Timetable

All talks are held in **Room 1'101**, Department of Physics, Newtonstraße 15, 12489 Berlin.
Coffee, lunch breaks, and dinner buffet are served in the **foyer** (ground floor).

Wednesday, August 31st	
09:00-09:15	Opening Remarks by Alexander Govorov (Ohio U) **Short review of the key points of the NANOPLAS Workshop**
09:15-10:15	Alexander Govorov (Ohio U) **Quantum theory of energetic (hot) plasmonic electrons and the role of hot spots in metal nanocrystals**
10:15-11:30	Garnett W. Bryant (NIST and U Maryland) **Approaching the quantum limit for nanoplasmonics**
11:30-11:45	Coffee break
11:45-13:00	N. Asger Mortensen (TU Denmark) **Nonlocal plasmonics: Beyond classical local-response electrodynamics**
13:00-14:30	Lunch buffet
14:30-15:45	F. Javier García de Abajo (ICFO and ICREA) **Graphene plasmons, quantum emitters, and light modulation at the nanoscale**
15:45-16:15	Coffee break
16:15-17:30	Kurt Busch (Humboldt U) **Theoretical aspects of quantum plasmonics in realistic systems**
17:30-18:15	Marten Richter (TU Berlin) **Plasmon-/cavity photon-emitter coupling: Non-perturbative numerical models and perspectives for non-canonical quantization**
18:30	Poster session and dinner buffet

Thursday, September 1st	
09:00-10:15	Javier Aizpurua (Center for Material Physics) **The quantum realm of nanoplasmonics for ultraresolution and active control of optoelectronics**
10:15-10:45	Coffee break
10:45-12:00	Stephen K. Gray (Argonne National Laboratory) **Quantum dynamics model for hybrid quantum dot/plasmonic systems**
12:30-14:00	Lunch buffet
14:00-15:15	Stephen Hughes (Queen's U) **Modelling the quantum optical properties of quantum-dot plasmonic resonator systems**
15:15-15:45	Coffee break
15:45-16:30	Yuan Zhang (Aarhus U) **Quantum theory for plasmonic nano-laser with multi-level emitters: Optical and electric pumping**
16:30-16:45	Closing remarks
17:00	Sightseeing tour

Abstracts of Invited Speakers

Quantum theory of energetic (hot) plasmonic electrons and the role of hot spots in metal nanocrystals

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The effect of generation of energetic (hot) electrons is a quantum phenomenon that occurs due to breaking of the momentum conservation of electrons. In small nanocrystal, this process occurs at the surfaces and in the hot spots [1-4]. To describe this class of quantum phenomena, we developed a novel kinetic approach (Kinetic DFT) that includes both the KS-equation and the master kinetic equation for the temporal evolution of the one-electron density matrix [4]. In the following steps, we applied this approach to several experimental realizations [5-8]. The efficiency of generation of energetic plasmonic carriers in metal nanostructures strongly depends on the optical design and material composition. Our studies demonstrated the ability to generate large numbers of hot plasmonic carriers in specially-designed hybrid nanostructures [5-8]. The hot-electron generation becomes especially efficient in plasmonic nanostructures with electromagnetic hot spots [3,5,8]. The energy distributions of optically-excited plasmonic carriers are very different in metal nanocrystals with large and small sizes [2]. For metal nanocrystals with smaller sizes or in nanostructures with hot spots, the excited state gets a large number of carriers with high energies [2,3,4]. Optical generation of electrons with high energies can be observed using ultrafast time-resolved spectroscopy, photocurrents and photochemistry [5-8].

[1] A. O. Govorov, H. Zhang, V. Demir, and Y. K. Gun'ko, **NanoToday**, 9, 85 (2014).

[2] A. O. Govorov, H. Zhang, Y. K. Gun'ko, **J. Phys. Chem. C**, 117, 16616 (2013).

