Three Dimensional Optical Nanoscopy with Excited-State Saturation Microscopy

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The study of interactions between single emitters has raised a lot of interest in the past few years since it opens the way to the realization of quantum logic gates. Among the interactions, coherent dipole-dipole interaction between two emitters would have a very high coupling efficiency at nanometric distances. Since conventional fluorescence imaging techniques fail to resolve two emitters at such distances, we aim at developing a new optical nanoscopy method to image single quantum emitter at a nanometer resolution in three dimensions. The super-resolution scanning confocal imaging technique that we developed at cryogenic temperatures is based on the optical saturation of the first singlet excited state of a single fluorescent molecule. The excitation beam is shaped such that it contains an isolated intensity zero at the focus of the microscope objective, around which a large intensity gradient develops. As we increase the excitation beam intensity, the non-linear response of the molecule results in a broadening of the fluorescence intensity spatial distribution in the confocal image, together with a sharpening of the central dark spot. In a first step, we developed a 2D super-resolution method, using a Laguerre Gauss (donut-shaped) excitation beam. At 2K, we achieved a lateral optical resolution of 4.4 nm with an excitation intensity of 13 kW/cm^2 (Fig 1a.)[1]. With this method, the longitudinal resolution is similar to that of conventional confocal microscopy. In order to achieve 3D super-resolution, the excitation beam is shaped with a phase mask with a circular central area of π-phase retardation [2], which produces a ring-shaped profile in the focal plane and dark central field with symmetric bright spots along the optical axis in any axial plane. We achieved 3D super-resolution of single molecules with an axial resolution of 30 nm and a lateral resolution of 60 nm with an excitation intensity of 1 kW/cm^2 (Fig 1b.), i.e. with an intensity ~10^6 times smaller than that needed to saturate optical transitions at room temperature in STED-based methods.

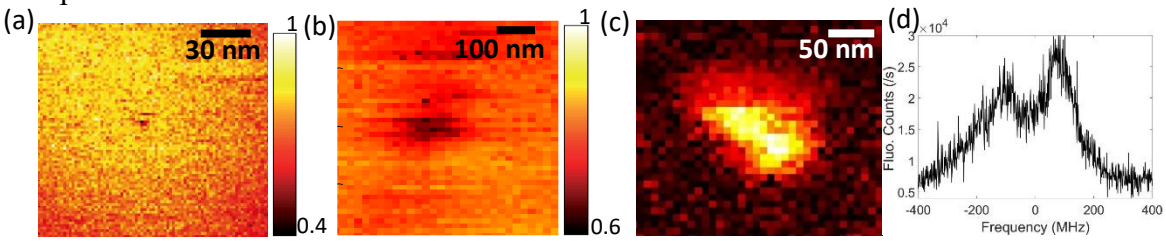

Fig.1 (a) ESSat image of a single DBATT molecule in Octadecane matrix in the (a) lateral plane (XY) using doughnut beam and (b) axial plane (XZ) using 0/pi beam (c) resolving two molecules with overlapping resonances and (d) fluorescence excitation spectrum. Color-bar giving the relative fluorescence intensity.

Our work will enable 3D-nanolocalization of single emitters close to nanostructures or dielectric surfaces, and open the way to the study of coherent dipole-dipole interactions between single emitters and their degree of entanglement [3].

References

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