

Single-Molecule Single-Nanoparticle Microscopy

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Based on a scanning near-field optical microscope (SNOM) we present measurements on the optical interaction of a single molecule and a single gold nanoparticle. The phenomena of field enhancement, emission quenching and fluorescence enhancement are studied with outstanding control over all key parameters.

We have attached a single gold nanoparticle to the extremity of a pulled glass fiber tip and approached it to within a few nanometers to a sample of highly dispersed molecules. Dielectric nanostructures can give rise to a localization of the electromagnetic field to small volumes. The resulting excitation enhancement for a fluorescent molecule depends its dipole orientation which - in a crystalline matrix - is defined by the guest-host system and can be chosen normal or at an angle to the substrate surface. An outstanding photostability of terrylene-molecules embedded in an ultrathin para-terphenyl film [1] allows us to study the details of the changing emission properties when the distance to the gold nanoparticle reaches down to 5-10 nm. If the incident wavelength is chosen to excite plasmon resonances in the nanoparticle then the local field may be amplified by a factor of 30 at the particle surface, thereby boosting the excitation rate of the molecule close by. The emission process of fluorescence light, however, can branch into additional radiative and non-radiative channels which are opening up in the presence of the nanoparticle. As a result, an increase of the observed fluorescence by up to 20 is observed, accompanied by a drastic reduction of the excited state lifetime from 24 to 1 ns. As expected, the effect is localized to a diffraction-unlimited volume of $65 \times 65 \times 15 \text{ nm}^3$ at the gold particle [2].

A thorough analysis shows deviations from a free-space model consisting of a sphere and a dipole in air. Also, changes in the spectral and spatial emission characteristics and the photostability are evidenced.

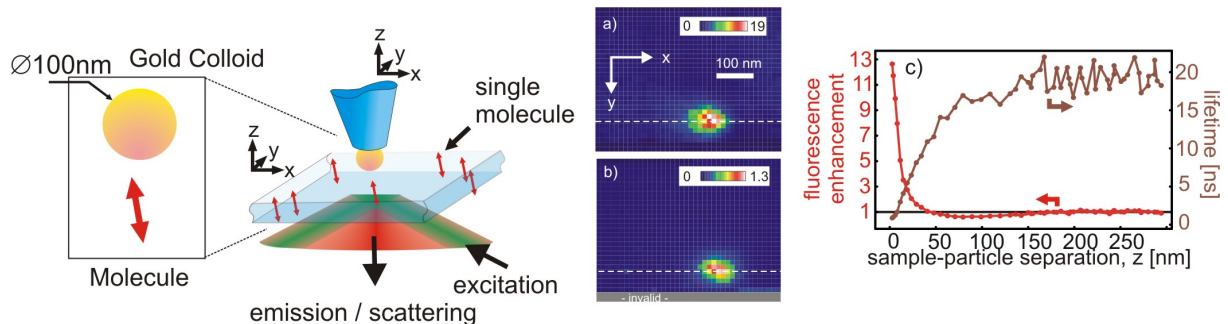


Figure 1: Sketch of our experimental setup, Surface scans: a) fluorescence intensity enhancement, b) excited state decay rate, c) Approach scan.

[1] R.J. Pfab et al., *Chem. Phys. Lett.*, **387** (2004) 490

[2] S. Kühn et al., *Phys. Rev. Lett. in print*, *arXiv*, *cond-mat/0604474*.