Q 27: Nano-Optics III

Time: Tuesday 14:30–16:30 Location: P 11

Q 27.1 Tue 14:30 P 11

Real-time propagation of a Na_{297} dimer as a coupled Maxwell-Schrödinger and time-dependent Kohn-Sham-Maxwell system — •Rene Jestaedt[1,2], Heiko Appel[1,2], and Angel Rubio[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Max-Planck-Institut für Struktur und Dynamik der Materie, Hamburg, Germany

External electromagnetic fields can induce non-negligible electric currents which influence the total Maxwell fields inside molecular systems. This backreaction affects the conductivity and the optical properties of molecular and nanoplasmonic systems. In the present work, we employ the Riemann-Silberstein vector of the electromagnetic field to cast Maxwell's equations into a spinor representation similar to the Dirac equation [1]. This representation allows us to use standard unitary propagation techniques [2], both for Maxwell's equations and the time-dependent Kohn-Sham equations. To illustrate our novel implementation of the coupled Maxwell-Kohn-Sham equations in the real-space code octopus [3], we show the effects of a large matter feedback to the Maxwell fields and vice versa a radiation feedback to electrons and nuclei for a Na_{297} nanoparticle.

[1] I. Bialynicki-Birula, Progress in Optics **36**, 245-294, (1996).
[2] A. Castro et al., J. Chem. Phys. **121**, 3425-3433, (2004).
[3] X. Andrade et al., J. Phys. Cond. Mat. **24**, 233202, (2012).
[4] R. Jestädt et al., (submitted)

Q 27.2 Tue 14:45 P 11

100-fold Enhancement of Spontaneous Emission from a Single Quantum Dot by a Gold Nanocone Antenna — •Hsuan-Wei Liu[1], Korenobu Matsuzaki[1], Stephan Götzinger[2,1], and Vahid Sandoghdar[1,2] — [1]Max Planck Institute for the Science of Light, Erlangen, Germany — [2]Friedrich Alexander University of Erlangen-Nürnberg, Erlangen, Germany

Control and enhancement of spontaneous emission has been an intriguing and active topic of research in the past few decades. Previously, we have theoretically suggested that a plasmonic gold nanocone antenna is promising for achieving spontaneous emission enhancements exceeding 1000 while maintaining a high quantum efficiency [1]. Here, we report on first experiments corresponding to this concept. The gold nanocones were fabricated by focused ion beam milling on a glass substrate [2]. By positioning a single semiconductor quantum dot with nanometer accuracy in the near field of the nanocone antenna, we observed large lifetime reductions for both monoexciton and biexciton emission. By performing saturation studies and analysing the photon statistics, we determined the radiative decay rate enhancement of the monoexciton and biexciton emission to be 109 and 100 folds with quantum efficiencies of 60% and 70%, respectively [3]. Such large enhancements open the door to many applications in light emitting technologies and fundamental research. [1] X. Chen, M. Agio, and V. Sandoghdar, *Phys. Rev. Lett.* **108**, 233001 (2012). [2] B. Hoffmann *et al.*, *Nanotechnology* **26**, 404001 (2015). [3] K. Matsuzaki *et al.*, *arXiv.*:1608.07843 (2016).

Q 27.3 Tue 15:00 P 11

Coherent interaction between a single molecule and a plasmonic nanoantenna — •Johannes Zirkelbach[1], Benjamin Gmeiner[1], Tobias Utikal[1], Stephan Götzinger[1,2], and Vahid Sandoghdar[1,2] — [1]Max Planck Institute for the Science of Light (MPL), D-91058 Erlangen, Germany — [2]Department of Physics, Friedrich-Alexander-University of Erlangen-Nürnberg, D-91058 Erlangen, Germany

We report on the coherent interaction of light with a coupled system consisting of a single quantum emitter and a plasmonic nanostructure. Here, we combine cryogenic high-resolution spectroscopy with localization microscopy to study single molecules in the vicinity of isolated plasmonic nanostructures embedded in a thin organic matrix. We discuss the spectral and spatial behavior of the extinction spectra recorded from the composite molecule-antenna system, leading to Fano profiles in the far-field interference signal. Future experiments will aim to observe transparency (cloaking) and ultrastrong absorption [1].

[1] X. Chen, V. Sandoghdar, and M. Agio, Phys. Rev. Lett. 110, 153605 (2013)

Q 27.4 Tue 15:15 P 11

Exceptional mode organization in a resonator microcavity based on a hyperbolic metamaterial — •Evgenij Travkin, Sascha Kalusniak, Sergey Sadofev, and Oliver Benson — Humboldt-Universität zu Berlin, Institut für Physik, Newtonstraße 15, 12489 Berlin

Metamaterials offer a variety of exciting possibilities for manipulation of light among which are i.e. the control of phase and group velocity and negative refraction. We investigate a hyperbolic metamaterial (HMM) based on stacked layer pairs of epitactically grown ZnO/ZnO:Ga embedded in a resonator microcavity. The highly anisotropic and frequency dependent HMM refractive index enables a unique distribution of resonant modes. Several modes of the same order can exist at different frequencies and the relative spectral positions of higher and lower order modes can interchange resulting in an anomalous mode organization. We present experimental spectra demonstrating unconventional mode emergence and reversal of the mode order in differently scaled HMM-based cavities and supplement them by transfer matrix calculations and dispersion relations derived from the cavity roundtrip condition. Our system can be fully tailored by tuning of the layer thickness ratio and doping level of the HMM.