[3] H. Zhang and A. O. Govorov, **J. Phys. Chem. C**, 118, 7606 (2014).

[4] A.O. Govorov and H. Zhang, **J. Phys. Chem. C**, 119, 6181 (2015).

[5] H. Harutyunyan, A. B. F. Martinson, D. Rosenmann, L.K. Khorashad, L.V. Besteiro, A.O.Govorov and G.P. Wiederrecht, **Nature Nanotechnology**, 10, 770–774 (2015).

[6] W. Li, Z. J. Coppens, L. Vázquez, W. Wang, A. O. Govorov and J. Valentine, **Nature Communications**, 6, 8379 (2015).

[7] L. Weng, H. Zhang, A. O. Govorov and M. Ouyang, **Nature Communications**, 5, 4792 (2014).

[8] A. Sousa-Castillo, M. Comesaña-Hermo, B. Rodríguez-González, M. Pérez-Lorenzo, Z. Wang, X.-T. Kong, A. O. Govorov, and M. A. Correa-Duarte, **J. Phys. Chem. C**

Approaching the quantum limit for nanoplasmonics

Garnett W. Bryant

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Hybrid systems of metallic nanoparticles (MNP) coupled to semiconductor quantum dots (QD) have drawn intense interest for the novel quantum optics which is possible when these quantum systems are strongly coupled and strongly interacting with light. A full quantum description of the entire system, treating the MNPs and QDs on an equal footing, is needed to account for size quantization, quantized excitations, coherent coupling, interparticle tunneling and nonlocal and nonlinear response. To achieve this, a quantum description of the nanoplasmonics is needed.

To this end, we have used real-space time-dependent density functional theory (TDDFT) for a quantum description of MNPs and linear chains of atoms. We discuss recent detailed analysis of the time dependence of driven MNPs and linear atomic chains to characterize fully these excitations. Modes have a "sloshing" character with charge oscillating between filled energy shells just below the Fermi level and empty shells just above the Fermi level and, at the same time, an "inversion" character with charge continuously emptying from levels far below the Fermi level and filling shells far above the Fermi level. The sloshing character is dominant in classical surface plasmon modes. The inversion character is more single-particle like and is dominant for other modes. This analysis allows us to identify the plasmonic modes.

While TDDFT yields information about the nature of the excitations in MNPs and linear chains, DFT can't address the quantum character of these excitations, ie whether the excitations are harmonic-like, bosonic, fermionic. To begin to address these issues, we have explored simple models for interacting electrons on a linear chain. Linear atomic chains, such as atom chains on surfaces, linear arrays of dopant atoms in semiconductors, or linear molecules, provide ideal testbeds for studying collective, plasmonic excitations in the quantum limit. We find the many-body spectra of finite (up to 25) atom chains. This allows us to identify the quantum plasmonic excitations of these atomic-scale systems.

Highly correlated, multi-excitonic states, strongly dependent on the electron-electron interaction strength, dominate the exact spectral response for short chains. The ubiquitous presence of excitonic many-body states in the spectra makes it hard to identify plasmonic excitations. A combination of criteria involving a many-body state's transition dipole moment, balance, transfer charge, dynamical response and induced charge distribution do strongly suggest which many-body states should be considered as plasmonic. This analysis is used to reveal the few plasmonic many-body states hidden in the dense spectrum of low-energy single-particle-like states and many higher-energy excitonic-like states. Excitonic states are the predominant excitations because of the many possible ways to develop local correlations. The plasmonic states are most apparent for strong interaction. Results for longer chains show how the importance of plasmonic excitations changes

with the increasing number of electrons and how systems start to evolve to the long chain limit where Luttinger liquid states should appear. With the identification of the plasmonic excitations, the quantization of the plasmonic near field and coupling to quantum light fields is determined to assess the quantum character of single- and multi-plasmonic excitations. The quantization of the near-field is compared with traditional models of field quantization to better understand the quantization of these polaritonic excitations. The influence of Pauli blockade, critical for the dynamics of 1D systems, is studied by also considering pairs of coupled chains, where blockade has less influence on the system dynamics. The effect of spin is also considered.

