ICFO International School on the Frontiers of Light

Photonics with Free Electrons

5-7 July, 2023

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University of Göttingen,
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Probing Electron-Photon Correlations with Integrated Photonics in an

1.Coulomb-correlated Few Electron States in a Transmission Electron Microscope Beam

Coulomb interactions between electrons play an important role in numerous phenomena in atomic, molecular, and solid-state physics. Anyway, in these systems, it is usually hard to observe Coulomb correlations on the single-particle level. Previously, this has also applied to free-electron beams, where few-body Coulomb effects have been concealed by event-averaged detection.

Recently, two-, three- and four-electron Coulomb correlations were characterized in the spectral and spatial domain [1]. Here, femtosecond-triggered photoemission from a nanotip [2, 3] enables temporal and spatial confinement of electrons. In the case of photoemission of a few-electron state, the initial Coulomb energies are acceleration-enhanced to about 2 eV at a beam voltage of 200 keV in an ultrafast transmission electron microscope [4].

In this poster contribution, I will discuss new ways of tailoring the few-electron Coulomb interaction. Generally, the implementation of few-electron states may foster new avenues for non-Poissonian electron beams [5], correlated probing, and free-electron quantum optics.

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2. Superposition of Laguerre-Gaussian modes with topological charge 2 in communication protocol

The development of fully functional quantum computers is an ongoing challenge, but researchers are making steady progress toward this ambitious goal. It is crucial to recognize that the advancement of quantum computers poses a potential threat to secure data transfer. Therefore, it becomes increasingly important to develop secure communication methods that not only match the ef>iciency of modern technology but also offer the same level of security as quantum communication protocols.

In this context, we propose the implementation of a simultaneous message-passing protocol based on optical vortices in free space. Our innovative solution has the potential to inspire novel applications in quantum optical systems. To achieve high-speed data transfer, we employ a digital micromirror device instead of the commonly used spatial light modulator (SLM) to encode information using a superposition of higher topological charge Laguerre-Gaussian modes.

Previously, we successfully compared two messages encoded with vortexes of charge 1 and -1 in binary (two-phase levels) [1] and ternary systems (three-phase levels). In our new scheme, we aim to expand the communication channels by incorporating the superposition of vortexes with charges 2 and -2. This enhancement enables us to compare four different images simultaneously. The functionality of our protocol was validated through two distinct experiments. The first experiment involved an original image and its three different corresponding distorted versions, while the second experiment encompassed two distinct original images and their corresponding distortions.

In our protocol, each image is represented as a perceptual hash and transmitted simultaneously. By calculating the Hamming distance between the images, we can determine the success of the comparison between these signals. To ensure security, we employ an optical implementation of the Fredkin gate, a three-qubit gate with one qubit serving as a control bit, along with two Hadamard gates that contribute to the reversibility aspect [2].

During this presentation, I will introduce the encoding and detection scheme of our protocol. Additionally, I will delve into the details of our experimental setup, including the results of a picture comparison experiment that highlights the significant potential of our protocol across various applications. Overall, we firmly believe that our work offers a valuable solution to address the evolving demands of the computational industry.

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3. Two-color current excitation in 2D materials using complex laser fields

We introduce a versatile and stable approach to drive ultrafast light field-induced currents in solids through two-color laser pulses at wavelengths 1550nm and 775nm. The pulses possess independent and controllable polarization states, allowing for the creation of intricate composite fields. By employing these bi-chromatic beams, we are able to manipulate the trajectories of electrons within 2D materials with the objective of investigating specific characteristics of their underlying band structure. Notably, when the waveform of the field exhibits asymmetry in relation to time inversion, an uneven distribution of excited electrons in momentum space occurs, resulting in a detectable photocurrent [1]. While conventional schemes typically involve a Michelson interferometer with the two-color laser beams spatially separated, our collinear approach offers superior stability with 3 mrad long-term phase jitter without the use of active stabilization techniques [2]. We anticipate that this highly stable configuration will serve as a foundation for exploring electronic phenomena in solids, such as spin polarization, Berry curvature and topological states, with unprecedented resolution at ultrafast timescales [3].

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4. Cooling times in femtosecond pump-probe experiments of phase transition materials with latent heat

Ultrafast pump-probe experiments can reveal the physics and chemistry of complex materials. Experiments are often limited by the need to limit the laser repetition rate to allow for the sample's back reaction between different laser pulses. However, few good methods exist for calculating this time. We investigate how sample geometry influences the cooling rate of VO2, a strongly correlated material with an ultrafast metal-insulator transition. We measure back reaction times of milliseconds, microseconds and nanoseconds in free-standing thin films, coated substrates and bulk crystals, respectively. We develop a simple latent heat model that reproduces the behaviour of all geometries and can therefore be used to predict the speed of the back reaction in other phase-change materials as well.

