HL 61: Focus Session: Tailored Nonlinear Photonics I

The research field of nonlinear photonics is driven by the tailoring and control of nonlinear light-matter interactions and by the application of nonlinear concepts for advanced light management. Current research activities are driven by concepts from quantum optics, coherent optics, and solid-state physics. The progress in the field strongly benefits from advanced solid-state materials, nanostructures, and photonic structures, as well as from extremely intense and efficient ps and fs laser sources. The application of new concepts paves technically viable routes towards advanced nonlinear photonic devices, which are indispensable for the implementation of efficient frequency conversion, conditional photonic functionalities, and photonic quantum technologies.

Organizers: Artur Zrenner (Universität Paderborn), Thomas Zentgraf (Universität Paderborn) and Manfred Bayer (TU Dortmund)

Time: Thursday 9:30–13:00 Location: POT 51

Invited Talk HL 61.1 Thu 9:30 POT 51

Supercontinuum second-harmonic generation spectroscopy of 2D semiconductors — •Steffen Michaelis de Vasconcellos — Institute of Physics and Center for Nanotechnology, University of Münster, Germany

Two-dimensional semiconductors such as MoS_2 or WS_2 and their heterostructures have recently emerged as promising material systems for novel optoelectronic devices due to their unique electrical, mechanical, and optical properties. Several of these atomically thin materials are particularly suited for nonlinear light-matter interactions. Their strong optical nonlinearity, broadband and tunable optical absorption, and ultrafast response have been successfully employed in all-optical modulators, lasers, wavelength converters, and optical limiters. The prototypical nonlinear process second-harmonic generation is a powerful tool to gain insight into nanoscale materials, because of its dependence on the crystal symmetry and electronic structure. We have developed a new method to perform ultra-broadband SHG spectroscopy, which provides access to the frequency-dependent nonlinear susceptibility $\chi^{(2)}$ of atomically thin materials and allows for the identification of excitonic resonances.

Invited Talk HL 61.2 Thu 10:00 POT 51

Quasi-instantaneous switch-off of deep-strong light-matter coupling — •Christoph Lange[1], Maike Halbhuber[1], Joshua Mornhinweg[1], Viola Zeller[1], Cristiano Ciuti[2], Dominique Bougeard[1], and Rupert Huber[1] — [1]University of Regensburg, Germany — [2]Université de Paris, France

Optical microresonators facilitate custom-tailored quantum states of matter by dressing electronic excitations with virtual cavity photons. Once the rate of energy exchange between light and matter modes exceeds the carrier frequency of light itself, 'deep-strong coupling' emerges, and the vacuum ground state is profoundly modified, giving rise to novel phenomena including cavity-mediated superconductivity and other phase transitions. While the exploration of the equilibrium properties of deep-strong coupling has just started, yet more unusual quantum dynamics are expected on subcycle scales. Here, we explore the intriguing dynamics that arises when deep-strong light-matter coupling is switched off quasi-instantly. The experiments employ custom-tailored THz nanoresonators coupled to cyclotron resonances of two-dimensional electron gases. Femtosecond photoexcitation of an integrated switch element extinguishes the fundamental cavity mode and decouples it from the cyclotron resonance, more than an order of magnitude faster than the oscillation cycle of light. The quasi-instantaneous extinction of the polariton states is hallmarked by sub-polariton-cycle oscillations of the transmission as confirmed by a quantum model.

HL 61.3 Thu 10:30 POT 51

Nonlinear optical susceptibility of potassium titanyl phosphate — •Agnieszka Kozub, Sergej Neufeld, Adriana Bocchini, Uwe Gerstmann, Arno Schindlmayr, and Wolf Gero Schmidt — Department Physik, Universität Paderborn, Paderborn, Germany

$KTiOPO_4$ (KTP) is a very important nonlinear optical material. Due to its high conversion efficiency, non-hygroscopicity, and cost-efficiency it is widely used for second harmonic generation (SHG). However, only very few experimental measurements on its SHG efficiency are available. In particular, its photon energy dependence has not been systematically studied. Previous theoretical works [1-3] focus mainly on the linear optical properties of KTP. Our present work aims at closing this gap.

Based on the Berry-phase formulation of the dynamical polarization [4] the second-order nonlinear optical susceptibility tensor $\chi^{(2)}$ is calculated and compared with the available experimental data. Particular attention is paid to the influence of non-stoichiometry and point defects on the KTP optical response.

