Ultrafast Quantum Phenomena in the Near Field

692. WE-Heraeus-Seminar

18 – 20 March 2019
at the Physikzentrum Bad Honnef/Germany

WILHELM UND ELSE
HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 692. WE-Heraeus-Seminar:

Progress in understanding light-matter interactions at the nanoscale and developments in experimental methods to address quantum systems, together with the possibility to fabricate well-defined nanostructures that can largely tailor coupling strengths, have generated a cross-fertilization in areas somewhat disconnected just a decade ago. For example, quantum-optical concepts such as cavity quantum electrodynamics (cQED) begin to merge with near-field optics and advanced coherent spectroscopies. This has just started and the seminar aims at identifying and facilitating such connections among these different research fields.

In nano optics strong local-field enhancements lead to efficient couplings with quantum emitters although the quality factors of the associated field modes are usually rather low. Hence strong dissipation and efficient decoherence mechanisms persist and the quantum realm of dynamics and interactions are intrinsically on ultrashort time scales. Attributed to this close connection between nanoscale quantum dynamics and ultrashort time scales the seminar will cover the two topics **efficient nanoscale energy and charge transfer mechanisms** and **quantum coherence and correlations**. The latter topic comprises among others few-photon optical nonlinearities, tailoring decoherence and nano-optical coherent control schemes. Whereas these issues are relevant on the single- or few-photon levels, the third topic, **electron–near-field interactions**, deals with strong-field phenomena. For example, the quantized energy exchange between near fields and electron pulses would pave the path to fundamentally new experiments and attosecond electron pulses.

Joining near-field optics with quantum optics and ultrafast spectroscopy opens new perspectives that shall be discussed and explored.

Scientific Organizers:

Prof. Mario Agio — Universität Siegen, Germany
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Program

Program

Sunday, 17 March 2019

17:00 – 20:00	Registration	
18:00	*BUFFET SUPPER and informal get-together*	

Monday, 18 March 2019

08:00	*BREAKFAST*	
08:55 – 09:00	Scientific organizers	**Opening remarks**
09:00 – 09:45	Niek F. van Hulst	**Ultrafast excitation & detection of single photon emitters**
09:45 – 10:30	Johannes Feist	**Ultrafast coupled photonic, electronic, and nuclear dynamics in molecular polaritons**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:45	Javier Garcia de Abajo	**Plasmonics in atomically thin materials**
11:45 – 12:10	James Lim	**Multi-color quantum control for isolating excited state coherences in 2D electronic spectroscopy**
12:10 – 12:40	Stefan Y. Buhmann	**The influence of retardation, dielectric environments and mediating atoms on interatomic Coulombic decay**
12:40	*LUNCH*	

Program

Monday, 18 March 2019

14:00 – 14:45	Matthew Otten	**Optical detection and storage of dissipation driven entanglement in plasmonically coupled quantum dots**
14:45 – 15:10	Bert Hecht	**Strong coupling of single quantum dots to plasmonic nano resonators at room temperature**
15:10 – 15:35	Markus Lippitz	**Plasmonic quantum circuits**
15:35 – 16:30	*COFFEE BREAK* **and Poster**	
16:30 – 17:15	Shaul Mukamel	**Probing elementary molecular events, conical intersections, and chirality by ultrafast x-ray pulses and quantum light in microcavities**
17:15-18:00	Doris Reiter	**Influence of electron-phonon interaction on optical signals from semiconductor quantum dots**
18:00 – 18:25	Alfred Leitenstorfer	**Charge and spin dynamics in single-electron compound nanosystems**
18:25 – 18:50	Christoph Lienau	**Coherent exchange of surface plasmons between a metal tip and a gold nanorod probed with nanometric resolution**
18:50 – 20:00	*DINNER*	
20:00	**Poster Session**	

Program

Tuesday, 19 March 2019

08:00	*BREAKFAST*	
09:00 – 09:15	Stefan Jorda	**About the WE-Heraeus-Foundation**
09:15 – 10:00	Nina Owschimikow	**Photon statistics beyond $g^{(2)}$**
10:00 – 10:45	Stephane Guérin	**Quantum optics and quantum control at the nanoscale via plasmonics**
10:45 – 11:15	*COFFEE BREAK*	
11:15 – 12:00	Libai Huang	**Long-range hot carrier transport in hybrid perovskites visualized by ultrafast microscopy**
12:00 – 12:25	Donghai Li	**Fluorescence-detected multidimensional spectroscopy of a 2D material**
12:25 – 12:50	Frank Schlawin	**Theory of coherent control with quantum light**
12:50 – 13:00	**Conference Photo** (in the front of the lecture hall)	
13:00	*LUNCH*	

Program

Tuesday, 19 March 2019

14:30 – 15:15	Jeremy Baumberg	**Extreme nanooptics with precision molecular assembly**
15:15 – 15:40	Marten Richter	**Nanoscale photon quantization in media using QNM**
15:40 – 16:05	Thomas Zentgraf	**Spatial entanglement of two-photon states with metasurfaces**
16:05 – 16:30	*COFFEE BREAK*	
16:30 – 17:15	Claus Ropers	**Quantum coherent electron-light interactions in ultrafast electron microscopy**
17:15 – 17:40	Nahid Talebi	**Quantum coherent interference paths in the generalized Kapitza-Dirac effect**
17:40 – 18:05	Giovanni M. Vanacore	**Attosecond coherent control of a free-electron wave function via semi-infinite light fields and plasmon polaritons**
18:05 – 18:30	Peter Baum	**Waveform electron microscopy of electromagnetic field cycles**
19:00	*HERAEUS DINNER* *(social event with cold & warm buffet with complimentary drinks)*	

Program

Wednesday, 20 March 2019

08:00	*BREAKFAST*	
09:00 – 09:45	Thomas Fennel	**New routes to imaging the classical and quantum dynamics of finite systems**
09:45 – 10:30	Vladislav Yakovlev	**Optical-field control of electrons in solids**
10:30 – 11:00	*COFFEE BREAK*	
11:00 – 11:45	Peter Hommelhoff	**Femtosecond two-color physics in the nearfield of nanometer sharp metal tips**
11:45 – 12:10	Boris Bergues	**Few-cycle laser driven reaction nanoscopy on isolated nanoparticles**
12:10 – 12:30	**Open Discussion and Closing Remarks**	
12:30	*LUNCH*	

End of the seminar and departure

NO DINNER for participants leaving on Thursday morning

Posters

Posters

Alexander Achtstein	**2D *k*-space spectroscopy and coherence in 2D nanoemitters**
Martin Aeschlimann	**Mixing the light-spin with plasmon-orbit**
Robert Bennett	**The virtual photon approximation for three-body interatomic Coulombic decay**
Alexander Block Pawel Woźniak	**Spatio-temporal mapping of energy transfer from solids to light harvesting complexes**
Edoardo Carnio	**Coherent control of two-photon absorption via entangled photons**
Antonietta De Sio	**Ultrafast dynamics through a conical intersection in an organic photovoltaic thin film probed by two-dimensional electronic spectroscopy**
Pascal Dreher	**Nonlinear photoemission from plasmonic vortices**
Rui E. Ferreira da Silva	**Study of non-Markovian dynamics in organic polaritons induced by a multimode coherent field**
Daniel Fersch	**Combining fluorescence and photoemission electron microscopy for the investigation of ultrafast surface phenomena**
Patrick Folge	**Coupling of quantum emitters to photonic modes in disordered nanotextured materials**
Bettina Frank	**Atomically flat gold flakes for surface plasmonics**
Veit Giegold	**Photoluminescence-detected coherent radial breathing mode oscillations of single carbon nanotubes**

Posters

Harald Gießen	**Plasmonic anyons and skyrmions as simulators of many-body quantum systems in artificial gauge fields on the nanoscale**
Petra Groß	**Dynamic imaging of plasmonic nanostructures with an ultrafast point-projection electron microscope**
Achim Hartschuh	**Deterministic SHG hotspot switching in plasmonic nanoantennas using phase-shaped laser pulses controlled by neural networks**
Jeanne Heintz	**Optimizing the coupling between fluorescent molecules and plasmonic nanoantennas using DNA**
Matthias Hensen	**Revealing the impact of quasinormal mode superposition on local field dynamics within a plasmonic cavity**
Georg Herink	**Ultrafast control of quantum systems via strong terahertz-nearfields**
Mohsen Janipour	**Switchable cavity-assisted energy transfer in the infrared via surface phonon polaritons**
Wouter Koopman	**Influence of transient cavity degeneration on strong coupling in hybrid exciton-plasmon nanorods**
Klas Lindfors	**Spectroscopy of graphene nanoribbons**
Andrea Mattioni	**Near feld induced speedup of exciton diffusion in light-harvesting arrays**
Alfred J. Meixner	**Enhanced radiative plasmon decay and superluminescence from a nanoscale plasmonic cavity**

Posters

Frank Meyer zu Heringdorf	**Quantum interference during the electron emission from surface plasmon polaritons**
Magnus Molitor	**Theoretical description of two-dimensional spectroscopy in a CdTe quantum dot doped with a single Mn ion**
Thomas Pertsch	**Towards the investigation of ultrafast nanoscale quantum phenomena by superfocusing plasmonic nanotips**
Erich Runge	**Hot-spots and plasmonic near-fields in Gold nanosponges**
Sina Saravi	**Photon-pair generation mediated by coupling of atoms to nonlinear photonic nanostructures**
Christoph Schnupfhagn	**First steps towards long-range coherent energy transfer in plasmonic lattices**
Josef Tiggesbäumker	**Polyanionic Silver clusters studied by photoelectron spectroscopy**
Zilong Wang	**Circularly polarized light-field driven current in solids and air**

Abstracts of Talks

(in chronological order)

Ultrafast Excitation & Detection of Single Photon Emitters

Matz Liebel[1], Lukasz Piatkowski[1], Esther Gellings[1], James T. Hugall[1], Anshuman Singh[1], Lisa Saemisch[1] and Niek F. van Hulst [1,2]

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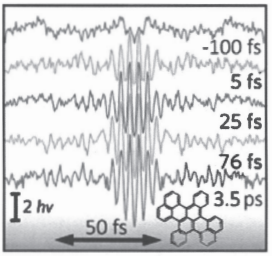
Tracing of individual molecules both in space and in time, provides super-resolution images of intra- and inter-molecular dynamic processes, all beyond the ensemble [1,2]. A host of important dynamic processes occurs on fs-ps timescale, such as electronic relaxation/dephasing, energy & charge transfer, vibrational relaxation, photo-dissociation-ionization, photo-isomerization. Yet capturing fast dynamics is fundamentally limited by the ns lifetime of the detected fluorescence. For the ensemble, fs-ps resolution is conventionally obtained by transient absorption and 2D electronic spectroscopy (2D-ES).



