O 31: Plasmonics & Nanooptics II: SHG and Dielectric Properties (joint session O/CPP)

Time: Tuesday 14:00–16:30 Location: H8

O 31.1 Tue 14:00 H8

Sum-Frequency Generation Enhanced by Localized Surface Phonon Polaritons — •Riko Kiessling[1], Yujin Tong[1], Alexander J. Giles[2], Joshua D. Caldwell[2,3], Martin Wolf[1], and Alexander Paarmann[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]U.S. Naval Research Laboratory, Washington, D.C., USA — [3]Vanderbilt University, Nashville, USA

Nanophotonic devices open the way to tailor light-matter interaction on the nano-scale by specifically designed structures of solid-state material. This is advantageous for, e.g., the sub-diffractional localization of light, which fosters amplification of nonlinear optical frequency conversion processes by enhancement of local electric fields.

Here, we employ localized surface phonon polariton modes in nanostructured polar dielectric media [1] to resonantly increase the nonlinear response. In contrast to surface plasmons, the optical phonon-based approach exhibits significantly lower optical losses. By means of IR-VIS sum-frequency generation spectroscopy [2], a strong modulation of the local electromagnetic fields, and thus, amplified upconversion radiation, is observed. Intensity and spectral position of the nonlinear emission can be manipulated by the nanostructure design. In this way, sub-wavelength sized architectures prove to be an efficient method for the control of localized fields associated with three-dimensionally confined optical modes in the mid-infrared spectral region.

[1] Razdolski et al., Nano Lett. 16, 6954 (2016)
[2] Kiessling et al., Phys. Rev. Accel. Beams 21, 080702 (2018)

O 31.2 Tue 14:15 H8

Nonlinear Response of Grating-Coupled Surface Phonon Polaritons — Marcel Kohlmann, Nikolai Christian Passler, Martin Wolf, and •Alexander Paarmann — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

Surface Phonon Polaritons (SPhPs) have recently attracted much attention in infrared nanophotonics, due to their low losses compared to plasmon polaritons. Specifically for nonlinear optics, SPhPs result in tremendous enhancement of local optical fields [1,2]. Here, we study the nonlinear response of grating-coupled SPhPs [3] in by means of second-harmonic generation (SHG), in sub-wavelength SiC grating geometries. Non-specular emission of SHG is observed for resonant SPhP excitation, demonstrating the peculiarities of nonlinear scattering for grating-coupled nonlinear nanophotonics [4].

[1] Razdolski et al., Nano Letters 16, 6954 (2016).
[2] Passler et al., ACS Photonics 4, 1048 (2017).
[3] Greffet et al., Nature 416, 61 (2002).
[4] Quail and Simon, J. Opt. Soc. Am. B 5, 325 (1988).

O 31.3 Tue 14:30 H8

Wavelength-dependent Third Harmonic Generation in Metallic Thin Films — •Vincent Drechsler, Joachim Krauth, Mario Hentschel, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

Harmonic generation in plasmonic systems has gained significant interest in the last years. While the plasmonic near-field enhancement has been studied in great detail, little attention was paid to the origin of the nonlinear signals which in fact lie in the microscopic nonlinearity of the involved plasmonic metals. In order to predict wavelength-dependent nonlinear processes it is therefore crucial to understand these microscopic nonlinearities. Utilizing an optical parametric oscillator as a tunable broadband light source, we study wavelength-dependent third harmonic generation from metallic thin films made of Gold, Copper, and Magnesium. We find that the linear properties of the metallic films, that is their absorption, strongly influence the third harmonic generation efficiency. Optical transitions between the different bands lead to a resonant enhancement of the third order susceptibilities. Utilizing hydrogen to switch metallic magnesium to dielectric magnesium hydride we can tune the nonlinearity of thin films and observe the metallic-dielectric phase transition in the linear and nonlinear regime. We find complex relations between the linear transmittance and the radiated third harmonic, which provide new insights into the phase transition.

O 31.4 Tue 14:45 H8

Plasmonic analogue of Electromagnetically Induced Absorption Boosts Third Harmonic Generation — •Joachim Krauth, Mario Hentschel, Bernd Metzger, and Harald Giessen — 4th Physics Institute and Research Center SCoPE, University of Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

In three-dimensional nanostructures, the appropriate choice of geometry and materials allows for tailoring the linear optical properties in a vast range. In the so-called plasmonic dolmen structure the coupling between a bright and a dark state can be manipulated to realize the plasmonic analogue of electromagnetically induced absorption (EIA), resulting in an enhanced absorbance of the coupled system. In analogy to solid state nonlinear optics, in which the linear absorption spectra largely determine the nonlinear interaction, plasmonic EIA is therefore expected to boost nonlinear conversion efficiencies. We are reporting third harmonic generation spectroscopy results on a plasmonic EIA system. The radiated third harmonic signals can be understood when taking the intricate phase relation and coupling strength dependence of EIA into account. An appropriately modified anharmonic oscillator model based on the linear absorption spectra can describe the nonlinear response of our three-dimensional plasmonic structure. The design freedom associated with three-dimensional nanostructuring can provide further insight into the nonlinear response of complex plasmonic systems.

