Scanning nanogap cavity microscopy

Plasmonic nanostructures have the ability to locally enhance optical fields in a tiny volume, which is essential for studying light-matter interaction on the nanoscale. Many such optical antennas with optimized properties have been developed over the years. However most of them have a planar structure, which makes them incompatible with conventional, tip-based scanning probe measurements.
Here we present a method to use planar nanostructures, in particular a Nanocube-on-mirror cavity, as scanning probes for nanoscopy. To achieve this we use a novel tip-less approach – a technique to scan a planar probe parallel to a planar sample at a distance of few tens of nanometers. We measure distance and tilt between the surfaces of probe and sample with 1 nm position and sub-0.1 mrad tilt resolution by using optical near- and far-field techniques. We show how we can deterministically place gold nanocubes on this probe to assemble a scanning nanogap cavity.

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Strong Quenching of Dye Fluorescence in Perylene Orange/TMDC Hybrid Structures

While monolayer transition metal dichalcogenides (1L-TMDCs) have emerged as 2D semiconductors with multiple applications in optoelectronics, their combination with dye molecules to form promising hybrid structures, since they should allow charge transfer after optical excitation as required for photodetectors or solar cells. Here, we discuss the preparation of such systems, i.e. the deposition of perylene orange (PO) onto substrates by means of spin coating, stamping, and thermal vapor deposition (TVD) and compare these methods regarding the quality of the dye layer and practicability of the process. For TVD-fabricated 1L-TMDC/PO hybrid structures, we observe a drastic
quenching of the dye fluorescence in terms of both intensity and lifetime reduction for all used TMDCs compared to hBN/PO references. This quenching is attributed to electron or hole transfer depending on the energy levels of the molecule and the specific TMDC, respectively.

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Signatures and Scaling of the Strong-Field Ionization Response in Low-Order Harmonic Generation

The understanding of nonlinear response in dielectric solids has been sucsessfully extended to the strong field ionization regime by linkinghigh-order harmonic generation (HHG) to intraband carrier dynamics and interband recombination [1,2]. Recently, it was found that thesemechanisms do not explain the emission of low harmonic orders and are, instead, generated by the strong field tunneling excitation that drivesBrunel [3] and injection currents [4]. So far it is not clear for which field and material parameters the injection mechanism remains as thedominant contribution. Using a semiclassical ionization radiation model, we examine the scaling behavior of ionization induced low-order harmonics and discuss for which field parameters injection harmonics become particularly strong. Finally, signal ratios from different polarizationconfigurations are examined that allow for robust experimental identification.

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Correlation effects in high harmonic generation from finite systems

Using the Hubbard model we study how the process of high-order harmonic generation (HHG) is modified by electron-electron correlation for both finite and bulk systems. A finite-size enhancement of the HHG signal is found and attributed to electrons backscattering off the lattice edges. Additionally, with the increasing strength of the electron-electron correlation an enhancement of the high-frequency regime of the HHG spectrum is found. This is attributed to the on-site Coulomb repulsion between electrons giving rise to a localized correlation-induced quiver motion of the electrons. The finite-lattice enhancement dominates the HHG spectra from a few harmonic orders until a threshold from which the correlational enhancement dominates. This threshold is determined by the degree of correlation and decreases into the low-frequency regime for increasing electron-electron correlation. This infers that as the electron-electron correlation increases, the finite-size effect on the electron dynamics decreases.

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Holographic Imaging and Reconstruction of Laser-Induced Modifications in thin films

Well controlled laser material processing with a spatial resolution on the scale of the laser wavelength is key to the realization of a large variety of applications. Respective developments will strongly benefit from a full spatial and temporal characterization of the laser-induced plasma evolution. To this end, we implemented an experiment based on coherent diffractive imaging (CDI). The probe pulse images the spatial plasma profile evolution induced by the pump pulse in a thin gold foil. The resulting scattering images are used for a reconstruction via phase retrieval. In contrast to typical CDI experiments, we record a superposition of scattered radiation and the radiation transmitted through the intact foil, leading to holographic signatures. Here, we present a systematic numerical analysis of the role of these holographic features for the object reconstruction as well as experimental optimizations. We further present a first successful application of the reconstruction method to experimental data.