[1] A. H. Reshak, I. V. Kityk, and S. Auluck, J. Phys. Chem. B 114, 16705 (2010).
[2] M. Ghoohestani, A. Arab, S. J. Hashemifar, and H. Sadeghi, J. Appl. Phys 123, 015702 (2018).
[3] S. Neufeld, A. Bocchini, U. Gerstmann, A. Schindlmayr, and W. G. Schmidt, J. Phys. Mater. 2, 045003 (2019).
[4] C. Attaccalite, M. Grüning, Phys. Rev. B 88, 235113(2013).

HL 61.4 Thu 10:45 POT 51

Systematic Investigation of the Nondegenerate Two-Photon Absorption Coefficient in ZnSe — •Laura Krauss-Kodytek, Claudia Ruppert, and Markus Betz — Experimentelle Physik 2, Technische Universität Dortmund, Otto-Hahn-Straße 4a, 44227 Dortmund, Germany

The two-photon absorption (TPA) coefficient β_{TPA} is strongly enhanced in case of extremely nondegenerate photon pairs ($\hbar\omega_1 \neq \hbar\omega_2$) when compared to the degenerate case ($\hbar\omega_1 = \hbar\omega_2$) [1]. However, a systematic study of the TPA strength as a function of the involved photon pair's energy ratio ω_1/ω_2 remains elusive.

To investigate this dependence we use an optical parametrical amplifier which provides photon pairs within energy ratios from unity to 3.5. The photon pair's sum energy of 3.1 eV slightly exceeds the bandgap of ZnSe ($E_g = 2.7$ eV at room temperature). The ZnSe samples have a thickness of 40 μm and a (100)- or (110)-orientation.

The setup is designed in a pump-probe fashion and the peak signals at time overlap reflect the TPA strength β_{TPA}. We observe an enormous increase of the nondegenerate TPA strength compared to the degenerate one. Further, the measured trend of the β_{TPA} as a function of the energy ratio is well in line with the theoretical predictions [2].

[1] D. Fishman et al., Nature Photonics **5**, 561-565 (2011)
[2] W.-R. Hannes and T. Meier, Physical Review B **99**, 125301 (2019)

30 min. break

Invited Talk HL 61.5 Thu 11:30 POT 51

Watching plasmonic skyrmions spin using ultrafast two-photon photoelectron spectroscopy — •Harald Giessen[1], Tim Davis[1,2], Bettina Frank[1], Pascal Dreher[3], David Janoschka[3], and Frank Meyer zu Heringdorf[3] — [1]4th Physics Institute and Research Center SCoPE, University of Stuttgart — [2]University of Melbourne — [3]CENIDE, University of Duisburg Essen

Plasmonic skyrmions are topological defects in the electromagnetic near-field on thin metal films, recently observed using scanning near-field optical microscopy. However, only one spatial component of the electric field was measured and one of the most intriguing features of skyrmions, namely their dynamics, was not assessed. Here we introduce a new technique, time-resolved vector microscopy, that enables us to compose entire movies on a sub-femtosecond time scale and a 10 nm spatial scale of the electric field vectors of surface plasmon polaritons. Specifically, we image complete time sequences of propagating surface plasmons as well as plasmonic skyrmions on atomically flat single crystalline gold films that have been patterned using gold ion beam lithography. This allows us to unambiguously resolve all vector components of the electric field as well as their time dynamics, enabling the retrieval of the experimental time-dependent skyrmion number, and indicating the periodic transformation from skyrmions to anti-skyrmions and back.

HL 61.6 Thu 12:00 POT 51

Quasiparticle and excitonic effects in the lithium niobate polaron optical absorption — •Falko Schmidt, Wolf Gero Schmidt, Arno Schindlmayr, and Uwe Gerstmann — Department Physik, Universität Paderborn, 33095 Paderborn, Germany

Lithium niobate ($LiNbO_3$), a perovskite-structure ferroelectric, is widely employed in nonlinear optical applications. The material employed in technical applications is usually grown from a congruent melt and therefore characterized by a high concentration of intrinsic defects which influence its optical properties. The latter are usually interpreted within the polaron concept [1]. Polarons tend to localize at crystal defects, especially at positively charged impurities, due to the attractive Coulomb potential and the additional lattice distortion. In the present study we perform many-body perturbation theory calculations in order to determine the influence of electron quasiparticle and electron-hole attraction effects on the polaron optical absorption. By means of comparison between simulated spectra and measured data [1] we are able to propose detailed atomic structures giving rise to both polarons and bipolarons.

