

Nanoscale Control of Single Photon Emitters by Optical Nano-Antennas and Tailored fs Pulses

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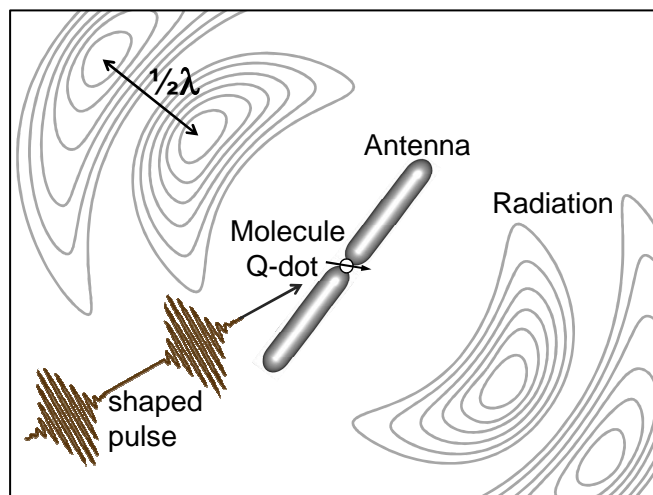
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Current trends in nanoscience now allow the exploration of light in and around nanostructures, single molecules, molecular complexes, etc. Indeed by proper control on the nm-scale sub-wavelength strong light fields are being created and detected. In the nanoworld single molecules or nanoparticles are the ultimate detectors of both local optical fields and interaction with the local environment. Here we focus on the control of single molecules using resonant nano-antenna and phase shaped fs pulses.



Figure, the concept:

A single molecule or quantum dot interacts with free optical radiation via a nano-optical antenna. In close proximity to the resonant nano-antenna the emitter exhibits enhanced excitation and decay rates and redirected emission [2]. Excitation by phase shaped fs pulses allows to control the excitation path.

We show how both excitation and emission of individual molecules is controlled by coupling to resonant optical nano-antennas. The molecule probes the local antenna field and here we show optical fields of a resonant monopole antenna, spatially localized within 25 nm [1]. Next the enhancement of the radiative and excitation rates is treated, particularly how the angular emission of the coupled system is highly directed, as the dominant antenna mode determines the angular emission. Thus arbitrary control over the main direction of emission is obtained, regardless of the orientation of the emitter [2]. A nano-Yagi-Uda antenna is discussed affording enhanced rates, strong unidirectional emission and, in reciprocity, efficient nano-focusing, making such antennas a promising candidate for compact easy-to-address planar sensors at the single molecule level [3].

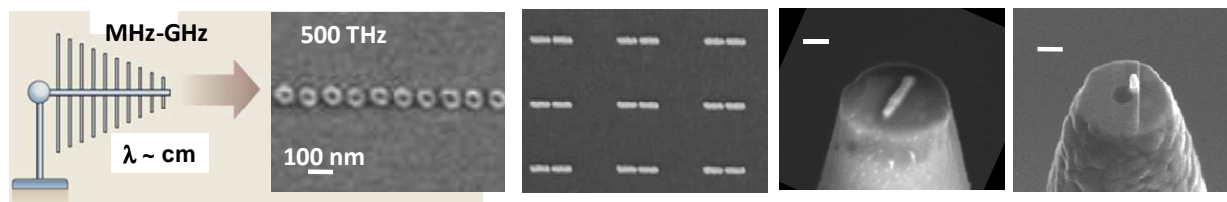


Figure:
 From radio to optical frequency, some characteristic nano-optical antennas.
 Scale bars 100 nm.

Next we address the femtosecond dynamics of single molecules at room temperature employing double pulse excitation scheme. By controlling both delay time and relative phase between the electric fields of the pulses we are able to manipulate the excited state population probabilities of single molecules. An analysis of these data based on the optical Bloch equations for two- and three level systems allows retrieving the dephasing time and Rabi frequencies of single molecules directly in the time domain.

The combined spatio-temporal control is promising for controlled single photon sources, light harvesting systems, efficient bio-sensors and optical imaging with 10 nm resolution.

References:

- [1]. Taminiau, T. H., Moerland, R. J., Segerink, F. B., Kuipers, L. & van Hulst, N. F. $\lambda/4$ resonance of an optical monopole antenna probed by single molecule fluorescence. *Nano Lett.* 7, 28–33 (2007).
- [2]. Taminiau, T.H., Stefani, F.D., Segerink, F.B. and van Hulst, N.F., Optical antennas direct single molecule emission, *Nature Photonics* 2, 234 (2008).
- [3]. Taminiau, T.H., Stefani, F.D. and van Hulst, N.F., Enhanced directional excitation and emission of single emitters by a nano-optical Yagi-Uda antenna, *Opt. Express* 16 10858 (2008).