Direct Near-Field Absorption Spectroscopy on a Single Molecule

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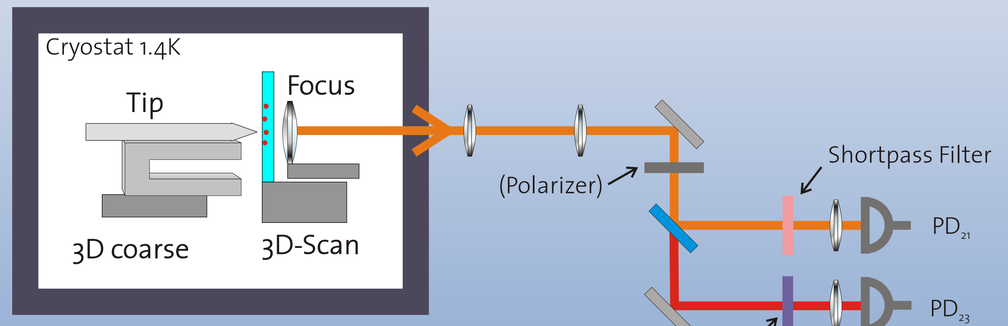
Abstract

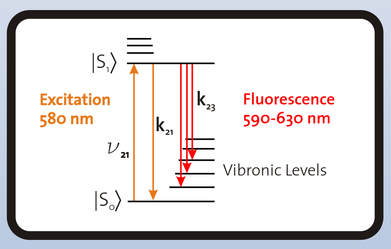
We discuss cryogenic experiments where a single molecule is excited by laser light under a metallic tip. Our setup consists of a combined confocal/near-field microscope that operates at T=1.4 K. Single organic dye molecules with absorption linewidths of a few tens of Megahertz are excited by a tunable narrow-band laser.

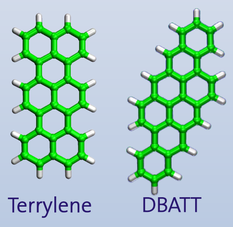
By studying the distance dependence and frequency dependence of the fluorescence and extinction caused by a single molecule, we investigate its coupling with the optical field. In addition, we discuss a scheme to control the line shape of the extinction signal by the use of a linear polarizer in the detection path.

Experimental Set-up

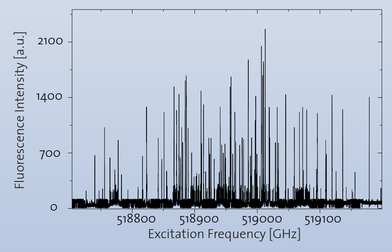


Molecules as single quantum systems

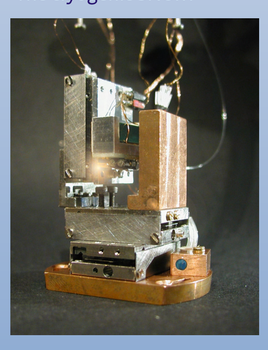




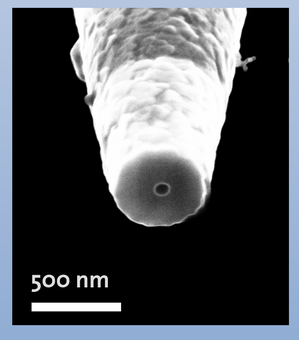
Inhomogeneous distribution of molecular resonance lines



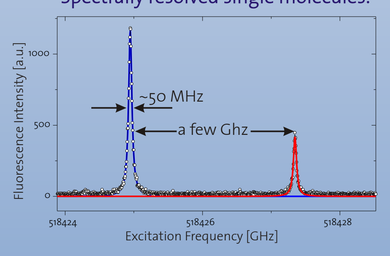
The cryogenic SNOM



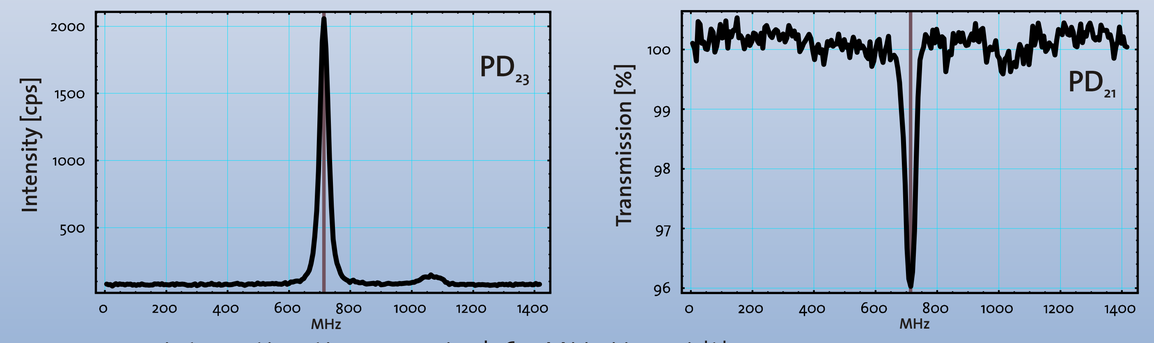
Nano fabricated tip



Spectrally resolved single molecules:

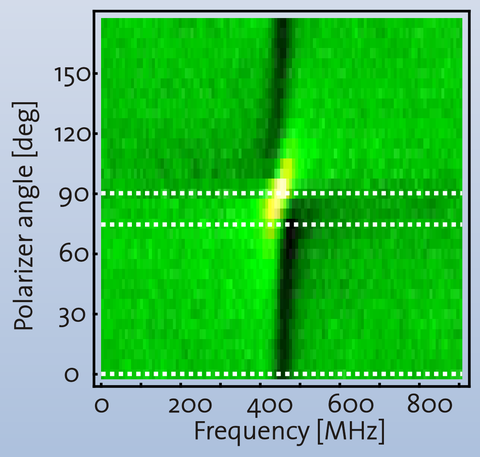


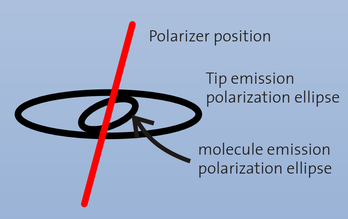
Direct Near-field absorption spectroscopy

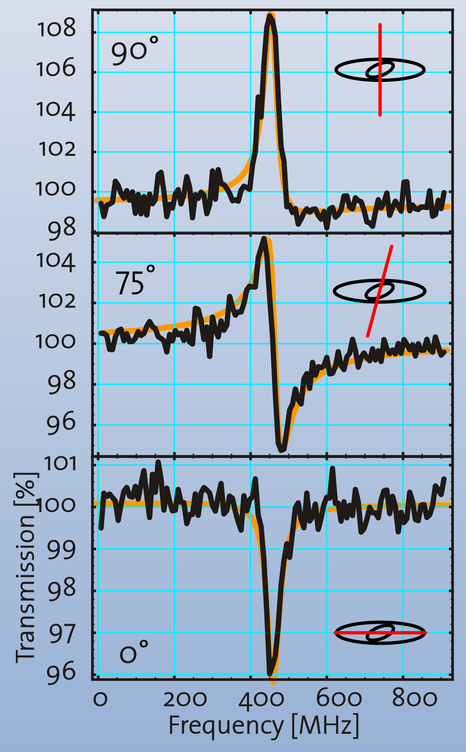


1 sec integration time per pixel, 60 MHz Linewidth

Polarizer control of the extinction line shape



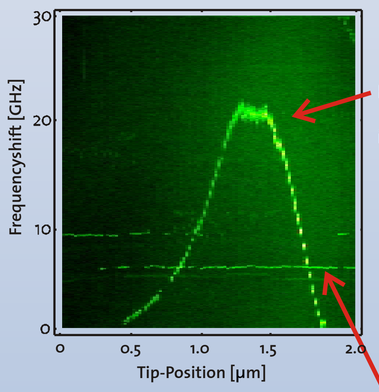




$$I_{\text{total}} = |E_{\text{tip}}|^2 + |E_{\text{mol}}|^2 + 2\Re(E_{\text{tip}} \cdot E^*_{\text{mol}})$$

Characterization of the system

a) Strong Stark shift due to proximity of the tip

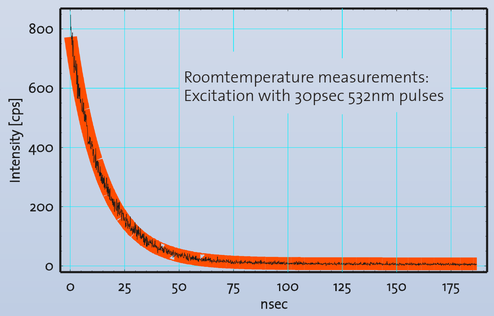


Resonance frequency of a molecule in the vicinity of the tip

Confocal illumination

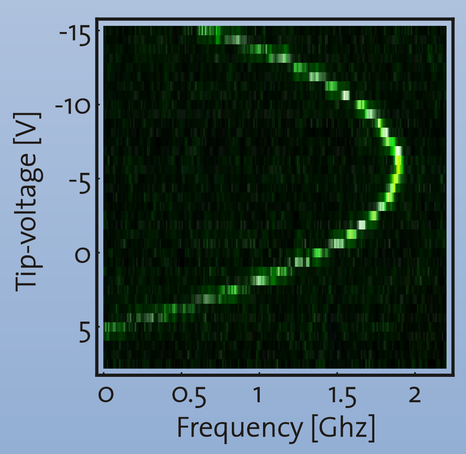
Molecules far away from the tip

b) Measuring the radiative lifetime

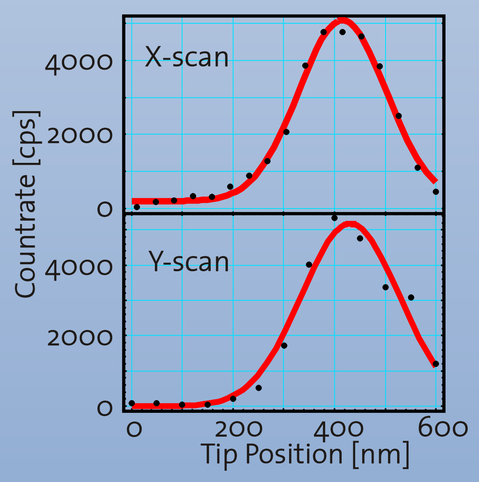


Lifetime of excited state ca. 15 nsec:
10 MHz homogeneous linewidth

lateral tip-molecule positioning and stray field compensation



e-field dependence of a single molecule resonance



Single molecule SNOM
(as presented on NFO 8)

Results & Outlook

At cryogenic temperatures, dye molecules in organic host crystals remain stable in space and frequency for an arbitrary time. This allows us to use them as model systems for quantum optical experiments.
We excite single molecules through the subwavelength aperture of near-field tips. The strength of interaction between light and molecule is controlled by the mode- and polarization overlap and the tip-sample distance.

Extinction can be interpreted as an interference between the excitation and coherently scattered light, and changes with the analyzed field components in the far-field. Here we present the first direct extinction measurements, without any noise suppressing elements. Further experiments to investigate and exploit the extinction effect are underway.

I. Gerhardt et. al., submitted (2006), ArXiv:quant-ph/0604177.