5. Real-time Wide-field broadband CARS microscopy

Coherent anti-Stokes Raman Scattering (CARS) is a powerful nonlinear vibrational imaging technique that provides label-free chemical maps of cells and tissues in their native state. This method involves the coherent excitation of vibrational modes in molecules using two pulses, called pump and Stokes, which are spatially and temporally overlapped. The source of the setup is an Ytterbium-amplified femtosecond laser, providing high-energy (≈ μJ) pulses at 1035 nm, with a repetition rate of 2 MHz. The pump has a narrow bandwidth of ≈1.1 nm (10 cm-1), ensuring sufficient spectral resolution. To generate the broadband Stokes beam, a YAG crystal is employed for white-light generation, resulting in a broadened spectrum in the 1100-1500 nm range. This region covers the so-called fingerprint region (500-1800 cm-1), the richest in chemical information. The Stokes is subsequently amplified employing a non-collinear optical parametric amplifier (NOPA). In this way it is possible to acquire hypercubes with large field of view in a few ms, ensuring video-rate level imaging capabilities, that may find applications in real-time fast biological dynamics.

6. A non-invasive technique for monitoring fuel production

Reducing carbon dioxide through plasmonic photocatalysis with the use of solar light and water has proved to be an efficient process for the generation of valuable fuels. Conventionally, the monitoring of alcohol production has relied on chemical methods such as gas chromatography, which can lead to potential losses in the measurement process. In our approach, we analyze the optical diffraction patterns produced by a thermal lens spectroscopic system and track changes in the refractive index of the sample using a Michaelson interferometer. We have developed two Matlab image processing algorithms to retrieve key information from both parts of the setup in real time. Our program optimizes fringe counting for the interferometric system, enabling us to accurately determine the sample's refractive index. Additionally, the thermal lens system provides information on the presence of binary mixtures within the sample. By coupling both systems, we are able to track fuel production noninvasively in real-time and compare the efficiency of different plasmonic catalysts, such as transition metal nanoparticles and iron-doped perovskites. This technique offers a way to optically measure the ratio of water to alcohol in a sample without the need for conventional chemical methods, providing a more accurate and efficient means of monitoring fuel production.

7. Optimizing Power Division using Metamaterial-Assisted Inverse Design in 1D Grating Waveguides

Power splitters are essential components that play a critical role in enabling signal merging, routing, and division to ensure the efficient operation of multi-channel systems in photonic integrated circuits. However, the use of conventional, bulky power splitters can lead to significant space consumption on the chip. To address this challenge, we propose the use of a metamaterial-assisted ultra-low-loss T junction power divider that integrates an input and two output grating waveguides, occupying a compact footprint of only 2.1 × 2.1 μm2. Furthermore, the presentation will elaborate on the significance of employing the proposed splitter structure for terahertz antenna array systems.

8. Laser deposited and structured 2D MoOx functional elements

Within this master thesis, we propose to investigate the implications of growing such α-MoO3 on Si substrates structured by laser processing. We expect that this approach will enhance the functional properties of the deposited MoO3 since it might induce the development of the α-MoO3 single crystals along a preferred orientation, thus obtaining large areas of large aligned 2D α-MoO3 crystals

9. Revolutionizing Photonic Technologies: The Fusion of Additive Manufacturing and Material Science

In this poster, we will delve into the transformative potential of the combination of photonics with additive manufacturing, a synergy that has the potential to redefine the relation of the fields of materials science and photonics. We will explore the emergent opportunities presented by the broad spectrum of materials that can be made compatible through additive manufacturing, highlighting how these can be leveraged to advance the development and application of photonic technologies. This discussion will further illuminate how additive manufacturing encourages innovation by enabling the exploration of novel 3D architectures in photonics, normally confined to the 2.5D capabilities of conventional CMOS cleanroom processes. Within this talk, we'll also show recent results in current projects carried out at the LAPD in EPFL, in particular the realization of photonic constructs via additive manufacturing by solution electrowriting of SU8 waveguides. More than just a manufacturing tool, additive manufacturing can be a catalyst for the design and discovery of new functionalities of light.