[1] OF Schirmer, M Imlau, C Merschjann, B Schoke, J. Phys.: Condens. Matter **21**, 123201 (2009).

HL 61.7 Thu 12:15 POT 51

PSF-analysis of ferroelectric domain-walls in waveguide structures — •Peter Mackwitz, Laura Padberg, Christof Eigner, Christine Silberhorn, Gerhard Berth, and Artur Zrenner — Universität Paderborn, Warburger Straße 100, 33098 Paderborn, Germany

The occurring nonlinear contrast mechanism of periodically poled waveguide structures in $LiNbO_3$ and $KTiOPO_4$ is examined via a combined experimental and numerical approach. The numerical calculation is based on a vectorial model, whereby the experimental access is realized by a direct imaging technique which records the spatial distribution of the second-harmonic signal in the back focal plane. As the model depends on the optical properties of the material, different scenarios of contrast mechanisms can be simulated and compared to the experimental data. It turns out that many features of the nonlinear signatures arise from destructive interference of phase-shifted wavelets at the domain walls as well as the waveguide boundaries. The phase is acquired due to the interaction of the focus with differentially poled domains, whose susceptibility tensor appears rotated by 180°. Another ingredient for the contrast are new tensor elements which do only occur at the domain walls. With this work a comprehensive explanation for the contrast mechanism in the coherent surface near regime is given.

HL 61.8 Thu 12:30 POT 51

All-optical switching of exciton polariton vortices — Xuekai Ma[1], •Bernd Berger[2], Marc Assmann[2], Rodislav Driben[1], Torsten Meier[1], Christian Schneider[3], Sven Höfling[3,4], and Stefan Schumacher[1,5] — [1]Department of Physics and Center for Optoelectronics and Photonics Paderborn (CeOPP), Universität Paderborn, Warburger Strasse 100, 33098 Paderborn, Germany — [2]Experimentelle Physik 2, Technische Universität Dortmund, 44227 Dortmund, Germany — [3]Technische Physik, Universität Würzburg, Am Hubland, 97074, Würzburg, Germany — [4]SUPA, School of Physics and Astronomy, University of St. Andrews, St. Andrews KY16 9SS, United Kingdom — [5]College of Optical Sciences, University of Arizona, Tucson, AZ 85721, USA

Vortices are elementary excitations in exciton polariton condensates that consist of a $2\pi \cdot m$ radial phase shift of the polariton wavefunction, where m is the topological charge of the vortex. This topological charge translates into the orbital angular momentum (OAM) state of the emitted light field, which we detect by application of a dedicated OAM spectroscopy technique. This technique gives us the opportunity to study the temporal dynamics of vortices in exciton polariton condensates without using complex interferometric techniques that also always require a phase reference. Here, we theoretically and experimentally demonstrate the switching of the charge of a localized vortex in a microcavity polariton condensate.

HL 61.9 Thu 12:45 POT 51

Semiclassical theory of multi-photon absorption in bulk zincblende type semiconductors — •Wolf-Rüdiger Hannes and Torsten Meier — Department of Physics and CeOPP, University of Paderborn, Warburger Str. 100, D-33098 Paderborn, Germany

Multi-photon absorption coefficients of zinc-blende type semiconductors are studied in the independent-electron approximation. Our theory is based on a gauge-invariant form of the semiconductor Bloch equations including inter- and intraband excitations. Extending our initial analysis [1,2] we use a GaAs bandstructure and complex dipole moments obtained from k·p theory. Considering a pump-probe scheme, the nonlinear-optical response is analyzed for varying excitation parameters, such as pulse frequencies, propagation directions, and polarization angles. Analytical solutions are obtained for near half-gap excitation in the cw-limit using a simplified multiband model valid in the vicinity of the Γ-point. We also investigate multi-photon absorption in a non-perturbative regime, where it shows Rabi-like oscillations as a function of the light intensity.

[1] W.-R. Hannes and T. Meier, Phys. Rev. B **99**, 125301 (2019).
[2] W.-R. Hannes, L. Krauß-Kodytek, C. Ruppert, M. Betz, and T. Meier, Proceedings of the SPIE **10916**, 109160O (2019).