Here first, we challenge ultrafast transient absorption of a single molecule and have set out to probe the non-linear ultrafast response of the single molecule using a broadband laser in an effective 3-pulse scheme with fluorescence detection. 2D-ES of single molecules is getting into reach [3].

Next, stimulated emission has the advantage of coherence and unity quantum efficiency, compared to fluorescence. Moreover depleting the excited state rapidly reduces photodissociation. Thus, we have set out to directly detect the stimulated emission from individual molecules and Q-dots at ambient conditions, to disentangle ultrafast charge dynamics in the excited state, on fs-ps timescale.

Finally, we enhance the sensitivity and resolution using the local optical near-fields of plasmonic nano-antennas. We apply both deterministic scanning and stochastic mapping of the nanoscale plasmon-molecule interaction, to optimize the coupling strength and ultrafast interaction [4,5]. We apply the enhancement to photosynthetic complexes (LH2) [6], to detect first single FMO complexes at room temperature and to control defect emitters in hexagonal boron nitride.

References

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Ultrafast coupled photonic, electronic, and nuclear dynamics in molecular polaritons

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Traditional nanophotonics is concerned with engineering material systems to control light on a nanometer scale. However, over the last years, it has become clear that the reverse is also possible, i.e., to engineer light modes so as to modify material properties and dynamics. Remarkably, this can be achieved even when no actual light is present in the system by engineering the electromagnetic vacuum and its fluctuations, based on quantum electrodynamics (QED). One powerful tool to achieve such modifications is **strong coupling**, which occurs when the coherent energy exchange between a (confined) light mode and material excitations becomes faster than the decay and decoherence of either constituent. This creates a paradigmatic hybrid quantum system with eigenstates that have mixed light-matter character, so-called polaritons. Organic molecules present a particularly favorable type of emitter to achieve this regime even at room temperature due to their large dipole moments and stability. Polariton formation leads to changes in the excited-state character and energy levels (i.e., potential energy surfaces), which can affect a wide range of properties, such as energy transport, photochemical reactions, and even thermally driven ground-state chemical reactions. At the same time, such systems display ultrafast femtosecond-scale dynamics in their coupled photonic, electronic, and nuclear degrees of freedom. I will discuss ways to observe and manipulate these dynamics, based on two separate numerical approaches: (1) a quantum mechanics / molecular mechanics (QM/MM) simulation that includes quantized light modes and is able to represent the full chemical complexity of the molecules, and (2) a tensor-network-based approached that allows a full quantum simulation of all degrees of freedom.

Plasmonics in atomically thin materials

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Two-dimensional polaritons have emerged as powerful tools to manipulate light at atomic scales in materials such as graphene, transition metal dichalcogenides, and atomically-thin metal films. In this talk, we will review recent experimental advances in the fabrication and performance of atomically-thin metallic films, as well as fundamental properties of these excitations, including their in/out-coupling to light and their potential for applications in sensing, nonlinear optics, and quantum physics.

Multi-color quantum control for isolating excited state coherences in 2D electronic spectroscopy

J. Lim[1], C. M. Bösen[1], A. D. Somoza[1], C. P. Koch[2], M. B. Plenio[1] and S. F. Huelga[1]

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[2] *Theoretical Physics, Kassel University, Kassel, Germany*

Quantum effects in the energy and charge transfer in molecular systems have been extensively studied by coherent multi-dimensional spectroscopy. Two-dimensional (2D) electronic spectroscopy has revealed the oscillatory features in the optical response of photosynthetic and artificial light-harvesters, demonstrating the presence of quantum coherences on the energy transfer timescale. The origin and implications of the observed oscillatory 2D signals have been the subject of vigorous discussion, as 2D oscillations may not only originate from coherent motions in the excited state potential, where energy and charge transfer occur, but could be induced by the vibrational motions in the ground state potential. The spectral overlap between ground and excited state coherences in 2D electronic spectra makes the unambiguous identification of a coherent energy transfer a challenging task. In this work, we provide a coherent control scheme where a multi-color pulse sequence suppresses the generation of the ground state coherences in rephasing 2D spectra, yielding oscillatory 2D signals that are dominated by excited state coherences [1].

References

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The influence of retardation, dielectric environments and mediating atoms on interatomic Coulombic decay

R. Bennett[1,2], S. Bang[1] and S. Y. Buhmann[1,2]

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[2] Freiburg Institute for Advanced Studies, Freiburg, Germany

Interatomic Coulombic decay (ICD) is a recently predicted and subsequently observed resonance energy transfer mechanism: An inner-shell excited donor ion emits a photon which ejects an electron from a neighbouring ground-state atom, ionising the latter. Being a source of slow electron's, this process is believed to strongly significantly contribute to radiation damage in biological tissue.
We employ macroscopic quantum electrodynamics [1] to study the impact of retardation and realistic environments on the ICD rate [2]. Our results in free space reveal that ICD may persist over a much larger distance range than previously believed on the basis of purely electrostatic considerations. We also predict that the efficiency of the process can be modified by means of intervening media such as water as well as surfaces. Single mediator atoms in the vicinity of the donor–acceptor pair may have a similar effect [3].

References

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Optical Detection and Storage of Dissipation Driven Entanglement in Plasmonically Coupled Quantum Dots

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A system composed of quantum dots coupled with a common surface plasmon mode is considered, with each quantum dot possibly coupled to a separate photonic cavity mode. Cavity quantum electrodynamics calculations are carried out to show that upon optical excitation by a femtosecond laser pulse, the quantum dot excitons naturally evolve to a long-lived entangled state through the dissipative plasmonic mode[1,2]. If the quantum dots are coupled to separate photonic cavity modes, the time evolution of the two-photon pair correlation function of the cavity photons is an indicator of the entanglement of the coupled quantum dot system. Furthermore, if coupling of the photonic cavity and quantum dot modes is large enough, the quantum dot entanglement can be transferred to the cavity modes to increase the overall entanglement lifetime[3]. This latter phenomenon can be viewed as a signature of entangled, long-lived quantum dot exciton-polariton formation. The preservation of total entanglement in the strong coupling limit of the cavity/quantum dot interactions suggests a novel means of entanglement storage and manipulation in high-quality optical cavities.

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Strong coupling of single quantum dots to plasmonic nano resonators at room temperature

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Recently the observation of strong coupling of a single emitter to ultra-confined light fields in plasmonic resonators at ambient conditions has been reported [1,2,3]. The fact that strong-coupling conditions may be reached at room temperature is of immense interest because it represents a clear route to a practical implementation of true quantum behaviour in nano-optical systems. Such quantum behaviour may manifest itself in few-photon nonlinearities, deterministic exchange of energy, as well as polariton physics at ambient conditions. As ultra-confined fields are associated with metallic resonators, comparatively low quality factors are a necessary consequence, requiring large spectral bandwidths and ultrafast temporal response which are both afforded by plasmonics.
We review the latest experimental results and emphasize consequences of ultrafast energy transfer in strongly coupled quantum systems.

References

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Plasmonic quantum circuits

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Plasmonic waveguides promise to shrink photonic circuits to micrometer dimensions. At the same time, the small mode volume increases light-matter interaction so that individual quantum emitters couple with high efficiency to the waveguide. At low temperature, epitaxial GaAs quantum dots form almost ideal single photon sources. We present efficient launching of single plasmons by these emitters around 800 nm wavelength. Towards the more visible wavelength range, single molecules are promising quantum objects, but gold as the usual waveguide material leads to large losses. We present single crystalline silver waveguides. Dye molecules are excited by the propagating plasmons and also re-emit propagating plasmons. These experiments form the first steps towards photonic quantum computing by plasmons and ultrafast plasmonic nonlinearities by single emitters.

Probing elementary molecular events, conical intersections, and chirality by ultrafast x-ray pulses and quantum light in microcavities

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Multidimensional spectroscopy uses sequences of optical pulses to study dynamical processes in complex molecules through correlation plots involving several time delay periods. Extensions of these techniques to the x-ray regime will be discussed. Ultrafast nonlinear x-ray spectroscopy is made possible by newly developed free electron laser and high harmonic generation sources. The attosecond duration of X-ray pulses and the atomic selectivity of core X-ray excitations offer a uniquely high spatial and temporal resolution. We demonstrate how stimulated Raman detection of an X-ray probe may be used to monitor the phase and dynamics of the nonequilibrium valence electronic state wavepacket created by e.g. photoexcitation, photoionization and Auger processes. Conical intersections (CoIn) dominate the pathways and outcomes of virtually all photophysical and photochemical molecular processes. Despite extensive experimental and theoretical effort, CoIns have not been directly observed yet and the experimental evidence is being inferred from fast reaction rates and some vibrational signatures. Novel ultrafast X ray probes for these processes will be presented. Short X-ray pulses can directly detect the passage through a CoIn with the adequate temporal and spectral sensitivity. The technique is based on a coherent Raman process that employs a composite femtosecond/attosecond X-ray pulse to directly detect the electronic coherences (rather than populations) that are generated as the system passes through the CoIn. Applications will also be made for time-resolved diffraction, X ray sum frequency generation, and detecting molecular chirality.Nonlinear optical signals induced by quantized light fields and entangled photon pairs will be presented.. Quantum light opens up new avenues for spectroscopy by utilizing parameters of the quantum state of light as novel control knobs and through the variation of photon statistics by coupling to matter. Entangled-photon pairs are not subjected to the classical Fourier limitations on their joint temporal and spectral resolution. Nonlinear optical signals induced by quantized light fields and entangled photon pairs are presented. Strong coupling of molecules to the quantum vacuum field of micro cavities that can be used to manipulate their photophysical and photochemical reaction pathways and polariton relaxation in photosynthetic antennae are demonstrated. Incoherent multidimensional photon coincidence signals in cavities provide a novel probe of exciton relaxation. These are demonstrated for the LHCII photosynthetic complex.

