peaks appearing above the linear absorption onset, a clear signature of quadrupole transitions. Critically, for molecules with appropriate symmetry, the quadrupole contribution to the absorption cross-section is maximized when a specific molecular axis aligns with the electric field gradient, analogous to the dipole case. By systematically rotating the molecular axis relative to the laser polarization vector, we calculate the orientation-dependent absorption cross-section and differentiate the contributions from dipole and quadrupole transitions. We quantitatively demonstrate that quadrupole transition is not a minor effect but a significant contributor to absorption spectra beyond the dipole approximation.

[1] C. Cocchi, et al., Phys. Rev. Lett. 112, 198303 (2014).

HL 1.9 Mon 11:45 POT/0006

Phase-matched electron-photon interactions enabled by 3D printed helical waveguides — •Masoud Taleb[1], Mohsen Samadi[2], and Nahid Talebi[1] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Department of Electrical and Information Engineering, Kiel University, 24143 Kiel, Germany

We demonstrate a new phase-matching mechanism between free electrons and guided optical modes using 3D-printed helical waveguides. The structure consists of a polymer micro-fiber coated with a thin gold layer, fabricated by two-photon polymerization. When an electron beam propagates parallel to the helix axis, the helical geometry extends the optical path and enables sequential, phase-synchronous coupling to a plasmonic hybrid mode. This interaction unifies elements of Smith-Purcell and Cherenkov emission and yields a strong, highly directional visible-light output. Cathodoluminescence measurements reveal a dominant emission near 2.1-2.2 eV, collimated along the electron trajectory and exhibiting circular polarization dictated by the helix handedness. The observed photon yield (~0.02 photons per electron for an 11-turn helix) exceeds that of planar electron-driven photon sources by more than two orders of magnitude. These results introduce helical micro-waveguides as an efficient platform for directional, polarization-controlled electron-photon interactions.

HL 1.10 Mon 12:00 POT/0006

Quantum-engineered CaAlSiN3 via terbium substitution: coupling spin, optical, and lattice responses in a rare-earth nitride host — •Sikander Azam and Jan Minar — University of West Bohemia New Technologies - Research Centre Univerzitní 8 301 00 Plzeň Czech Republic

Rare-earth nitrides attract interest because they link optical activity, magnetism, and mechanical stability. CaAlSiN3 is a robust wide-gap host, but the microscopic role of terbium substitution is not well established. We study pristine and Tb-doped CaAlSiN3 (8.5 and 17 percent) using spin-polarized density functional theory within GGA+U and spin-orbit coupling in WIEN2k. Tb narrows the bandgap and introduces 4f-derived mid-gap states that enhance visible-range absorption. The densities of states show hybridization between Tb 4f levels and the surrounding network. The optical response shows higher absorption, a larger refractive index, and shifts in dielectric features that follow these electronic changes. Simulated Tb L*-edge X-ray absorption spectra display white-line shifts and peak broadening with increasing Tb content, consistent with the modified unoccupied 4f states. Tb carries a strong local moment, raising the total moment to about 11.8 μB at higher doping. All compositions remain mechanically stable, and bulk and shear moduli increase moderately. The piezoelectric response also strengthens, with d33 and e33 roughly doubling compared to the undoped host. These results show that Tb substitution tunes the electronic, magnetic, optical, and mechanical behaviour of CaAlSiN*, supporting its potential in photonic and spin-related applications.

HL 1.11 Mon 12:15 POT/0006

Accurate computation of Berry connection purely limited by basis set completeness — Martin Thümmler[1], •Alexander Croy[1], Thomas Lettau[2], Ulf Peschel[2,3], and Stefanie Gräfe[1,3,4] — [1]Institute of Physical Chemistry, Friedrich Schiller University Jena — [2]Institute of Condensed Matter Theory and Optics, Friedrich Schiller University Jena — [3]Abbe School of Photonics, Friedrich Schiller University Jena — [4]Fraunhofer Institute for Applied Optics and Precision Engeneering Jena

Wannierization provides a reliable framework for interpolating operator matrix elements in the Brillouin zone. This Fourier-type interpolation of the matrix elements of **k**-local operators, e.g., the Hamiltonian, is well understood. However, the Berry connection is computed from the overlap matrices of the cell periodic parts of the Bloch functions with different **k** and hence non-local. So far, all proposed interpolation schemes do not account for the matrix structure of the overlap matrices, but treat the matrix elements as independent when computing the Berry connection. In this work, we propose a consistent interpolation scheme based on the matrix logarithm. Its accuracy is only limited by the basis set incompleteness of the considered bands in the ab-initio calculation. The quality of the new interpolation scheme is demonstrated by an improved agreement between the velocity and momentum operator over the full Brillouin zone. As an application, we demonstrate how this refinement impacts the predicted linear and non-linear optical properties of semiconductors.

HL 1.12 Mon 12:30 POT/0006

Focused Ion Beam (FIB) patterned Microcavities for Topological Lasers — •Lorenz Löffler[1,4], Dominik Horneber[1,4], Jakob Lindenthal[2,4], Johannes Düreth[1,4], Markas Sudzius[2,4], Monika Emmerling[1], Martin Kamp[3], Simon Betzold[1,4], Johannes Benduhn[2,4], Sven Höfling[1,4], Karl Leo[2,4], and Sebastian Klembt[1,4] — [1]University of Würzburg, Physikalisches Institut, Lehrstuhl für Technische Physik — [2]Dresden Integrated Center for Applied Physics and Photonic Materials (IAPP), and Institute of Applied Physics, Technische Universität Dresden — [3]University of Würzburg, Physikalisches Institut and Wilhelm-Conrad-Röntgen-Research Center for Complex Material Systems — [4]Würzburg-Dresden Cluster of Excellence ct.qmat

Topological lasers can be realized by coupling individual photonic resonators in topologically non-trivial lattice geometries. Optical microcavities have been found to be a promising platform for studying topological laser interfaces with long-range coherence. The fabrication of such structures typically involves etching of the microcavity and through the emitter layer. In our work, we create the photonic potential by FIB milling of a glass substrate prior to depositing dielectric Bragg reflectors. This technique allows the potential to be engineered while keeping the emitter layer intact. Here, the emitter is either semiconductor quantum wells or the organic semiconductor Alq_3 doped with DCM dye. By exploring modified lattice geometries that enhance the interface-to-bulk ratio as well as temperature-dependent studies, we aim for the next step towards practical topological lasers.