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Modeling ultrafast plasma formation in dielectrics using FDTD

The Finite-Differences-Time-Domain (FDTD) method solves Maxwell's equations on a spacial grid that can be easily extended by rate equations for e.g. ionization and is therefore optimally suited for modeling nonlinear laser-material interaction and plasma formation in dielectrics close to the damage threshold. The material response is modeled using nonlinear Lorentz oscillators for Kerr-type nonlinearities and Brunel as well as injection currents associated with the excitation of electrons into the conduction band for higher order nonlinearities. Along with strong field ionization, plasma formation is induced by impact ionization which is strongly dependent on the electron velocities. To avoid simulating the full electron velocity distributions required for the calculation of the impact ionization rates, we apply an effective rate equation model for the electron temperatures and drift velocities. First simulation results for strong and ultrashort laser pulses tightly focused into thin fused silica films show the formation of a pronounced ionization grating.

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Analysis of x-ray single-shot diffractive imaging using the propagation multislice method

Single-shot wide-angle x-ray scattering has enabled the three-dimensional characterization of free nanoparticles from a single scattering image. Key to this method is the fact, that the scattering patterns contain information of density projections on differently oriented projection planes. Wide-angle scattering typically requires XUV photon energies where absorption and attenuation cannot be neglected in the description of the scattering process. The multislice Fourier transform (MSFT) method, which provides a fast scattering simulation within the Born approximation, can be extended to also include these propagation effects. In this presentation the performance of conventional MSFT and propagation MSFT will be discussed and compared to exact results obtained from Mie theory. As a first application, selective resonant scattering from core shell systems is explored.

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Vibrational Sum-Frequency Generation Spectroscopy: Spectroscopic interrogation of thinnest of layers

Vibrational Sum-Frequency Generation (VSFG) spectroscopy is a non-linear vibrational spectroscopic technique that combines chemical specificity of vibrational spectroscopy with interface sensitivity of even-order non-linear spectroscopy. Unlike conventional vibrational spectroscopy, the signal beam (sum-frequency photons) is generated at the sample, thus it not only provides the energetics of the vibrational bands, but also the information about the spatial symmetry of those bands. We aimed at exploiting this property to probe the molecularly functionalized surface. Any information about the orientation and interaction of the molecules with surface can be crucial to tailor its efficiency. Our results suggest that the molecule is adsorbed differently on a gold surface and on alumina coated gold surface due to different chemical entities involved in binding, further a change in the thickness of alumina layer influence molecular orientation significantly possible due to apparent change in the surface roughness or different packing density.

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Laser-driven liquid bubbles for studying elasticity, and sample delivery

Laser ablation of pure liquids in vacuum is a sparsely studied area due to the difficulties in experiment design, at the same time this technique is ideal for studying the behavior of fluids in extreme conditions. Here we present the results of time-resolved bright field imaging of liquid glycerol plumes in vacuum following ablation with a picosecond infrared laser. This experiment reveals previously unobserved surface bubbles formed at the liquid-vacuum interface as a result of photomechanical effects. The sustenance and rupture of these bubbles need to be explained using theorized and sparsely studied phenomena of the existence of elasticity in liquid glycerol. Glycerol which is widely regarded as a Newtonian fluid shows behaviors unexplainable satisfactorily with surface tension alone while forming liquid bubbles following the ablation.

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Impact of coherent phonon dynamics on high-order harmonic generation in solids

We theoretically investigate the impact of coherent phonon dynamics on high-order harmonic generation (HHG), as recently measured [Hollinger et al, EPJ Web of Conferences 205, 02025 (2019)]. A method to calculate HHG in solids including phonon excitation is developed for a model solid. Within this model we calculate the signal of specific harmonics as a function of a pump-probe delay in the pico-second range. The characteristic behavior of the harmonic signal is traced back to underlying phonon dynamics.