References

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Influence of electron-phonon interaction on optical signals from semiconductor quantum dots

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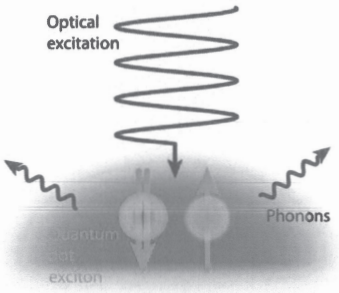
Semiconductor quantum dots are nano-objects with a discrete energy structure. Using optical manipulation the electronic state of a quantum dot can be well controlled and accordingly quantum dots are highly debated candidates in applications in quantum information technology. By using linear and non-linear optical spectroscopy techniques, the properties of a quantum dot can be revealed. In contrast to atomic systems, quantum dots are embedded in the solid state matrix and accordingly are subject to electron-phonon interaction.

In this talk, I will discuss optical signals resulting from optical excitation of a quantum dot under different excitation conditions and focus on the impact of the electron-phonon interaction. In the linear spectrum, the phonon influence is seen as phononic background and can be traced back to the emission of phonon wave packets from the quantum dot [1]. These phonon wave packets also lead to dephasing in optical state preparation schemes are Rabi oscillation or adiabatic rapid passage [2]. Using pump-probe techniques [3] or four wave mixing signals [4] the damping of Rabi oscillations can be revealed and the properties of the quantum dot can be studied.



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Charge and Spin Dynamics in Single-Electron Compound Nanosystems

Alfred Leitenstorfer

Department of Physics, University of Konstanz

The strong interband transition dipole of II-VI semiconductor quantum dots enables ultrafast transient transmission experiments resonantly exciting and probing only a single electron with strong quantum confinement at a time. Therefore, we have developed hybrid particles with functional polymer shells showing excellent long-time stability and negligible blinking even under nonlinear excitation. Latest advances in single-electron femtosecond spectroscopy techniques combined with broadband plasmonic antennas for maximum light-matter coupling allow us to monitor the dynamics of individual charges and spins. We find e.g. an intraband coherent electron dynamics lasting more than 100 ps which is enabled by the spin blockade for energy relaxation in the singly-charged dots. The ultrafast onset of biexcitonic absorption and single-photon gain at the fundamental trion resonance are readily detected and controlled. This work is aiming at a number of advanced ideas for manipulation and control of single electrons and photons which become available with this type of ultrafast quantum technology.

Coherent exchange of surface plasmons between a metal tip and a gold nanorod probed with nanometric resolution

Christoph Lienau

Carl von Ossietzky Universität Oldenburg

The coherent exchange of optical near fields between two neighboring dipoles plays an essential role for the optical properties, quantum dynamics and thus for the function of many naturally occurring and artificial nanosystems. These interactions are inherently short-ranged, extending over a few nanometers only, and depend sensitively on relative orientation, detuning and dephasing, i.e., on the vectorial properties of the coupled dipolar near fields. This, so far, has made it extremely challenging to analyze them experimentally.

Here, I will use a new and highly sensitive approach to probe these interactions with unprecedented precision. By illuminating the sharp apex of a gold tip by plasmonic nanofocusing, we create an essentially background-free, broadband and coherent light source with 5 nm dimensions and 300 nm spectral bandwidth. By approaching this tip with subnanometric precision to the surface of a 10 nm x 40 nm gold nanorod, we cannot only probe the local optical density of states of a single nanorod, but we can very nicely see how an ever-increasing number of surface plasmons is exchanged between tip and nanorod. This not only allows us to follow nicely the transition from weak to intermediate and strong coupling but also shows that and how these couplings depend on the coupled near fields. I will show that approach offers a new paths towards nano-optics with subnanometric resolution and may give fundamental new insight into quantum-plasmonic properties of hybrid nanosystems.

Photon statistics beyond $g^{(2)}$

Nina Owschimikow

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I will discuss an approach to integrate photon number sampling into a heterodyne detected pump-probe experiment with ultrafast time resolution [1]. The detection method is amplitude- and phase-sensitive, and consequently, after careful calibration, the mean values and the fluctuations of amplitude and phase can be converted into either Wigner functions or the full photon number statistics of the probe light field [2]. It is thus possible to access the quantum optical state of the probe light field with high time resolution. We demonstrate the capabilities of the method using a quantum-do-based semiconductor optical amplifier. We show the thermal nature of the amplified spontaneous emission in the photon number distribution, and the mixing of thermal and coherent quantum-optical features as a short laser pulse is amplified by the device. The transient change in the population inversion of the quantum dots during the amplification process is extracted from the time-resolved changes in the Wigner functions of the transmitted light pulse.

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Quantum optics and quantum control at the nanoscale via plasmonics

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The quantum control of emitters is a key issue for quantum information processing at the nanoscale. This generally necessitates the strong coupling of emitters to a high Q-cavity for efficient manipulation of the atoms and field dynamics (cavity quantum electrodynamics or cQED). Since almost a decade, strong efforts are put to transpose cQED concepts to plasmonics in order to profit of the strong mode confinement of surface plasmons polaritons [1]. Despite the intrinsic presence of lossy channels leading to strong decoherence in plasmonics systems, it has been experimentally proven that it is possible to reach the strong coupling regime [2]. In this work, we derive effective Hamiltonians [3,4], which allows us to describe the metallic nanoparticle-emitter interaction in full analogy with cQED formalism using a multimodal lossy cavity. We discuss (i) the concept of dressed states of quantum emitter strongly coupled to a metal nanoparticle [5], leading for instance to efficient/ blockade population transfers or superradiance/subradiance effects, and (ii) the multi-emitter adiabatic control via quantum plasmonics, for instance via stimulated Raman adiabatic processes [3,6]. Cooperative emission by quantum plasmonic superradiance will be also addressed [7,8]. The quantization of plasmons is usually analysed under the assumption of an infinite-sized bulk medium interacting with the electromagnetic field. We show how to reformulate it for finite-size nano-structures, highlighting sharp differences [9].

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Long-Range Hot Carrier Transport in Hybrid Perovskites Visualized by Ultrafast Microscopy

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The Shockley-Queisser limit for solar cell efficiency of ~ 33% can be overcome if hot carriers can be harvested before they thermalize. Recently, carrier cooling time up to 100 picoseconds was observed in hybrid organic-inorganic lead halide perovskites, but it is unclear whether these long-lived hot carriers can migrate long distance for efficient collection. We report direct visualization of hot carrier migration in $CH_3NH_3PbI_3$ thin films by ultrafast transient absorption microscopy, demonstrating three distinct transport regimes. Quasi-ballistic transport was observed to correlate with excess kinetic energy; resulting in up to 230 nanometers transport distance in 300 fs that could overcome grain boundaries. The nonequilibrium transport persisted over tens of picoseconds and ~ 600 nanometers before reaching the diffusive transport limit. These results suggest potential applications of hot carrier devices based on hybrid perovskites.

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Fluorescence-detected multidimensional spectroscopy of a 2D material

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Electronic 2D spectroscopy is an established technique to resolve ultrafast quantum phenomena and it is employed, e.g., to study excitonic dynamics in supramolecular systems [1]. In a nanooptical near-field context, however, the applicability of the conventional noncollinear phase-matching condition is limited. An alternative method is phase cycling, in which each laser pulse of a sequence is scanned systematically through a number of discrete phase steps. We have developed rapid-scan one-quantum [2] and two-quantum [3] fluorescence-detected 2D and 3D spectroscopy using a pulse shaper in shot-to-shot-modulation mode, and we have transferred the method also to a high-NA optical microscope offering additional spatial resolution [4].

Here we present a recent study on exfoliated monolayer $MoSe_2$. Monolayer transition metal dichalcogenides currently receive extensive attention due to their extraordinary optoelectronic properties. We perform and study the first 2D fluorescence spectra of an exfoliated $MoSe_2$ monolayer. The observed quantum beating behavior of cross peaks reveals a theoretically predicted [5] but previously undetected dark state near the electronically excited state and an additional unpredicted one near the ground state, activated by exciton–phonon scattering.

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Theory of coherent control with quantum light

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Spectroscopy with entangled photons promises to become a novel tool to reveal information about complex molecules. Nonclassical intensity fluctuations enhance nonlinear optical signals, such that e.g. two-photon absorption scales linearly with the photon flux, enabling nonlinear spectroscopies at low light intensities. Classical constraints for the simultaneous time and frequency resolution can be circumvented using entangled photons [1,2], thus allowing the investigation of samples in genuinely novel ways.

In this talk, we will explore when, and quantify by how much, entanglement can enhance an optical signal [3]. By formulating this problem in the language of coherent control, we find optimal classical and quantum pulse shapes, which can be directly compared.

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Extreme Nanooptics with Precision Molecular Assembly

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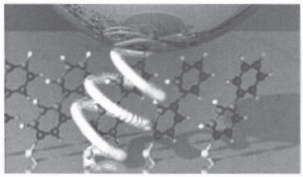
Coupling between coinage metal 'plasmonic' nano-components generates strongly red-shifted optical resonances combined with intense local light amplification on the nanoscale. I will show how we now create ultralow volume plasmonic cavities trapping light to the atom scale <1nm^3, and are routinely able to watch individual molecules and bonds vibrating. Using DNA origami we couple 1-4 dye molecules together optomechanically, and produce strong-light matter coupling that changes their quantum emission properties. We also watch redox chemistry in real time, watching single electrons shuttle in and out of single molecules, as well as 2D materials confined in the same gap. Prospective applications range from (bio)molecular sensing to fundamental science. I particularly focus on applications to nanomachinery actuation by light.



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Nanoscale photon quantization in media using QNM

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(i) Many nanoscale quantum emitters like quantum dots are coupled to localized photon or plasmon modes. For quantum applications a (modified) Jaynes-Cummings model is desirable. So far Jaynes-Cummings model (JCM) are formulated using phenomenological coupling elements, a systematic mode quantization in absorptive media was missing. In semi classical Maxwell calculations using few quasi normal modes (QNM) was successful for single photon properties such as radiative decay. Here, a microscopic mode quantization using QNM for a JCM in absorptive media is presented[1].
(ii) Furthermore algorithms for extracting many particle properties from coherent multidimensional spectroscopy may be shown[2] and (iii) prospects for using tensor networks for quantum photon propagation [3].