Nonlocal plasmonics: Beyond classical local-response electrodynamics

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I will discuss ongoing experimental and theoretical efforts to explore the plasmonic response in metallic nanoparticles, where nanometric dimensions are expected to promote a nonlocal plasmonic response beyond that of the usual classical electrodynamics. I will review efforts to make semi-classical hydrodynamic extensions [1,2], as well as our developments beyond these simplified models [3,4]. I will also address our most recent single-particle EELS observations of high-order modes in silver nanoparticles [5]. I will discuss insight from single-particle spectroscopy versus ensemble-averaged properties. As an intriguing example, nonlocal response may potentially promote inhomogeneous broadening even deep inside the quasi-static regime [6], where classical electrodynamics would otherwise predicts a size-independent dipole resonance, i.e. homogenous broadening only. Finally, I will discuss means to quantify the degree of nonclassical effects from an energy perspective [7].

[1] N.A. Mortensen, S. Raza, M. Wubs, T. Sondergaard, and S.I. Bozhevolnyi, **Nature Communications**, 5, 3809 (2014)

[2] S. Raza, S.I. Bozhevolnyi, M. Wubs, and N.A. Mortensen, **J. Phys. Cond. Matter**, 27, 183204 (2015)

[3] G. Toscano, J. Straubel, A. Kwiatkowski, C. Rockstuhl, F. Evers, H. Xu, N. A. Mortensen, and M. Wubs, **Nature Communications**, **6**, 7132, (2015)

[4] W. Yan, M. Wubs, and N. A. Mortensen, **Phys. Rev. Lett.**, 115, 137403 (2015)

[5] S. Raza, S. Kadkhodazadeh, T. Christensen, M. Di Vece, M. Wubs, N.A. Mortensen, and N. Stenger, **Nature Communications**, 6, 8788 (2015)

[6] C. Tserkezis, J.R. Maack, Z. Liu, M. Wubs, and N.A. Mortensen, **arXiv**:1602.00874

[7] W. Yan and N.A. Mortensen, **Phys. Rev. B**, 93, 115439 (2006)

Graphene plasmons, quantum emitters, and light modulation at the nanoscale

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Plasmons in highly doped graphene are highly tuneable by external means (electrically, thermally, magnetically, nonlinearly) and exhibit record levels of confinement. Consequently, the interaction with optical quantum emitters is extraordinarily intense, rendering these excitations an excellent platform for the implementation of quantum optics phenomena at the nanometer scale. In particular, we predict that the strong-coupling regime is reached at the single plasmon level. In this presentation, we will discuss the physics and phenomenology of quantum optics phenomena enabled by graphene plasmons and their application to light modulation at the nanoscale.

Theoretical aspects of quantum plasmonics in realistic systems

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Modern experiments and technological applications require an accurate description of the interaction between radiation and matter. Depending on the kind of system under consideration, this interaction may need to be described using a full quantum treatment. While a quantum description of the electromagnetic field can be found in many textbooks, the description of matter and its interaction with radiation encounter several difficulties as soon as the microscopic world (e.g. single charges, simple atoms) is abandoned in order to proceed to mesoscopic or even the macroscopic regimes. The effective description of systems with a large number of degrees of freedom as well as the consideration of associated phenomena such as dissipation turn out to be far from trivial a has led to different (but mostly equivalent) approaches.

In this talk, we focus on a specific aspect of the light-matter interaction, namely the existence of resonances induced by the collective behavior of conduction electrons in plasmonic systems. We provide a (necessarily biased) review of certain conceptual aspects and developments as well as numerical and analytical techniques which provides an approach to light-matter interaction in plasmonic systems within a quantum-mechanical framework.