O 31.5 Tue 15:00 H8

Symmetry-forbidden second-harmonic generation in a fully centro-symmetric plasmonic nanocircuit — •Julian Obermeier[1], Tzu-Yu Chen[2], Thorsten Schumacher[1], Fan-Cheng Lin[2], Jer-Shing Huang[3,4,5], Chen-Bin Huang[2,5], and Markus Lippitz[1] — [1]University of Bayreuth, Germany — [2]National Tsing Hua University, Taiwan — [3]Leibniz Institute of Photonic Technology, Germany — [4]National Chiao Tung University, Taiwan — [5]Research Center for Applied Sciences, Taiwan

Nonlinear optical frequency conversion on the nanoscale remains a challenge, but may pave the path towards multi-functional optical circuits. Surface plasmon fields are highly spatially confined near the metal/dielectric interface and exhibit giant field enhancement. These two attributes are invaluable for nonlinear optics with plasmons. However, second-harmonic generation in bulk is forbidden in centro-symmetric materials such as typical noble metals. Past efforts concentrated on the broken symmetry at the surface in combination with an asymmetric shape of the particle. We introduce a new way of breaking the symmetry by a propagating mode of a plasmonic waveguide, a two-wire transmission-line. We demonstrate that an optical mode of correct symmetry is sufficient to allow SHG even in centro-symmetric structures made of centro-symmetric material. This is a new degree of freedom for on-chip nonlinear signal processing in nanophotonics.

O 31.6 Tue 15:15 H8

Resonant Field Enhancement of Epsilon Near Zero Berreman Modes in an Ultrathin AlN Film — •Nikolai Christian Passler[1], Ilya Razdolski[1], Joshua D. Caldwell[2], Martin Wolf[1], and Alexander Paarmann[1] — [1]Fritz-Haber-Institute of the MPG, Berlin, Germany — [2]Vanderbilt University, Nashville, USA

In nanophotonics, the strongly enhanced local optical fields of polaritonic modes are the driving force of nonlinear optical phenomena. In sub-wavelength thin polar dielectric films, a polaritonic Berreman mode arises at the longitudinal optical phonon frequency where the dielectric permittivity crosses zero [1]. Complementary to this radiative epsilon near zero (ENZ) mode, also an evanescent ENZ polariton is supported at large in-plane momenta [2], exhibiting similar properties. At resonance, the Berreman mode features an immense field enhancement, enabling to boost the efficiency of nonlinear nanophotonic applications like all-optical ultrafast switching.

Employing a mid-infrared free-electron laser, we investigate the second harmonic generation (SHG) of an ultrathin AlN layer on top of bulk SiC. The employed SHG spectroscopy [3] serves as a direct experimental probe of the tremendous field enhancement of the high-quality, strongly confined ENZ Berreman mode. With these properties, the AlN thin-film Berreman mode offers an appealing platform for the development of novel infrared nanophotonic devices.

[1] Vassant et al., Optics Express 20, 23971 (2012)
[2] Passler et al., Nano Letters 18, 4285 (2018)
[3] Paarmann et al., Applied Physics Letters 107, 081101 (2015)

O 31.7 Tue 15:30 H8

Enhanced second harmonic emission from zinc oxide nanoparticles infiltrated into the pores of gold nanosponges — •Jue-Min Yi[1], Dong Wang[2], Felix Schwarz[3], Jinhui Zhong[1], Abbas Chimeh[1], Anke Korte[1], Peter Schaaf[2], Erich Runge[3], and Christoph Lienau[1] — [1]Carl von Ossietzky Universität, 26129 Oldenburg — [2]TU Ilmenau,98693 Ilmenau. — [3]TU Ilmenau, 98693 Ilmenau

We introduce zinc-oxide (ZnO)-functionalized porous gold (Au) nanoparticles which exhibit strong second harmonic (SH) generation around 400 nm by coupling surface plasmons (SPs) to ZnO excitons. The hybrid nanosystem consists of a thin layer (~10 nm) of ZnO incorporated into individual porous Au nanoparticles percolated with a three-dimensional (3D) network of 10-nm sized ligaments acting as nanoantennas and nanocavities.We have utilized a broadband and few-cycle ultrafast laser to generate coherent nonlinear emission from individual bare nanosponges and from ZnO-functionalized sponges. The third harmonic (TH) emission spectrum of Au/ZnO hybrid particles reveals a distinct red shift with respect to pure Au sponges. In contrast, a substantial broadening of the SH spectra and a distinct blue-shift is seen in the ZnO-functionalized nanosponges. It is demonstrated that SH emission around 400 nm, close to the ZnO band gap, is 15 times stronger with respect to bare Au nanosponges. Such enhanced SHG emission is attributed to plasmon-enhanced two-photon excitation of ZnO excitons.