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Spatial entanglement of two-photon states with metasurfaces

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Optical metasurfaces open new avenues for precise wavefront control of light for integrated quantum technology. Here, we demonstrate a hybrid integrated quantum photonic system that is capable to entangle and disentangle two-photon spin states at a dielectric metasurface. By interfering single-photon pairs at a nanostructured dielectric metasurface, a path-entangled two-photon NOON state with circular polarization is generated that exhibits a quantum interference visibility of 86 ± 4%. Furthermore, we show with a metasurface-based interferometer that nonclassicality, phase sensitivity, and disentanglement can be obtained. The results demonstrate that metasurfaces can manipulate the spatial wavefront while preserving quantum characteristics and perform quantum interference without altering the phase information. Our findings provide a promising way to hybrid-integrated quantum technology with high-dimensional functionalities in various applications like imaging, sensing, and computing.

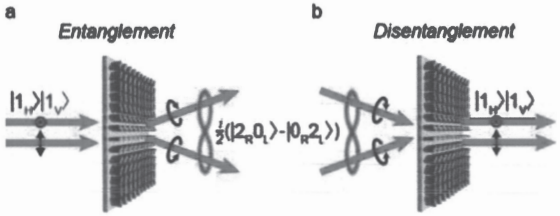

Fig. 1 Spatial entanglement and disentanglement of a two-photon state at a dielectric metasurface made of silicon nanofins. By enforcing a representation in the circular polarization base a two-channel path entanglement is achieved for a two-photon state. Experimentally, we characterized the two-photon state by analysing the photon-bunching effect in the two spatially separated paths. Passing the metasurface a second time, the entanglement is retracted and the original state recovered.

Quantum Coherent Electron-Light Interactions in Ultrafast Electron Microscopy

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Ultrafast Transmission Electron Microscopy (UTEM) is an emerging technique to study structural and electronic dynamics on the nanoscale. Besides its use as an analytical tool with simultaneous femtosecond temporal and nanometer spatial resolution [1], UTEM also provides for a unique test bench to study quantum optics phenomena with free electrons.

This talk will discuss several examples of free-electron beams interacting with optical near-fields at nanostructures, emphasizing quantum coherent processes. Specifically, for swift electrons traversing intense optical near-fields [2,3], we observe multilevel Rabi-oscillations on a ladder of quantized free-electron states [4], and implement Ramsey-type dual interactions in polarization-controlled, spatially separated near-fields [5]. Employing phase-locked two-color fields, coherent control of free-electron states is demonstrated [6], and we introduce a scheme to characterize the quantum state of such phase-modulated free-electron states in terms of their density matrix or Wigner function [6]. We demonstrate various new possibilities in the coherent manipulation of the longitudinal and transverse degrees of freedom of free-electron wave functions, including the optical preparation and characterization of attosecond electron pulse trains [6].

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Quantum Coherent Interference Paths in the Generalized Kapitza-Dirac Effect

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In the Kapitza-Dirac (KD) effect, propagating electron wavepackets are diffracted off a standing wave pattern of light created by two counter propagating light waves [1]. The momentum distribution of the final electron wave packet after the interaction is populated into distinct diffraction orders specified by the even harmonics of the momentum of the light waves. This effect has been intensively studied both theoretically and experimentally and is known to be an outcome of merely ponderomotive forces acting on the electron wave packet in free-space [2]. Here, we study a case for the generalization of the KD effect to include two inclined and focused laser beams interacting with a slow-electron wavepacket (Fig. 1). We show that the KD effect can be generalized to the realization of arbitrary momentum states of the electron wavepacket by controlling the interference between quantum pathways originating from distinctly different parts, absorptive and ponderomotive, of the interaction Hamiltonian. This offers fundamentally new degrees of freedom for designing light-controlled phase masks for free-space electron pulses [3].

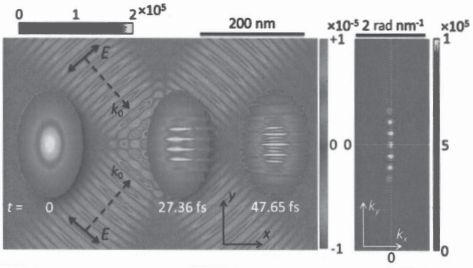

Fig. 1. An electron wave packet at the velocity of 0.03c, where c is the velocity of light, interacting with two mutually coherent CW laser waves at the wavelength of 30 nm. (Left Panel) Spatial distribution of the wavepacket at depicted times during the interaction with light waves and (Right Panel) Final distribution of the electron wavepacket at the momentum space.

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Attosecond coherent control of a free-electron wave function *via* semi-infinite light fields and plasmon polaritons

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The interaction between light and electrons can be exploited for generating radiation, such as in synchrotrons and free electron lasers, or for controlling electron beams in applications such as time-resolved electron microscopy for the dynamical investigation of materials and molecules. Using electromagnetic fields the coherent control of the electron wave function can be pushed to unexplored timescales, even below the attosecond regime, which would enable new applications in light-assisted quantum devices and diagnostics at extremely small timescales, such as those governing intramolecular electronic motions and nuclear processes.

In this contribution, we report on a generalized method for the coherent manipulation of free electrons with attosecond precision, and show that it can be pushed down to the zeptosecond regime with existing technology. A relativistic pulsed electron beam was made to interact with an appropriately synthesized electromagnetic field. The field was generated either by a sequence of two fs laser pulses reflected at the surface of a mirror (semi-infinite field), or by the coherent superposition of the surface plasmon polaritons (SPPs) optically-generated from nanofabricated structures (near field). The energy-momentum exchange resulting from the electron-field interaction was directly mapped via momentum-resolved ultrafast electron energy-loss spectroscopy, demonstrating the ability to efficiently manipulate both the longitudinal and the transverse phase distribution of the free electron wave function.

The potential of our approach to reach the attosecond timescale and below should pave the way to achieve unprecedented insights into non-equilibrium phenomena in advanced materials, and should play a decisive role in the rational design and engineering of future applications.

Waveform electron microscopy of electromagnetic field cycles

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Abstract: Nanophotonic devices and metamaterials can shape light-matter interaction in such a way that macroscopic effects are produced that are impossible to achieve with natural materials. Here we report our latest advanced in waveform electron microscopy [1] with THz-compressed single-electron pulses [2] in tilted and non-tilted configuration [3]. Sub-wavelength and sub-cycle electric and magnetic fields can be visualized in space and time and vectorial direction (see Figure). We will present our advances with increasing the spatial and temporal resolutions [4] and report advanced measurement methodologies such as sub-cycle scanning transmission electron microscopy (STEM). Waveform electron microscopy is a viable tool for understanding modern and future nonlinear-optical metamaterials and nanophotonic devices from a sub-element, sub-cycle, electromagnetic field-vector perspective.

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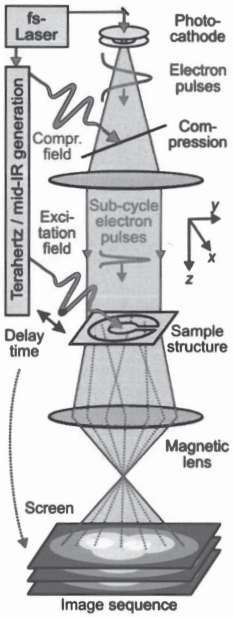

New routes to imaging the classical and quantum dynamics of finite systems

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When compared to atoms and solids, the collective and correlated electron dynamics in finite systems under intense light fields can be substantially modified, enhanced and controlled by local near-fields [1-3]. Near-fields result from electronic polarization and charge separation and can unfold on femtosecond or even attosecond time scales [4]. In this talk, two novel routes for characterizing the classical and quantum aspects of the underlying light-matter interactions at the nanoscale will be discussed. Though being fundamentally different conceptually, both schemes have come in reach with current short-wavelength FEL sources providing multicolor pump-probe pulses with exquisite control over the timing.

The first scenario – the *"nanoplasma oscilloscope"* – is motivated by previous theory work [5-7] on the XUV ionization of clusters and aims at tracing the complex evolution of space-charge potentials in laser-illuminated nanostructures. Such information is inaccessible with scattering methods such as coherent diffractive imaging but important for the understanding of non-linear plasma formation dynamics, radiation damage and relaxation processes in finite systems. Preliminary experimental data from a recent beam time at FERMI and the related theoretical analysis will be discussed.

The second scenario – the "*quantum coherent diffractive imaging*" (QCDI) - aims at exploring the non-linear response of extended nanosystems through near-field driven coherent quantum dynamics. A promising route to tracing spatiotemporal population dynamics is the analysis of respective signatures in single-shot diffraction images. We simulated the nonlinear response of Helium droplets under resonant 1s-2p excitation as a model using a coupled quantum-electromagnetic simulation based on a few-level approximation and utilizing the finite-difference time-domain method. The nonlinear modifications of the diffracted field through coherent bound state dynamics will be presented [8]. Our results illustrate the potential for spatiotemporal characterization of collective excitation dynamics in nanosystems and motivate new metrologies in the emerging field of quantum coherent diffractive imaging, paving the way into the realm of attosecond quantum imaging.

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Optical-field control of electrons in solids

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Physical speed limits of metrology and signal processing are defined by how fast electric or optical properties of materials can be manipulated. In this context, highly nonlinear processes induced in dielectrics and semiconductors by intense few-cycle laser pulses are of particular importance—these processes allow one to significantly change the state of electrons within a fraction of an optical cycle. A better understanding of how electrons behave driven by a nonperturbatively strong electric field may open up new paths toward ultrafast measurements and performing electronic operations with unprecedented speed [1]. This talk reviews recent progress in this direction at the Laboratory for Attosecond Physics [2-9].

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Femtosecond two-color physics in the nearfield of nanometer sharp metal tips

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Two-color laser excitation can lead to the excitation of currents based on quantum path interference [1,2]. We have shown that quantum path interference can lead to a large current enhancement and suppression in femtosecond two-color photoemission from nanometer sharp needle tips depending on the relative phase of the two colors. We observe current modulation visibilities of 94% [3], and more recently even up to 97.5% [4].

In simulations, we find that the perturbative pictures of quantum path interference can change quite dramatically if we go to strong fields. In that case, not only the photoexcitation process but also the trajectories of the photoemitted electrons are strongly influenced by the two-color phase [5]. We will show the current status of the ongoing joint experimental and theoretical work.