Plasmon- / Cavity photon -emitter coupling: Non-perturbative numerical models and perspectives for non canonical quantization

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The quantum realm of nanoplasmonics for ultraresolution and active control of optoelectronics

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The optical response of plasmonic nanosystems has been traditionally well described within the framework of local dielectric response theory. However, practical configurations in surface-enhanced microscopies and spectroscopies are reaching the possibility to establish subnanometric control of metallic morphologies and architectures which expose the importance of addressing the quantum nature of electrons and photons to correctly describe a complex variety of optoelectronic processes that give rise to ultraresolution, nonlinearities and active control of the optical response in metallic nanoenvironments. In this talk, the interplay between electronic transport and optics will be introduced, with a description of the quantum nature of the electron gas within the time-dependent density functional theory (TDDFT) [1,2]. Furthermore, atomistic quantum calculations of plasmonic configurations will be shown to provide extreme electric field confinements opening the door to ultraresolution [3,4]. Finally, the possibility to modify the optical response of plasmonic structures by application of an external DC bias [5], and by particle charging [6] will be also theoretically addressed in the context of active control of nanoplasmonics.

[1] R. Esteban, A. G. Borisov, P. Nordlander, and J. Aizpurua, **Nature Communications**, 3, 825 (2012).

[2] D.C. Marinica, A.K. Kazansky, P. Nordlander, J. Aizpurua, and A. G. Borisov, **Nano Letters**, 12, 1333 (2012) .

[3] M. Barbry, P. Koval, F. Marchesin, R. Esteban, A. G. Borisov, J. Aizpurua, D. Sánchez-Portal, **Nano Lett.**, 15, 3410 (2015).

[4] F. Marchesin, P. Koval, M. Barbry, J. Aizpurua, D. Sánchez-Portal, **ACS Photonics**, 3, 269 (2016).

[5] D. C. Marinica, M. Zapata, P. Nordlander, A. K. Kazansky, P. M. Echenique, J. Aizpurua, A. G. Borisov, **Science Advances**, 1, e1501095 (2015).

[6] M. Zapata, J. Aizpurua, A. K. Kazansky, and A. G. Borisov, **Langmuir**, 32, 2829-2840 (2016).

Quantum Dynamics Model for Hybrid Quantum Dot/Plasmonic Systems

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A time-dependent quantum density matrix formalism for one or more quantum dots interacting with a plasmonic system is discussed [1]. The model, an extension of previous work on a single quantum dot interacting with a plasmonic system [2], includes dephasing, spontaneous emission and dissipation. The strengths and limitations of the formalism are emphasized, as well as computational approaches for obtaining time-dependent and frequency-resolved information. Numerical simulations focusing on the generation of bipartite quantum entanglements [3] in the cases of two and three quantum dots interacting with a plasmonic system are discussed, and an analytical solution for an arbitrary number of quantum dots, but in the limit of no quantum dot dephasing, is presented. Possible extensions of the approach to other problems, including those involving nitrogen-vacancy centers [4] as opposed to quantum dots, are discussed.

[1] M. Otten et al., **Phys. Rev. B**, 92, 12543 (2015).

[2] R. A. Shah et al., **Phys. Rev. B**, 88, 075411 (2013).

[3] W. K. Wootters, **Phys. Rev. Lett.**, 80, 2245 (1998).

[4] R. Schirhagl, K. Chang, M. Loretz, and C. L. Degen, **Annu. Rev. Phys. Chem.**, 65, 83 (2014).

Modelling the quantum optical properties of quantum-dot plasmonic resonator systems

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The study of quantum light-matter interactions near metal cavities can be used to explore fundamental quantum optical regimes such as modified spontaneous emission of a single photon emitter. Metal resonators create localized surface plasmons which give rise to pronounced resonances in a similar way to high-Q cavity structures, but with incredibly small mode volumes. However, metal nanostructures are significantly more complicated to model because of material losses and it is well known that standard mode expansion techniques fail.