O 31.8 Tue 15:45 H8

Revealing Plasmon-Exciton Coupling for SHG Enhancement by Interferometric Frequency Resolved Autocorrelation — •Jinhui Zhong[1], Juemin Yi[1], Dong Wang[2], Anke Korte[1], Abbas Chimeh[1], Peter Schaaf[2], and Christoph Lienau[1] — [1]Institute of Physics, Carl von Ossietzky University Oldenburg, 26129 Oldenburg, Germany — [2]Institut für Mikro- und Nanotechnologien Macro-Nano and Institut für Werkstofftechnik, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Plasmon-enhanced excitonic nonlinear emission from semiconductors is interesting because the coupling of plasmon and exciton may boost the nonlinear generation efficiency. Nevertheless, to date, a detailed characterization of the electron dynamics of metal(plasmon)-semiconductor(exciton) composites is lacking. Herein, we present interferometric frequency-resolved autocorrelation (IFRAC) measurements on individual porous gold nanoparticles (nanosponges) incorporated with ZnO that allows us to track spectrally and temporally the response of the generated second-harmonic fields. We propose a method to distinguish homogeneous and inhomogeneous spectral broadening of plasmon resonances by Fourier-transform of the IFRAC traces to get two-dimensional (2D) IFRAC spectra. More importantly, we observed enhanced excitonic SHG emission at 390 nm from ZnO by coupling to localized plasmons of gold nanosponges. Pronounced features of coherent plasmon coupling are observed, proving that localized plasmons enhance the nonlinear ZnO excitonic emission, as revealed from the fundamental (FM) sidebands of the 2D spectra.

O 31.9 Tue 16:00 H8

Low-temperature infrared dielectric function of hyperbolic α-quartz — •Christopher J. Winta, Martin Wolf, and Alexander Paarmann — Fritz-Haber-Institut der Max-Planck-Gesellschaft

Natural hyperbolic materials, where the principal components of the dielectric tensor have opposite signs, like hexagonal boron nitride [1], have recently attracted much attention due to their unique characteristics for polaritonic nanophotonics [2]. Here, we show that also the common uniaxial crystal α-quartz exhibits multiple hyperbolic bands in the far-infrared (far-IR), which support low-loss hyperbolic modes at low temperatures.

We determine the IR dielectric properties of α-quartz in the temperature range from 1.5 K to 200 K. Far-IR reflectivity spectra of a single crystal y-cut were acquired in 8 distinct configurations. Fitting a multi-oscillator model globally to these data allows for extraction of frequencies as well as damping rates of the in-plane and out-of-plane, the longitudinal and transverse IR-active optic phonon modes, and hence the temperature-dependent ordinary and extraordinary dielectric functions, $\varepsilon_\perp(\omega)$ and $\varepsilon_\parallel(\omega)$, respectively.

The results are consistent with previous high temperature studies [3] and indicate remarkably high quality factors, Q, for polaritons at low temperatures in α-quartz's hyperbolic spectral region.

[1] Caldwell et al., *Nature Communications* **2014**, *5*.
[2] Liu, Lee, Xiong, Sun, Zhang, *Science* **2007**, *315*, 1686–1686.
[3] Gervais and Piriou, *Phys. Rev. B* **1975**, *11*, 3944–3950.

O 31.10 Tue 16:15 H8

Tunable Low Loss 1D Surface Plasmons in InAs Nanowires — •Yixi Zhou[1], Jianing Chen[1], and Thomas Taubner[2] — [1]Institute of Physics, CAS — [2]Institute of Physics (IA), RWTH Aachen

Due to the ability to manipulate photons at the nanoscale, plasmonics has become one of the most important branches in nanophotonics [1]. The prerequisites for the technological application of plasmons include high confining ability (λ_0/λ_p), low damping, and easy tunability. However, plasmons in typical plasmonic materials, i.e. noble metals, cannot satisfy these three requirements simultaneously, therefore limiting their overall applicability [2].

Here, indium arsenide (InAs) nanowires are identified as a material that satisfies all three prerequisites, providing a platform for modern nanophotonics. The dispersion relation of InAs plasmons is determined using the nanoinfrared imaging technique, and show that their associated wavelengths and damping ratios can be tuned by altering the nanowire diameter and dielectric environment. The launched plasmons simultaneously exhibit high confinement factor ($\lambda_0/\lambda_p = 34$) and low damping rate ($\gamma^{-1} = 25$) [3]. The observation of InAs plasmons could enable novel plasmonic circuits for future subwavelength applications.

[1] J. A. Schuller et al., Nat. Mater. 9, 193 (2010).
[2] D. K. Gramotnev et al., Nat. Photonics. 4, 83 (2010).
[3] Y. Zhou et al., Adv. Mater. 30, 1802551 (2018).