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Few-cycle laser driven reaction nanoscopy on isolated nanoparticles

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The chemical composition of atmospheric aerosols is a crucial factor in their contribution to air pollution and their impact on health. Strong laser fields offer a route for single particle chemical analysis, where molecular fragments are created in the laser interaction and spectroscopically identified. Strong-field induced processes in molecules such as ionization and dissociation have been subject to theoretical and experimental investigations for many decades [1]. These processes include, e.g., above threshold ionization, high harmonic generation, and laser induced electron diffraction. Since these effects strongly rely on the exact spatial and temporal evolution of the electric fields, they are also influenced and controlled by the presence of enhanced near-fields in the proximity of a nanostructure [2]. Under irradiation with light, the associated near-fields can induce, enhance, and control molecular adsorbate reactions on the nanoscale. So far, however, there is no simple method available to spatially resolve the near-field induced reaction yield on the surface of nanoparticles. Here, we close this gap by introducing *reaction nanoscopy* based on three-dimensional momentum resolved photoionization.

We have developed a nanotarget reaction-microscope based on recoil-ion-momentum spectroscopy (nanoTRIMS) [3], which permits recording both ions and electrons from the interaction of light pulses with molecules on a nanoparticle surface in coincidence. Nanoparticles are injected into the interaction region using an aerosol technique and are illuminated by 5 fs laser pulses at 720 nm with a peak intensity of ~3×10^{13} W/cm². We have studied the dissociative ionization of water, ethanol (and silanol) molecules on SiO_2 nanoparticles of various diameters, leading to the ejection of high-energy protons (see Fig. 1(a,c)). For large nanoparticles, the data show a clear propagation effect [4] and the influence of the laser-induced near-field on the yield of the dissociative ionization. The results from the nanoTRIMS experiment are modelled by semi-classical Monte-Carlo trajectory simulations [3], including Mie fields and charged particle interactions, cf. Fig. 1(b,d).

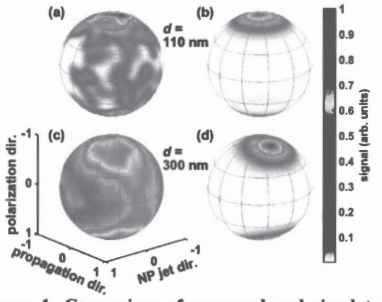

Figure 1. Comparison of measured and simulated angular proton distributions. The 3D (φ,θ,r) momentum distributions of protons are integrated along the radial coordinate and the retrieved two dimensional (φ,θ) density map is spanned over a unit sphere. The number of protons per solid angle is encoded in the color scale: (a) and (b) show, for 110 nm particles, the measurement and Monte-Carlo trajectory simulations, respectively, and (c) and (d) the analogues for 300 nm particles.

The results open the door towards strong near-field induced chemical reactions on nanoparticles and a route towards nanometer-scale spatio-selective chemical analysis of molecular adsorbates on aerosols. Reaction nanoscopy is suited for a wide range of isolated nanosystems and can provide spatially-resolved ultrafast reaction dynamics on nanoparticles, clusters, and droplets. Finally, the reaction nanoscope is suited to trace spatio-temporal variation in reaction yields in time-resolved pump-probe experiments.

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Abstracts of Posters

(in alphabetical order)

2D *k*-space Spectroscopy and Coherence in 2D Nanoemitters

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We introduce the electronic and exciton dynamic properties of 2D CdSe nanoplatelets and show that they are intrinsically directional light emitters with 2D degenerate transition dipoles, potentially important for applications in photonics including lasing and energy efficient display technology. We discuss the impact of coherence on the exciton-phonon interaction and recombination dynamics of these nanoemitters, demonstrating (quasi independent) tunability of emission energy, lifetime and exciton-phonon interaction.

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Mixing the Light-Spin with Plasmon-Orbit

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Transformation of light carrying spin angular momentum (SAM) to optical field vortices carrying orbital angular momentum (OAM) has been of wide interest in recent years. The interactions between two optical fields, each carrying one of those degrees of freedom, and furthermore, the transfer of the resulting angular momentum product to matter is seldom discussed. Here we measure the interaction between light carrying axial SAM and plasmon-polariton vortices carrying high order transverse OAM. The interaction is mediated by two-photon absorption within a gold surface, imprinting the resulting angular momentum mixing into matter by excitation of electrons that are photo-emitted into vacuum. We experimentally demonstrate that, counter-intuitively, the spatial distribution of the emitted electrons carries the signature of a subtraction of the spin from the orbit angular momenta. We show experimentally and theoretically that the absorptive nature of this interaction leads to both single and double photon-plasmon angular momentum mixing processes by one- and two- photon interactions.

The virtual photon approximation for three-body interatomic Coulombic decay

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Abstract— Interatomic Coulombic decay (ICD) is an ultrafast mechanism by which atoms and molecules can redistribute energy amongst themselves. In order to describe the process a wide variety of *ab initio* quantum chemical approaches have been developed, which are very effective at short distances. At longer distances (those where wavefunction overlap becomes negligible), it is possible to describe ICD by a 'virtual photon' approach [1], which was recently extended to include the influence of retardation and macroscopic dielectric environments [2]. Recent *ab initio* calculations have shown that ICD can be enhanced by an ICD-inactive mediator [3], which can also be considered in the new framework derived in [2]. Using our formalism we find analytic expressions for the rate in all geometries, and observe a surprising conclusion that the combined effects of retardation and placement of the mediating atom can almost completely suppress the ICD rate [4]. In this presentation I will outline the theory behind our three-body ICD formalism, summarise the results we have found so far, and provide some suggestions for future work.

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Spatio-temporal mapping of energy transfer from solids to light harvesting complexes

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Light-induced energy transport is a crucial process in photovoltaics, optical detectors, and even photosynthetic light harvesting. Whether described as exciton diffusion or excitation energy transfer, these processes occur on nanometer length and (sub-) picosecond time scales and are therefore difficult to observe directly. Only recently, transient absorption microscopy, combining femtosecond with nanometer resolution, has emerged as a powerful tool to study carrier migration in semiconductors [1]. In particular, the diffusion of non-equilibrium ("hot") carriers has been investigated as a possible route to overcome the Shockley-Queisser limit in solar cells [2].

Here, we combine super-resolved imaging with pump-probe techniques to directly visualize non-thermalized carrier diffusion in a thin gold film [3]. We track the spatio-temporal hot electron diffusion with 20 nm spatial and 0.25 ps temporal resolution. We observe an initial fast spatial diffusion within the first few picoseconds, followed by an abrupt transition to an orders of magnitude slower diffusion. Theoretical analysis with a two-temperature model identifies two distinct diffusion regimes. Initially, hot electrons dominate the diffusion process, followed by phonon-limited diffusion due to electron-phonon coupling. Finally, we extend our transient microscopy to synthesized membranes of light-harvesting complexes. By studying the nanoscale inter-complex transport across purified and reconstituted LH2 monolayers, we aim to answer open questions regarding the origin of long-range energy transfer [4] by resolving the process, for the first time, in space and time. The gained insights could be applied to the development of artificial light-harvesting systems in the future.

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Coherent control of two-photon absorption via entangled photons

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Coherent control exploits the coherence of classical light to drive an initial quantum state to a desired final state. In two-photon absorption (TPA), in particular, two photons are used to excite a molecule from the ground to an excited state. Under certain conditions, frequency-entangled photons drive the transition more efficiently than in the classical case [1]. We first show how this enhancement correlates with the entropy of entanglement of the two-photon state, both in the transient and in the steady states. We then extend our analysis to the case of a manifold of intermediate states, where we aim to control specific excitation pathways with the appropriate design of the two-photon states.

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Ultrafast dynamics through a conical intersection in an organic photovoltaic thin film probed by two-dimensional electronic spectroscopy

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Organic photovoltaic (OPV) materials are complex molecular systems that may present many vibrational degrees of freedom. In such large molecules, vibronic coupling may result in conical intersections (CIs) of potential energy surfaces. Large vibronic couplings close to CIs may induce efficient ultrafast transition between electronic states and, hence, they may profoundly influence the dynamics and yield of photoinduced charge transfer processes. So far, however, the importance of CIs for OPV materials has not yet been discussed. Here we use ultrafast two-dimensional electronic spectroscopy (2DES) to study the light-induced dynamics in an acceptor-donor-acceptor oligomer thin film employed in efficient OPV devices [1]. Upon impulsive excitation, we detect a grid-like peak pattern in the 2DES maps suggesting coherent wavepacket motion in the excited state with a dominant period of 30fs. Interestingly, pronounced peak shifts with decreasing peak splittings are observed at early times. After <50 fs, the 2DES pattern transforms into a broad, nearly featureless peak, strikingly different than the ones at earlier times. The dynamics show an abrupt amplitude drop concurrent with the spectral changes, indicating ultrafast depopulation of the excited state as the peak structure vanishes. At the same time, weaker oscillatory features with different period appear and persist for the rest of the investigated timescale, suggesting the emergence of a new, distinct wavepacket. No significant changes are observed at later times. All these experimental features do not match with a single-mode vibronic coupling model, as we recently reported for some polymers [2,3]. Supported by molecular dynamics calculations of stacked dimers of this oligomer, our results show that intermolecular transfer in this OPV thin film involves passage of the optically excited wavepacket through a CI within <50 fs [4].

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Nonlinear Photoemission from Plasmonic Vortices

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Observing strong-field effects in the interaction of light with condensed matter systems requires particularly high field intensities on the femtosecond time-scale. Nanostructures such as nanotips or nanoantennas provide an intrinsic local enhancement and spatial confinement of optical near-fields which is often utilized to obtain the necessary field intensities. Even though such enhancement is absent on flat surfaces, high near-field intensities can be achieved on metal surfaces using tailored surface plasmon polariton pulses.

Here, we use the angular momentum of surface plasmon polaritons in Archimedean vortex lenses to create transient plasmonic nanofoci. Performing our experiments on atomically flat gold surfaces provides us with precise control over the near field distribution. We employ spectroscopic photoemission electron microscopy on the optical skyrmion formed in the center of the Archimedean vortex lens with an orbital angular momentum of one and a total angular momentum of zero. We observe strong-field effects which are driven completely by the strong plasmonic near-field.