This talk will introduce several theoretical techniques for modelling quantum light-matter interactions in hybrid systems of quantum dots or "artificial atoms" coupled to plasmonic resonator systems. I will first describe how the electric-field Green function from classical optics can elegantly connect to quantum field theories in lossy plasmon systems. I will also describe how to derive useful time-local quantum master equations, and present a modal theory for understanding localized plasmon resonances. Various applications of these theories will be shown, including vacuum Rabi splitting, field driven quantum dots in the Mollow triplet regime, electron energy loss spectroscopy, single photon emitters, and hyperbolic metamaterials. Finally, I will discuss some open challenges and newly evolving fields such as plasmon-assisted quantum Raman scattering.

Quantum theory for plasmonic nano-laser with multi-level emitters: Optical and electric pumping

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Conventional laser design with a cavity and many quantum emitters does not allow constructing a laser of sub-wavelength size because of light diffraction limit. This constraint can be overcome by replacing the cavity with metallic films or nano-particles, which explore the light confinement by surface plasmon. However, to achieve lasing, we need enough emitters in a sub-wavelength region and pump them very hard to compensate the huge plasmon damping. In order to minimalize the required pumping power, we have to describe the pumping mechanisms of the emitters more accurately rather than using effective pumping rates. To achieve this goal, we model the system with Cavity-QED theory of multi-level emitters and solve the corresponding quantum master equation with exact and approximate method. The solutions of different levels reveal how the system output, like emission intensity and line-width as well as second-order correlation function of photons, are connected with the input, i.e. the power of the pumping laser or the current. In addition, we have also investigated saturation effect and mode competition.

Poster Contributions

Design Principles for "Next Level" Quantum Plasmonics in the Lab

***Günter Kewes*[1], *Andreas Ott*[1,2], *Max Schoengen*[2], *Oliver Neitzke*[1], *Pietro Lombardi*[3,6], *Rolf-Simon Schönfeld*[1], *Giacomo Mazzamuto*[3,6], *Andreas W. Schell*[4], *Jürgen Probst*[2], *Janik Wolters*[5], *Bernd Löchel*[2], *Yan Lu*[2], *Matthias Ballauff*[1,2], *Costanza Toninelli*[3,6], *and Oliver Benson*[1]**

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A promise of "quantum plasmonics" is to construct ultra-compact devices with new functionality. Here, by quantum plasmonics we think of hybrid systems [1], in which active quantum systems (emitters) interact with plasmonic nanostructures. In the last two decades fundamental quantum features of surface plasmons or localized plasmons have been studied [2]. Experimental results have so far not differed from classical electrodynamics. Essentially, a plasmon seems to be simply "a photon with damping".

In order to reveal potential differences between photons and plasmons and also to explore new functionalities, future experiments have to significantly reduce any "preventable" damping. Three major issues can be identified:

- coherence of the emitters
- direct quenching of emitters
- short lifetime of the plasmons

In our poster we present recent plasmonics experiments in our lab that cover the range from single plasmonic excitations to investigation of coherent processes in highly emitter-doped systems. Further, we discuss and propose experiments that are intended to bring quantum plasmonics to the "next level" - essentially to the strong coupling regime (with individual emitters) [3].

[1] Benson, **Nature**, 480, 193–9 (2011).

[2] Tame et al., **Nature Physics**, 9, 329–340 (2013).

[3] Kewes et al., **Sci. Rep.**, accepted (2016).

Creation and annihilation operators for quasinormal modes in open optical systems

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Exact treatment of emitter-plasmon and emitter-photon coupling in mesoscopic quantum devices with many quantum emitter

Michael Gegg

Institute of Theoretical Physics, Technische Universität Berlin

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Exciton Formation and Quenching in an Au/CdS Core/Shell Nano-Structure

Dirk Ziemann

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Near-field light-matter interaction in tip-enhanced Raman scattering

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