10. On-Chip Frequency Conversion with a GaAs platform

Frequency conversion is a well-studied effect of nonlinear optics, in which the nonlinear dielectric polarization of a material describes the response to an incident optical field. The result is the generation of new secondary fields of different frequencies, enabling a variety of applications. For instance, in the field of quantum communication it is used to produce entangled photon pairs and photons of telecommunication wavelength.

An approach to achieve high conversion efficiency is to include a highly responsive material. This is the motivation behind GaAs, since it has one of the largest second and third order nonlinear optical coefficients. Now the case of GaAs is well documented in academic literature, where several techniques has been demonstrated. The research interest of the project is to transfer this phenomenon in an integrated setup, where compact size and better scalability are exploited compared to the free-space alternative.

11. Improved electron optics for detecting PINEM orders inside of an SEM

Photon-induced near-field electron microscopy (PINEM) is an electron microscopy technique used for imaging electromagnetic fields with femtosecond time resolution [1]. Up until recently all PINEM works have been performed in transmission electron microscopes (TEM) which provide high-energy electrons (100-300 keV) for interaction with an excited near-field. However, scanning electron microscopes (SEMs) are more compact and less expensive than TEMs and provide an entirely different range of electron energies (1-30 keV), so it is of interest to perform PINEM inside an SEM as has been shown in [2]. Different electron energy range puts significantly stricter requirements on electron beam manipulation and control.

The goal of this project is to design a system to control low-energy electrons (0.1-1 keV) custom made for time-resolved experiments with high spatial resolution. This involves the simulation, fabrication and testing of electrostatic lenses of various designs. These lenses will be used to image energy resolved spectra to imaging screen of a microchannel plate with high magnification.

12. Quantum control of plasmon-induced extraordinary optical transmission

Hira Asif

Understanding the ultrafast processes at their natural-time scale is crucial for controlling and manipulating nanoscale optoelectronic devices under light-matter interaction. In this study, we demonstrate that ultrafast plasmon resonances, attributed to the phenomenon of Extraordinary Optical Transmission (EOT), can be significantly modified by tuning the spectral and temporal properties of the ultrashort light pulse. In this scheme, all-optical active tuning governs spatial and temporal enhancement of plasmon oscillations in the EOT system without device customization. We analyze the spectral and temporal evolution of the system analytically through coupled harmonic oscillator model and discuss time-resolved spectral and spatial dynamics of plasmon modes through the 3D-FDTD simulation method and wavelet transform. Our results show that optical tuning of oscillation time, intensity, and spectral properties of propagating and localized plasmon modes yields a 3-fold enhancement in the EOT signal. The active tuning of the EOT sensor through ultrashort light pulses paves the way for the development of on-chip photonic devices employing high-resolution imaging and sensing of abundant atomic and molecular systems.

13. Spatio-temporal resolution of plasmons in one-dimensional nanostructures with electrons

Controlling light-matter interaction at the nanoscale requires overcoming the well-known Abbe diffraction limit, which dictates the spatial resolution that we can achieve with direct illumination. However, electron beams interacting with nano-sized materials enable both the spatial precision to resolve the nanoscale details and the temporal resolution to study the rapid dynamics of the excited electromagnetic fields in the system. Specifically, we are interested in metallic structures that support plasmons, the collective oscillation of conduction electrons in metals, which are localized in the particle and that can be excited with electrons. Therefore, one way to characterize the plasmonic modes of the system is to study the energy loss of the electrons as they travel near the structure (i.e., electron energy loss spectroscopy (EELS)) due to the interaction with the material.
Here, we consider a swift electron passing by/through a long silver nanowire with a length of the order of microns and tens of nanometer in diameter. To describe the dielectric properties of the nanowire, we use a local Drude model for the metal and solve Maxwell equations using the Discontinuous Galerkin method in the time domain (DGTD), which allows us to achieve spatio-temporal resolution of the plasmonic modes in the nanometer and femtosecond range, as has been experimentally observed for planar surfaces, and recently investigated in metallic wires. By comparing the numerical and experimental results, we aim to model the corresponding EELS experiments to characterize the plasmonic modes of the nanowires. In this context, we collaborate with the experimental group of Prof. Christoph T. Koch at the Humboldt University of Berlin, which is able to acquire the EELS spectra by shooting electrons through/past the nanowire using a state-of-art transmission electron microscopy (TEM) featuring an energy resolution in the meV range. In this work, we develop a theoretical and numerical framework for treating quantitatively the interaction of swift electrons with plasmonic nanostructures with high spatiotemporal resolution.