Study of non-Markovian dynamics in organic polaritons induced by a multimode coherent field

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Organic polaritons are hybrid light-matter states that present several advantages with respect to semiconductor polaritons. Among them, they present larger binding energies and allow for larger Rabi splitting. For a deeper understanding, the study of the dynamics of these systems is desirable, and little is known about their non-Markovian dynamics. In particular, the excitation from the groundstate of the system to the polaritonic states is the first step in any experiment and its modelling is the target of our work. To that purpose, the use of tensor network states is a fascinating tool that allows exact propagation of the system, treating thousands of degrees of freedom (plasmonic, phononic and radiative) on an equal foot. In this work, we show the results of the simulation of a prototype organic molecule (Rhodamine800) that is excited with a coherent multimode laser field. To see how the absorption spectrum changes we do a scan over the central frequency of the pulse and we analyse the influence of the phononic degrees of freedom.

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Combining fluorescence and photoemission electron microscopy for the investigation of ultrafast surface phenomena

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To learn something about individual quantum systems and their interaction with the environment it is advantageous to look beyond the optical diffraction limit on ultrashort timescales. For this purpose, we use a scanning fluorescence microscope as well as an aberration-corrected photoemission electron microscope combined with a NOPA-based laser source, giving us an effective temporal and spatial resolution of sub-20 fs and sub-10 nm, respectively. Here we present results on the polarization-dependent fluorescence enhancement of cyanobacterial Photosystem I thin films by gold nanorods, as well as first photoemission images of the protein dropcasted on monocrystalline gold flakes. We further show fluorescence and time-resolved photoemission from low-dimensional nanostructures performed with our pulse shaper. Finally, we want to use our microscopes to compare fluorescence-based multidimensional spectroscopy and coherent two-dimensional nanoscopy [1] on the same samples.

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Coupling of quantum emitters to photonic modes in disordered nanotextured materials

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Disordered nanotextured absorber materials support localized photonic modes. This modes for example find application in random lasing and the question of how individual emitters interact with such modes is of vital interest. Here we show field simulations for such photonic modes in randomly nanotextured absorber layers and investigate the dipole coupling strength of quantum emitters placed at different positions close or inside the photonic mode. As an outlook we compare the field simulations to actual experiments monitoring these modes and explore the possibility for future nanoscale cavity QED experiments based on the excitation with ultrashort laser pulses.

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Atomically flat gold flakes for surface plasmonics

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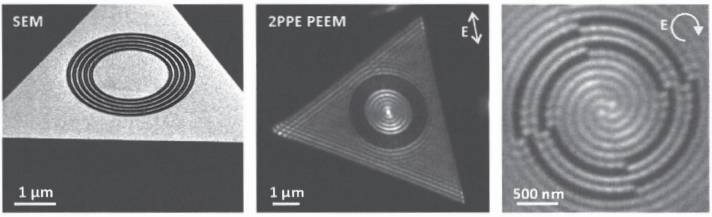
We use single crystalline gold flakes on atomically flat silicon substrates to generate ideally suitable interfaces for plasmon propagation. By electrochemical means, the thickness is tunable from a few tens to over 100 nm. Using sub-20 fs laser pulses around 800 nm, we excite surface plasmons, whose dynamics can be observed using time-resolved two-photon excitation electron emission (PEEM).

Plotting the dispersion of surface plasmons in a thin gold slab on silicon, one finds that excitation at 800 nm can lead to extreme wavelength reduction due to the dispersion slope of over five. Using focused ion beam for cutting rings with appropriate periodicity into the samples (see left image), we can excite concentric surface plasmons that create a nanofocus of only 60 nm width for 800 nm excitation (see center image) [1].



Archimedean spirals with broken n-fold radial symmetry excite surface plasmons with angular orbital momentum on the gold flakes (see right image). This leads in case of 4-fold symmetry to cloverleaf-type nanofoci of approximately 100 nm, which rotate during four optical cycles by 360 degrees [2].

Two-pulse experiments with a subwavelength-stabilized Michelson interferometer, allow investigation of the surface pattern dynamics with (sub-) femtosecond resolution, thus giving insight into the dynamics of the nanofocus formation as well as on the plasmonic spin-orbit coupling.

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Photoluminescence-Detected Coherent Radial Breathing Mode Oscillations of Single Carbon Nanotubes

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Vibrational resonances in single nanoobjects are mostly observed in the spectral domain. We show that the radial breathing mode (RBM) oscillation in single-walled carbon nanotubes (SWCNTs) can be probed in real-time using two-pulse femtosecond photoluminescence (PL) correlation spectroscopy. After the initial signal dynamics with strong modulations mainly resulting from spectral interference (Fig. 1b)), the PL intensity oscillates with a period of ~90 fs corresponding to the duration of a single RBM oscillation. We show that the signal contrast can be further optimized by spectral shaping of the laser pulse (Fig. 1c)). Our results help to clarify conflicting interpretations in the literature regarding exciton-phonon coupling and the initial phase of the RBM oscillations, i.e. the question if the nanotube's diameter first expands or shrinks upon impulsive excitation.

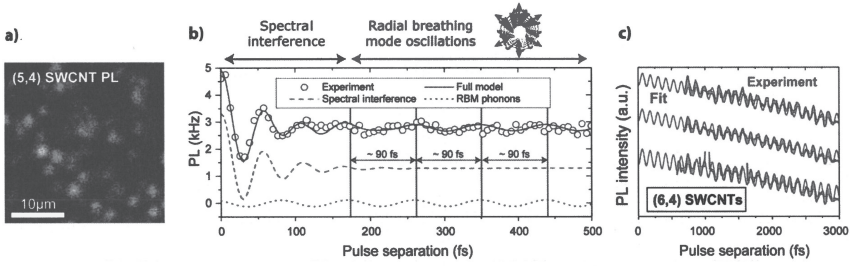

Fig. 1: a) Confocal PL image of (5,4) SWCNTs. b) PL from a single (5,4) SWCNT upon excitation with two 20 fs laser pulses for varying pulse separation. The signal oscillations for pulse delays > 150 fs reflect the excitation of coherent RBM oscillations. c) PL traces detected for three different (6,4) SWCNTs upon two-pulse excitation.

Plasmonic Anyons and Skyrmions as simulators of many-body quantum systems in artificial gauge fields on the nanoscale

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Frank Meyer zu Heringdorf, *Universität Duisburg-Essen*

Tim Davis, *University of Melbourne*

In condensed matter systems like MnSi, excitations of topological states exist that describe quantum phenomena such as the fractional quantum Hall effect, magnetic vortices, and their interactions [1-10].We are going to investigate the plasmonic analog of such excitations.

The resulting optical skyrmions and other topologically protected states are relevant for a number of applications such as quantum computing. Optically excited plasmonic systems with the correct topology and geometrical shape can simulate a variety of novel quantum systems which have not been found in nature. This way they become accessible to experimental observation and ultrafast time-resolved analysis.

We will use time-resolved ultrafast Photoemission Microscopy (PEEM), which has been well-established to observe plasmonic fields in the time domain [E2, E4]. Using PEEM it is possible to detect the subfemtosecond temporal dynamics of the formation, interaction, and destruction of vortices with different topological charge [E1, E3], which is currently not possible in solid state many-body systems. The experimental observations are accompanied by theory and numerical simulations [E3].

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[E1] **Revealing the subfemtosecond dynamics of orbital angular momentum in nanoplasmonic vortices**
G. Spektor, D. Kilbane, A. K. Mahro, B. Frank, S. Ristok, L. Gal, P. Kahl, D. Podbiel, S. Mathias, H. Giessen, F.-J. Meyer zu Heringdorf, M. Orenstein, and M. Aeschlimann
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[E2] **Short-range Surface Plasmonics: Localized Electron Emission Dynamics from a 60 nm Spot on Atomically Flat Single Crystalline Gold**
B. Frank, P. Kahl, D. Podbiel, G. Spektor, M. Orenstein, L. Fu, T. Weiss, M. Horn-von Hoegen, T. J. Davis, F.-J. Meyer zu Heringdorf, and H. Giessen
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[E3] **Subfemtosecond and Nanometer Plasmon Dynamics with Photoelectron Microscopy: Theory and Efficient Simulations**
T. J. Davis, B. Frank, D. Podbiel, P. Kahl, F.-J. Meyer zu Heringdorf, and H. Giessen
ACS Photonics **4**, 2461 (2017).

[E4] **Imaging the Nonlinear Plasmoemission Dynamics of Electrons from Strong Plasmonic Fields**
D. Podbiel, P. Kahl, A. Makris, B. Frank, S. Sindermann, T. J. Davis, H. Giessen, M. Horn-von Hoegen, and F.-J. Meyer zu Heringdorf
Nano Lett. **17**, 6569 (2017).

Dynamic imaging of plasmonic nanostructures with an ultrafast point-projection electron microscope

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We observe the emission and motion of photoelectrons in a single nanoantenna using an ultrafast point-projection electron microscope (UPEM). Employing ultrashort probe electron pulses of low kinetic energy, we measure the streaking of probe electrons by the free charges photoemitted from the nanoantenna.

Probing electrons are released from a plasmonic nanofocusing taper [1] and reach a plasmonic nanoantenna cut into a free standing, 30-nm thick gold film. Previously, we recorded the photoemission from such a structure and the subsequent motion of the electrons with 20-nm spatial and 25-fs temporal resolution [2]. To observe streaking, we have equipped the UPEM with a time-of-flight delay-line (ToF/DLD) detector that records position as well as kinetic energy of each probe electrons. We observe clear a streaking signature for probing electrons that traverse the gap region. The sensitivity is sufficient to resolve changes in kinetic energy <1%, and the transition from acceleration to deceleration (Fig. 1e) occurs within one delay step of 12 fs.