Q 27.5 Tue 15:30 P 11

Coherent coupling of a single molecule to a scanning Fabry-Pérot microcavity — •Daqing Wang[1], Hrishikesh Kelkar[1], Diego Martin-Cano[1], Tobias Utikal[1], Stephan Götzinger[2,1], and Vahid Sandoghdar[1,2] — [1]Max Planck Institute for the Science of Light, D-91058 Erlangen, Germany — [2]Friedrich Alexander University Erlangen-Nuremberg, D-91058 Erlangen, Germany

We report on the coherent coupling of a single organic molecule to a scannable, tunable and broadband microcavity. The cavity consists of a planar distributed Bragg reflector and a micromirror with a radius of curvature of 5 μm fabricated with focused ion-beam milling and coated with silver. By integrating an organic matrix in the microcavity at liquid helium temperature, we are able to coherently couple individual molecules to the single mode of the cavity. Our results show that a single molecule can attenuate 38% of the light stored in the cavity. We also demonstrate four-fold improvement of single-molecule stimulated emission compared to free-space coupling in a tight focus. Our experimental approach based on a microcavity with low mode volume and low quality factor paves the way for the realization of a series of nonlinear and collective quantum optical effects.

Q 27.6 Tue 15:45 P 11

Extreme single-emitter interaction with two-coupled bad resonators — •Burak Gurlek, Vahid Sandoghdar, and Diego Martin-Cano — Nano-Optics Division, Max Planck Institute for the Science of Light, Erlangen, Germany.

Nonlinear photon generations require strong interactions between light and matter. Nanoantennas and optical cavities are two complimentary and common approaches for enhancing these interactions at extreme levels, the first relying on extremely small volumes whereas the second on small bandwidths (high Q-factors). Recently, it has been proposed that hybrid combinations between nanoantenna and a high Q cavity can further enhance the spontaneous emission of a single quantum emitter [1, 2]. Here, we explore a hybrid system based on a low-Q cavity and a nanoantenna, leading to non-intuitive additional enhancements and quenching behavior. Taking a confocal cavity and a nanocone as an example, we make use of the Quasi Normal Mode (QNM) approach [3] to study the interference mechanism that creates these interesting properties. Our results provide new possibilities for improving the efficiency of single emitters and entering extreme regimes of photophysics. References: [1] Y. Xiao et al., Phys. Rev. A 85, 031805(R) (2012). [2] H. M. Doeleman et al., ACS Photonics 3 (10), 1943-1951 (2016). [3] C. Sauvan et al., PRL 110, 237401 (2013).

Q 27.7 Tue 16:00 P 11

Strong coupling between surface plasmon polaritons and molecular vibrations — •Hala Memmi, Sascha Kalusniak, Sergey Sadofev, and Oliver Benson — Institut für Physik, Humboldt Universität zu Berlin

The hybridization of different quasi-particles is an extensively studied subject; both from a practical as well as fundamental point of view.

The resultant hybrid excitations exhibit new properties that are not available by the isolated constituents.

Here, we report on hybridization of surface plasmon polaritons and molecular vibrations in an organic/inorganic plasmonic hybrid structure. The structure consists of a poly-vinyl-methyl-ketone layer deposited on top of a silver layer. Attenuated-total-reflection measurements allow us to reconstruct the dispersion relation of the hybrid. The system exhibits two polariton branches clearly demonstrating anticrossing behavior in vicinity of the carbonyl stretching vibration of the polymer with the coupling strength of 14 meV. Systematic tuning of the carbonyl group density confirms the square root dependence of the energy splitting. We also present some of the characteristics of this hybrid species and discuss its potential applications.

Q 27.8 Tue 16:15 P 11

Attosecond time-resolved photoelectron spectroscopy of hybridnanoresonators — Julia Hengster and •Thorsten Uphues — CFEL, Attosecond Research and Science Group, Hamburg University, Luruper Chaussee 149, 22761 Hamburg

Understanding plasmons as collective oscillations of the free-electron gas density important questions related to their propagation, damping, charge and energy localization came up. Nevertheless the behaviour of hybrid nanostructures approaching the monolayer limit raises a new type of questions concerning their plasmonic behaviour in space and time, following the complex dynamics of the electromagnetic field. Attosecond time-resolved experiments are on the way to resolve subcycle electron dynamics from plasmonic interaction of ultrashort driving pulses in surfaces and nanostructures. Our approach of attosecond photoscopy demonstrates a reliable route to extend attosecond technology to surface and nanostructure dynamics. Hybrid nanostructures exhibit complex plasmonic properties sensitive to parameters as geometrical aspect ratios or material compositions. Vertically aligned disk nano-resonators belong to a group of tailored systems demonstrating field enhancement with strong localization. We found a remarkably sensitive behaviour in the coupling of surface and bulk plasmons of the resonators with ultrafast subcycle dynamics. As a proof-of-concept we demonstrate attostreaking from gold films with significant deviation to gas-phase streaking. Furthermore we developed non-destructive preparation procedures for nanoparticle samples as a basic requirement for attosecond photoscopy.