14. Photon Transverse Orbital Angular Momentum in High Harmonic Generation

Light field with transverse orbital angular momentum, i.e., the so-called spatiotemporal optical vortex, was recently proposed and realized. However, since the spatiotemporal optical vortex was just recently discovered with few theoretical studies and experimental observation, many of its properties remain elusive. As known, high harmonic generation (HHG) is a cornerstone of nonlinear optics. How does the spatiotemporal structure of lights affect the generation of high-harmonic radiations? Does the spatiotemporal singularity survive the highly nonlinear process? Moreover, what kind of conservation rules does the transverse orbital angular momentum follow during the HHG process? These questions lay the foundation for the studies of interaction between structured light fields and matters, while these crucial questions have not been answered. Here, we study on the HHG driven by spatiotemporal optical vortex pulses. We show that the spatiotemporal optical vortex pulses allow one to impose optical orbital angular momentum on both the microscopic and macroscopic aspects of HHG, in which the electron dynamics has been significantly modulated as well as the spatial structure of HHG radiation, giving rise to several unique structures in the produced spatial-resolved harmonic spectra. Such structures have never shown up in previous HHG phenomena. We also demonstrate the conservation rule of photon transverse orbital angular momentum in the HHG process. Based on this conversion rule, we further show a robust method to control the spatiotemporal topological charge and the spectral structure of high harmonics.

15. Inverse Design of Smith-Purcell radiators and dielectric laser accelerators

Inverse design is a computational optimization technique and a powerful tool for designing nanophotonic structures. In inverse design, the structure's performance is evaluated through an objective function, which is then is optimized using gradient descent-based techniques. One highly efficient way to calculate the gradient of the objective function is the adjoint method, where only two simulations of the system are necessary to calculate the gradient for an arbitrary number of parameters. Since the computational cost of this method is virtually independent of the number of parameters, a large parameter space can be explored efficiently, which often leads to intricate structures differing significantly from designs based on human intuition.

Using inverse design, multiple applications have already been shown, such as highly efficient waveguides, complex optical demultiplexers, and different kinds of optical circuitry [1], as well as highly efficient light coupling to photonic nanostructures, used in dielectric laser acceleration [2].

In our work, we use inverse design on dielectric laser accelerators as well as Smith-Purcell radiators [3]. Our aim is to improve the performance of the current generation of structures with this novel technique, while staying within the well-tested confines of silicon photonics.

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16. Towards a Yb:fiber laser based frequency comb for nonlinear enhancement cavity experiments

Femtosecond enhancement cavities are used in high-sensitivity linear and nonlinear gas spectroscopy, as well as for efficient nonlinear optical frequency conversion. To seed such an enhancement cavity a fully tunable frequency comb with high pulse energies is necessary. Therefore, the repetition rate and the carrier-envelope offset frequency need to be controlled. We demonstrate a set-up that allows both, the stabilization of the repetition rate and the carrier-envelope offset frequency. The set-up was first characterized and we were able to extract the mode, which is most suitable for low noise properties. As a next step, the generation of an octave spanning supercontinuum was required to finally measure the carrier-envelope offset. This was attempted with a photonic crystal fiber, though we were only able to reach a pulse spectrum from 800 nm to 1400 nm. This goal could be achieved, for example, by the usage of higher pulse energies or an optimized photonic crystal fiber.

17. Probing Electron-Photon Correlations with Integrated Photonics in an

The combination of photonics and electron microscopy enables various applications ranging from the shaping of electron beams to novel spectroscopy techniques. The underlying free electron-light interaction also constitutes a cornerstone for future hybrid quantum technologies with theoretical works predicting possibilities for both the probing [1] as well as the creation and shaping of quantum optical excitations [2]. Here, the implementation of a platform based on the combination of an ultrafast transmission electron microscope (UTEM) and an integrated photonic microresonator suitable for non-classical experiments with single electrons and single photons is presented.

A silicon nitride waveguide-coupled ring resonator on a photonic chip is used to couple photons into and out of the microscope and the electron beam adjusted to a few nanometers distance to the resonator waveguide surface [3,4]. The efficient electron-light interaction is characterized by means of combined optical and electron spectroscopy [3]. Due to an increased photon-electron coupling strength caused by the ring resonator the energy gain and loss of the free electrons is visible in the electron energy spectrum [3].

Additionally, it was possible to identify correlated electron-photon pairs, that result from inelastic electron scattering with the vacuum-cavity mode [4]. The detection of the electron energy allowed a correlation of the energy loss to the photon existence.

The here presented setup enables the probing of single photon-single electron correlations and is a step towards new hybrid quantum technologies. In the future it might be usable in combination with external optical state generation to probe photon quantum states or facilitate electron wave function shaping.

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