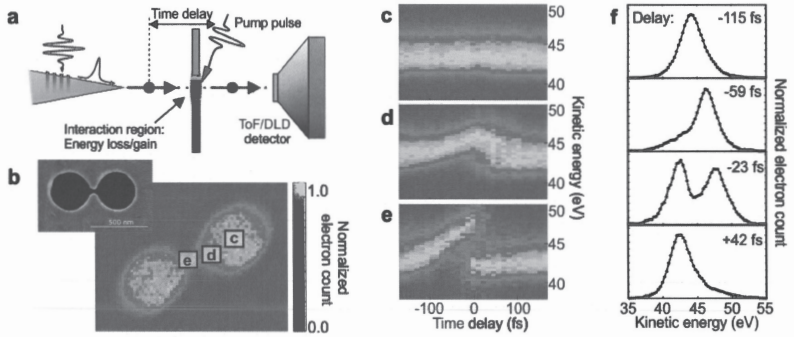

Fig. 1. Streaking of electrons emitted from a plasmonic nanofocusing taper by free charges. **a.** The UPEM setup, equipped with a space- and energy-resolved detector. **b.** Spatially resolved map of the energy count after transmission through a plasmonic nanoantenna (inset: SEM image of the structure). The squares indicate the regions where the streaking signals in **c-e** were recorded. **c.** Away from the antenna gap, no streaking is apparent. **d, e.** Closer to the gap, streaking is clearly visible. **f.** Kinetic energy spectra recorded in the gap region (position **e**) for different delay times.

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Deterministic SHG Hotspot Switching in Plasmonic Nanoantennas Using Phase-Shaped Laser Pulses Controlled by Neural Networks

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Plasmonic nanoantennas can efficiently focus broadband optical fields to form nanometer sized ultrafast hotspots [1]. The local field response of a metallic nanostructure, however, depends on its geometry in a nontrivial fashion, further complicated by the fact that this geometry can only be known to a certain extend in an experiment. We present an efficient procedure for the deterministic switching of hotspots in nanoantennas using neural network-controlled laser pulse-shaping [2]. We first numerically show that a trained neural network (NN) can be used to predict the relative intensity of the SHG hotspots in L-shaped nanoantennas for a given spectral phase and that it can be employed to deterministically switch individual hotspots on and off on sub-diffraction length scale. We then demonstrate that a NN trained on a specific L geometry can efficiently predict the hotspots in an antenna with different aspect ratio after minimal further training. Finally, we use the numerically trained NN to implement hotspot switching in the experiment.

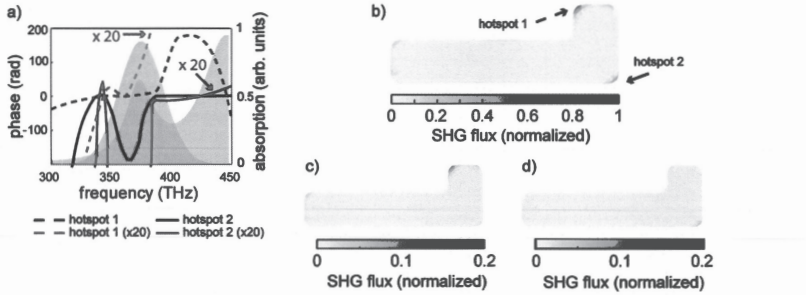

Fig. 1: Hotspot switching in a 90 nm - 250 nm gold nanoantenna. (a) Spectral phases maximizing the SHG at hotspots '1' and '2'. Laser and nanoantenna absorption spectrum are shown as red and green shaded area, respectively. (b)-(d) SHG for a flat phase laser pulse (b) and for the optimized phases for hotspots '1' (c) and '2' (d).

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Optimizing the coupling between fluorescent molecules and plasmonic nanoantennas using DNA

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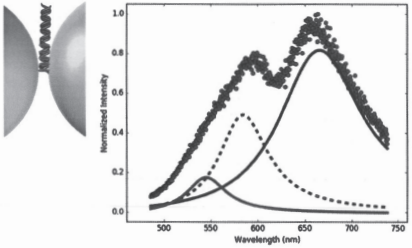
DNA self-assembly is a flexible and robust technique to controllably couple fluorescent molecules with plasmonic resonators. In particular, DNA-templated gold particle dimers enhance the spontaneous emission rates of quantum emitters by more than two orders of magnitude (Nat. Commun. **3** (2012) 962), providing bright single-photon sources with lifetimes below 10 ps and quantum yields above 50% (ACS Nano **10** (2016) 4806).

We will discuss recent results on the optimization of the interaction between multiple identical dye molecules and a single plasmonic resonator to reach a strong coupling regime. In particular, by screening the electrostatic repulsion between negatively charged gold colloids, we can reach interparticle spacings below 2 nm in DNA-templated dimers (ACS Nano **9** (2015) 978). This allows us to observe reproducibly, in scattering spectroscopy, plasmon mode splitting in the longitudinal resonance of single 40 nm gold particle dimers coupled to 5 ATTO647N or Methylene Blue molecules.



Preliminary result of strong coupling observed in scattering spectroscopy for a single DNA-templated gold dimer coupled to 5 dye molecules.

Revealing the impact of quasinormal mode superposition on local field dynamics within a plasmonic cavity

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Plasmonic cavities are known for supporting discrete modes upon resonant excitation with light. The associated characteristic pattern of intense electromagnetic hot-spots can be exploited to enhance light-matter interaction and to enable strong coupling of distinct cavities over a micrometer distance [1] or to strongly couple single excitons and plasmons at room temperature [2]. Here, we disentangle the local field dynamics of individual hot-spots within a nanoslit resonator with a spatial resolution of <10 nm by combining time-resolved photoemission electron microscopy (PEEM) and a 1 MHz NOPA system. Interestingly, we detect local differences of rather global properties such as the *Q*-factor and resonance frequency. By using the concept of quasinormal modes we explain these notable local differences, which will be experienced by, e.g., quantum emitters, with a non-negligible influence of adjacent resonator modes.

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Ultrafast Control of Quantum Systems via Strong Terahertz-Nearfields

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In this project, we want to explore field-induced dynamics in luminescent 1D- and 2D-quantum materials. We are interested in field-driven dynamics that occur instantaneously with a strong biasing E-field and in the form of field-induced modifications of fluorescence after optical excitation. In particular, we focus on the impact of strong-field charge injection and carrier acceleration within the micro-environment of the nanoprobe. Experimentally, we combine strong-field THz- and mid-infrared biasing with tailored nanostructures, enabling the local application of sub-picosecond electric fields to embedded quantum systems, sketched in Figure 1. The study is expected to advance the control schemes of optical properties in quantum systems and to provide new means for detection of local electric fields and charges via the sensing of visible photons.

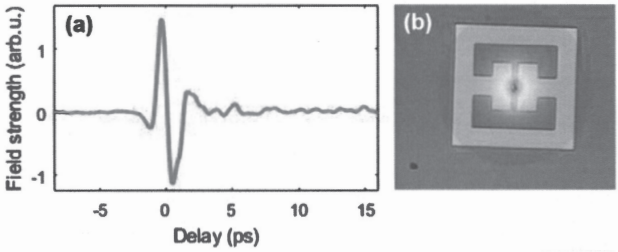

Figure 1: (a) Single-cycle THz waveform for strong-field biasing, recorded via electro-optic sampling. **(b)** Microstructure to enhance local fields with a sketched quantum emitter embedded in the gap.

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Switchable cavity-assisted energy transfer in the infrared via surface phonon polaritons

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Devices providing switchable energy and/or signal transfer in the infrared frequency range are essential to realize novel integrated photonic circuits. In the infrared and terahertz regime, Silicon-Carbide (SiC) supports the low-loss surface phonon polaritons. Based on finite difference time domain simulations we investigate high-quality tunable photonic modes in an elliptical metal–SiC heterostructure cavity which serves as a low-loss transfer channel for efficient coupling between two spatially separated locations in the cavity. Tuning of such modes in the Reststrahlen band of SiC is achieved via the longitudinal optical phonon resonance that depends on the carrier concentration. We demonstrate photonic mode shifts larger than 20 times the mode line width and conclude that these cavity modes are well suited to realize optically switchable resonant energy transfer channels in the range between 24 – 29 THz.

Influence of transient cavity degeneration on strong coupling in hybrid exciton-plasmon nanorods

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In strongly coupled exciton-plasmon systems the plasmon resonator presents the dominating loss channel, expressed by a short plasmon lifetime in the range of some tens of femtoseconds [1]. When exciting nano-plasmonic systems by femtosecond pulses, the energy dissipated via the plasmonic losses makes the situation even worse. The dissipation of the plasmon into high energy electrons and the associated heating of the electronic system, leads to a further transient reduction of the cavity quality. This effect must be considered in any experiment trying to measure the transient behavior of "plexcitons". However, a full simulation of the optical spectra of transiently excited strongly coupled exciton-plasmon systems has not been presented in literature so far.

Here we present an investigation on the influence of nanoparticle heating in the strong coupling regime in TDBC coated gold nanorods [2][3]. By selective excitation of the nanoparticle core we measured the transient behavior broadening of the coupled resonances. We simulated the influence of the nanoparticle temperature on the differential absorption by a modified Mie-Gans-Theory in combination with a temperature-dependent permittivity. In order to achieve a good fit to experimental data, saturation of the TDBC absorption by the electric vacuum field had to be included. Fluence dependent measurements show a transient broadening of the coupled resonances by the generation of nanoparticle heat as well as a decrease of the Rabi splitting width with increasing pump intensity.

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Spectroscopy of Graphene Nanoribbons

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One-dimensional (1D) graphene nanoribbons (GNRs) are promising materials for future electronics and optoelectronics. Different from graphene, armchair-edge GNRs (AGNRs) are semiconducting with a direct bandgap, whose size can be tuned through the width of the ribbon. Until now the photophysical characterization of AGNRs has not been addressed properly due to the metal substrate on which they are grown. We have during the last three years applied fluorescence and absorption spectroscopy [1], and plasmonic enhancement [2,3] to reveal the fundamental optical properties of 7-atom wide AGNRs. We have mapped the spectral positions of the excitons in 7-AGNRs and shown how dark excitons influence their emission properties. A key result of our work is the observation that inducing defects to the ribbons can be used to significantly boost the fluorescence. By using plasmonic enhancement, even single photon emission can be obtained from isolated ribbons.

