CPP 36 Single molecules II

Zeit: Mittwoch 11:15–12:30 Raum: TU C243

CPP 36.1 Mi 11:15 TU C243

Enhanced chromophore performance and single-molecule detection near a metal surface — •Maximilian Kreiter, Fernando Stefani, Krasimir Vasilev, and Volker Jacobsen — Max Planck Institut für Polymerforschung, Ackermannweg 10 D-55128 Mainz

A nearby metal film is known [1] to influence the excitation and emission rates of nearby chromophores, for both effects surface plasmons play a major role. We present a detailed quantitative study of these effects which became possible due to an improved sample preparation with ultraflat layers. Intensity as a function of excitation and emission direction is studied and found to be in quantitative agreement with the classical predictions down to a chromophore-metal separation of 10 nm where a significant deviation is found [2]. The photostability of the dyes turns out to be enhanced by the nearby metal [3]. In a scanning confocal optical microscope, detection of single fluorescent molecules is possible thorugh the metal. The observed patterns are modeled quantitatively and it is found that the main excitation and emission channels are via intermediate excitation of surface plasmons, leading to a preferential detection of dipoles perpendicular to the surface. This is experimentally proven by choosing different illumination modes. It turns out that the total number of photons that can be obtained from one chromophore is enhanced by a factor of 1.4 by the presence of the metal film.

[1] K. H. Drexhage et al. Ber. Bunsenges. Phys. Chem. **70**, 1179 (1966)
[2] K. Vasilev et al. J. Chem. Phys. **120**, 3439 (2004)
[3] K. Vasilev et al. J. Chem. Phys. **120**, 6701 (2004)

CPP 36.2 Mi 11:30 TU C243

A Common Source for Emission Intensity Fluctuations of Single Chromophores — •Frank Cichos[1], Abey Issac[2,1], Jörg Schuster[2], and Christian von Borczyskowski[2] — [1]Photonik und Optische Materialien, Institut für Physik, TU Chemnitz, 09107 Chemnitz — [2]Optische Spektroskopie und Molekülphysik, Institut für Physik, TU Chemnitz, 09107 Chemnitz

The emission intermittency of quantum dots and dye molecules is one of the striking phenomena of single quantum objects. Its occurrence is unwanted and wanted at the same time since it intermits the observation of the chromophore but resembles to be one of the simplest switching events possible in a nano-object. However, both issues require control over the intermittency, which has to be based on a detailed understanding of this phenomenon.

Our experimental results demonstrate, that both, emission intensity fluctuations of organic dye molecules and semiconductor quantum dots reveal power law statistics at timescales far beyond typical triplet state lifetimes. The similarity of the blinking statistics for both types of emitters suggests a common source. Experiments on single emitters in various dielectric environments reveal, that the dark periods during the intensity fluctuations are related to an ionization of the emitter including a self-trapping of charges in the environment. The duration of the bright periods is however largely uncoupled from the dielectric properties of the environment and involves intermediate states of the quantum dot or dye molecule.

CPP 36.3 Mi 11:45 TU C243

Near-field tip-enhanced Raman spectroscopy – towards single molecule sensitivity — •Catalin C. Neacsu and Markus B. Raschke — Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, D-12489 Berlin

The local-field enhancement at a sharp metallic tip in combination with resonance Raman spectroscopy provide an optical scanning probe method with ultrahigh spatial resolution and in principle sensitivity down to the single molecule level. Illuminating the apex of a Au wire tip in close proximity to the sample the resonance Raman response of malachite green and rhodamine 6G adsorbed on flat gold surfaces has been studied. The strong near-field coupling between Au tip and sample results in a high degree of field-confinement within several nm of tip-sample distance and is responsible for the Raman intensity enhancement. Spectral Raman line narrowing compared to the ensemble average and spectral diffusion is observed for submonolayer coverages: temporal fluctuations of spectral position and relative peak intensities as well as transient line splitting in time series of sequentially recorded spectra are indicative of probing only a small ensemble or cluster of molecules. These results indicate that with further improvements in tip preparation and illumination/detection efficiency single molecule Raman microscopy and spectroscopy is achievable in scattering-type near-field microscopy.

CPP 36.4 Mi 12:00 TU C243

Quantification of photo-induced and spontaneous quantum-dot photoluminescence blinking — •Fernando D. Stefani, Xinhua Zhong, Wolfgang Knoll, and Maximilian Kreiter — Max-Planck-Institut für Polymerforschung
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Photoluminescence blinking of $Zn_{0.42}Cd_{0.58}Se$ QDs on glass and conducting substrates was investigated experimentally as a function of the excitation intensity and modeled via a Monte-Carlo method. One (spontaneous) process is active for switching on and off, and has a power-law probability distribution for the length of both bright and dark periods. In addition, a single rate (photoinduced) process is evoked for the on→off transition. From the modeling of the data it is possible to extract independent information about both processes. The spontaneous blinking is completely independent of excitation intensity and the electronic properties of the environment. Spontaneous on- and off-switching processes show identical statistics which indicate a symmetry of the underlying physical mechanism, and a weak but noticeable memory effect. The photoinduced on→off transition is defined by its characteristic time τ_{PI} and can account for all the experimentally observed photoinduced effects. On glass, τ_{PI} is found to be inversely proportional to the excitation power. This indicates the presence of a 1-photon process in agreement with earlier studies on isolating matrices. On ITO, τ_{PI} shows an unprecedented exponential dependence on excitation intensity.

CPP 36.5 Mi 12:15 TU C243

Local field corrections for molecules in nanocavities — •Carsten Henkel — Universität Potsdam, Germany

Atoms, molecules, and other light emitters embedded in dielectrics are strongly coupled to their immediate environment. For a homogeneous, dense dielectric, the Lorenz-Lorentz or Clausius-Mossotti relation provides a link between the microscopic properties of its constituent molecules and its macroscopic properties (dielectric constant). We review recent results on these so-called “local field corrections” that differ for interstitial and substitutional emitters, respectively [1]. Our approach is motivated by experiments with emitters embedded in sub-wavelength sized particles [2]. For soft matter dielectrics, where the surrounding medium is allowed to have a continuously varying dielectric constant $\varepsilon(r)$, we show that the local field problem is actually ill-defined: the correction depends on the way the function $\varepsilon(r)$ approaches the homogeneous background. The “empty-cavity” model is recovered for a step-wise variation. We discuss implications for impurities embedded in Bose-Einstein condensates.

[1] F. J. P. Schuurmans, P. de Vries, and A. Lagendijk, Phys. Lett. A 264 (2000) 472.

[2] A. Beveratos et al., Phys. Rev. A 64 (2001) 061802(R); H. Schniepp and V. Sandoghdar, Phys. Rev. Lett. 89 (2002) 257403; S. F. Wuister, C. de Mello Donegá, and Andries Meijerink, J. Chem. Phys. 121 (2004) 4310; L. Rogobete and C. Henkel, Phys. Rev. A (2004, in press).