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Design of an electrochemical cell for VSFG

Vibrational sum-frequency generation (VSFG) using an infrared (IR) laser pulse as probe and a visible laser pulse for upconversion is a powerful technique to study surfaces. Due to its 2nd order nonlinearity VSFG is sensitive towards surfaces, while ignoring bulk media. Combining VSFG with various electrochemistry techniques enables direct probing of vibrational modes of the absorbed molecule on electrode surfaces and its oxidized/reduced species. To practically study these processes a measurement cell has to be designed to combine VSFG and electrochemistry. In my poster I will show the development and advantages, disadvantages of two different types of cells with them.

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Investigating Interexcitonic Coherences with Two-Dimensional Electronic Spectroscopy in Different Environments

The detection and control of coherence between different quantum systems, e.g., the interexcitonic coherence of nanomaterials, is key to applications in quantum technology. However, characterizing these coherences is difficult due to the rapid dephasing and broad spectral features resulting from the polydispersity of the samples. Here, we address this challenge by applying two-dimensional electronic spectroscopy (2DES), which allows us to follow the dynamics of the density matrix of the quantum system and thus also the dynamics of the coherences. In addition to room-temperature measurements in liquid phase, we have also performed low-temperature measurements in solid organic glasses to achieve a reduction of the homogeneous linewidth. As an outlook, we propose to combine 2DES with the spatial resolution of a photoelectron emission microscope in the nanometer range to investigate quantum systems at interfaces, i.e., to study actual quantum devices.

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Tunnelionization induced generation of low harmonics at the interface of media

In the strong-field region, in addition to Kerr nonlinearities, tunneling ionization processes are crucial for understanding nonlinear phenomena in dielectric media. In this poster, the influence of tunnel ionization processes on the reflectance spectrum of dielectric solids, is investigated. The simulations are performed using the finite-difference time-domain method. It is shown that tunnel ionization processes, in particular the injection current, is the dominant term for the generation of low harmonics.

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Sub-10 fs direct laser writing of broadband surface waveguides

Direct laser writing is a well established technique for the fabrication of three-dimensional structures in transparent materials. However, fabricating optical waveguides near the surface of glasses remains a challenge due to ablation. The current work aims to overcome this limitation by focusing sub-10 fs laser pulses at a quasi-grazing incidence. In fused silica, the microstructures obtained support broadband waveguiding in the visible and near-infrared spectral regions. The modes supported in these structures exhibit an evanescent component enabling the probing of the optical properties in the vicinity of the surface. As an application, we use these surface waveguides to demonstrate direct refractive index sensing and plasmonic sensing with sensitivities up to 3793 nm/RIU.

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Light-matter interaction within the framework of stochastic equations

The detailed description of light-matter interactions usually involves the quantum description of both constituents. A realistic description further requires the inclusion of the environment in the form of decoherence and dissipation. Without any assumptions and approximations, such a system has an exponentially large number of degrees of freedom. In some cases, the system is permutationally symmetric, and the number of degrees of freedom is significantly reduced [1]. However, even this reduction is not sufficient for a large number of particles. Phase-space methods are capable of resolving this curse of dimensionality. A high-dimensional density matrix is replaced by a quasi-probability distribution function [2]. In some cases, this distribution function satisfies the Fokker-Planck equation and may be sampled by stochastic trajectories, with quantumness inscribed at the correlation properties of stochastic processes. The price of this is the intrinsic instability of stochastic equations. In this poster, we show how to move from the quantum picture to the stochastic one, and share our successes and challenges in modeling light-matter interactions.

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Single-Cycle Electron Emission from Metal Nanotips

Exposing nanostructures to strong fields enables the emission of energetic electrons via near-field driven elastic backscattering [1]. In LiMatI, the availability of intense single-cycle or sub single-cycle waveforms [2,3] will enable to explore the formation and propagation of attosecond electron pulses in previously inaccessible regimes of the strong-field interaction. Here, the electron emission from tungsten nanotips under intense single-cycle pulses is inspected theoretically via one-dimensional TDSE simulations. The calculated carrier-envelope phase-dependent photoelectron energy spectra reveal prominent signatures with pronounced differences to previous studies performed with many-cycle pulses. The physical origins behind the observed spectral features are disentangled by extending the famous Simple Man's Model of strong-field physics.

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