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Near field induced speedup of exciton diffusion in light-harvesting arrays

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The observation of long-range energy propagation along nano-engineered arrays of bacterial antenna complexes has been recurrently addressed as a result of ballistic, hence, coherent propagation. We show that the experimentally determined quantities relevant for these dynamics hinder the formation of large coherent domains and inevitably lead to classical diffusion in these densely packed arrays, which nevertheless benefit from the spectral complexity of single antenna complexes and near field interactions to result in the propagation observed. We show that the packing density bridges the apparent incompatibility between observed energy transfer rates in bacterial photosynthetic membranes and nano-engineered antenna arrays, which together with exciton delocalization across individual antennas and non-equilibrium transport, provides useful design strategies to optimize energy transfer in the presence of typical room temperature conditions.

Enhanced radiative plasmon decay and superluminescence from a nanoscale plasmonic cavity

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In the nanoscale cavity of an optical antenna formed between a sharp gold tip and a flat gold substrate covered with a monolayer of Raman active molecules, coupled plasmon oscillations can be excited optically leading to enhanced photoluminescence (TEPL) and Raman scattering (TERS) [1-3]. Alternatively, the coupled plasmon oscillation can also be excited purely electrically by inelastic electron tunneling with an increased bias voltage applied between the tip and the substrate, leading to electroluminescence. Combining both excitation schemes can lead to almost three orders of magnitude higher electron-to-photon conversion efficiency than it is with inelastic tunneling alone [2]. The emission spectra recorded from the nanoscale plasmonic cavity as a function of bias voltage and excitation laser power show a significant intensity increase that is associated with spectral narrowing. This observation suggests that radiative eh recombination is feeding photons back into the cavity by stimulated emission resonant with the gap mode in analogy to a superluminescent diode [3].

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Quantum interference during the electron emission from surface plasmon polaritons

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During nonlinear photoemission electrons are liberated from a solid by simultaneous or subsequent absorption of multiple photons. Recently it was demonstrated that electron emission can also be achieved by nonlinear absorption of surface plasmon polaritons (SPPs) alone. The absorption of either photons or SPPs may only be considered independent if both are separated in space and time. If this is not the case, the electron emission arises from the nonlinear absorption of quanta from the coherent superposition of the light's and the SPP's electromagnetic fields at the surface.

To disentangle the individual contributions of photons and SPPs we employ pump-probe photoemission electron microscopy (PEEM) in normal-incidence geometry. We record the spatially modulated electron yield that arises from the interference of an isolated femtosecond SPP pulse and a probing femtosecond laser pulse with nanometer resolution. A Fourier analysis of the PEEM images enables us to separate all relevant electron emission pathways and we find evidence for quantum interference of SPP and photon absorption. By independently adjusting the strength of the SPP and the laser field we can directly assign the quantum contributions of photons and SPPs to each unique electron emission pathway.

Theoretical Description of Two-Dimensional Spectroscopy in a CdTe Quantum Dot doped with a Single Mn Ion

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Quantum dots (QDs) doped with a single magnetic ion offer new opportunities in the field of spintronics and quantum information [1]. In such applications knowledge of decoherence and radiative life-times, fine-structure splitting and mechanisms of coherent coupling is of crucial importance. Non-linear optical measurements like four-wave mixing spectroscopy are able to provide such insights [2].

In Mn doped QDs the Mn ion behaves as a spin M= 5/2 with six eigenstates Mz=$\pm$5/2, $\pm$3/2, $\pm$1/2 resulting in a photoluminescence (PL) spectrum consisting of six equally spaced lines even in the absence of a magnetic field. By increasing the magnetic field, states which are coupled by the exchange interaction are brought into resonance and multiple anticrossings are observed in the PL spectrum. However, using PL alone one cannot unambiguously distinguish if these resonances arise from different non-interacting states or from a coupled system. In contrast, two-dimensional (2D) four-wave mixing (FWM) spectroscopy allows us to disentangle involved spectra.

In this contribution we study theoretically multipulse excitations to probe the coherence dynamics of a QD doped with a single Mn ion. We model the system as an ensemble of QDs in different Mn spin states, whereby each member is given by a few-level system. Our study shows that 2D FWM is able to identify and investigate coherent coupling mechanisms between the excitonic excitations and the magnetic dopant in the QD.

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Towards the investigation of ultrafast nanoscale quantum phenomena by superfocusing plasmonic nanotips

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The investigation of ultrafast nanoscale quantum phenomena in the near field requires dedicated experimental methods, which allow controlling the optical nearfield on femtosecond time scales to manipulate and enhance its interaction with quantum objects, as e.g. quantum dots or vacancy centers. Plasmonic superfocusing at metallic nanotips offers ultra-strong nearfield confinement combined with the versatility of a tip scanning method. The precise positioning control of these tips enables their application particularly to the study of quantum systems, which can't yet be fully integrated into the nanotechnologies realizing the complete system. Moreover, when the superfocusing plasmon is coupled resonantly to the low loss guided modes of an optical fiber efficient far-to-nearfield coupling is achieved with ultralow noise background in the volume of the quantum object. Hence, plasmonic nanotips are considered an indispensable tool for the systematic study of quantum phenomena in nanosystems. We will report about our current progress towards the realization of such plasmonic tips and their performance parameters as well as about our research vision to integrate structure and property resolving measurement techniques with these tips.

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Hot-spots and plasmonic near-fields in gold nanosponges

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Gold nanosponges, i.e. porous gold particles of hundreds of nm size, can be used as an easy-to-produce versatile test-bed for light-matter coupling in strongly enhanced plasmonic fields. The intrinsically disordered porous particles can be considered as an ensemble of hundreds or thousands of tiny local nano-antennae [1]. Their geometrical structure - and thus the statistical distribution of antennae properties - can be optimized by metallurgical tricks.

For any given excitation conditions, almost all possible local modes will either not be excited or hybridize too strongly to collective electromagnetic modes of the spongy particle to be useful. However, there will always be some 'hot spots' for which the conditions are 'incidentally' 'just right': On the one hand, nanoscale-energy transfer from collective Mie-type modes into these hot-spot modes is sufficiently efficient to funnel a noticeable part of the total energy into highly localized modes of ~20 nm size. On the other hand, the mode volume is very small and Purcell factors of single hot-spot modes reach up to 106 [2]. Strong ultrafast electron emission confirms experimentally the predicted strong light-matter coupling in the hot spots [1].

* *based on experiments from the Lienau group (Institut für Physik and Center of Interface Science, Carl von Ossietzky Universität Oldenburg, Germany) and samples from the Schaaf group (Institut für Mikro- und Nanotechnologien MacroNano® and Institut für Werkstofftechnik, Technische Universität Ilmenau, Germany)*

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Photon-pair generation mediated by coupling of atoms to nonlinear photonic nanostructures

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We investigate hybrid nonlinear systems of an atom-like emitter coupled to a nonlinear photonic nanostructure [1]. Using the spectral selectivity and the fermionic nature of the atom's transition together with the ability to shape the optical density of states (DOS) in nanostructures, we suppress the individual responses of the two constituents to achieve regimes where the response of the hybrid system is dominated by coupling-induced nonlinear effects. In particular, photonic nanostructures allow to realize spectral gaps with zero DOS, such that nonlinear effects like spontaneous photon-pair generation are prohibited if the frequency of one of the generated photons is in a gap. Using a rigorous Green's function quantization method to model the hybrid source, we show that a coupled atom with a transition frequency in the gap of the photonic structure mediates the otherwise forbidden generation of exactly one photon pair, where the idler photon directly excites the emitter and the signal photon occupies an optical mode. This effect can serve as a sensitive indicator for the presence and excitation of single atoms. Furthermore, this scheme could be used to implement quantum memories or deterministic and tunable single-photon sources.

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First steps towards long-range coherent energy transfer in plasmonic lattices

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Taking photosynthesis as an example, light harvesting is based on an efficient energy guiding of electronic excitations to reaction centers. This is mediated by near-field coupling of transition dipole moments and limited to a few nanometers. The goal of our project is to scale the coherent energy transfer up to the micrometer range using plasmonic lattices. The Bragg grating generates a spatially extended mode while the plasmonic hotspots boost light-matter interaction locally. Coherent energy transfer then results from the strong coupling between the hybrid plasmon lattice mode and emitters. Here we present our first measurements on the dispersion relations of surface lattice resonances in gold nanoparticle arrays and their interaction with emitters.

Polyanionic Silver Clusters studied by Photoelectron Spectroscopy

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Abstract

Multiple negatively charged silver clusters Ag_N^{z-} ($N = 40 - 800$, $z = 1 - 6$) have been investigated in order to study the electronic and optical properties in a so far not explored parameter range, i.e., in particular for those complexes where a part of the electron ground state density is confined above the vaccuum level by the presence of a repulsive Coulomb wall. Tunable laser pulse excitation in combination with electron spectroscopy is applied to extract photoeelectron spectra and photodetachment efficencies. In the analysis, tunneling through the Coulomb barrier after photoexcitation has to be taken into account. We found evidence for plasmonic motion in all selected polyanionic silver clusters. The development of the plasmon energies as function of size agree well with former work on the Ag_N^+, Ag_N and Ag_N^- and highlights the impact of electron spill-out and reduced *s-d* interaction at the surface on the optical activity in silver polyanions.

Circularly polarized light-field driven current in solids and air

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The electro-magnetic field of few-cycle laser pulse with precisely controlled carrier-envelope phase (CEP) has been widely used to explore the nonlinear response of materials under strong field excitations. In gas-phase media, the response can be well described by the tunneling ionization process; while in condensed matter, the situation is more diverse due to the complex charge carrier dynamics in the context of crystal band structures. Experiments show that ultra-short laser pulses can generate a finite current in dielectrics and wide bandgap semiconductors, which can be understood as the strong-field induced semi-metallization due to the Wannier-Stark localization in the material [1]. On the one hand, this provides a new method, in addition to optical signal detection or photoelectron detection, to measure the CEP correlated photocurrent in actual electronic devices. On the other hand, the CEP correlated signal measured in this kind of devices could also be used to precisely determine the actual CEP of a single few-cycle laser pulse, which usually requires a sophisticated vacuum apparatus. Here, we explore the possibility of using a circularly polarized light for single-shot stereo-ATI phase-meter, which has a series of advantages over linearly polarized excitations [2]. We demonstrate experimentally that circularly polarized few-cycle pulses induces photocurrent in both solid state (SiO_2) and in air-gap quadrupole electrode devices. The photocurrents measured with our devices exhibit CEP, polarization and light intensity dependence. Our results provide a novel scheme for the measurement of the CEP of few-cycle laser pulses.

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