Room Temperature Extinction Imaging of Single Emitters:
Fluorescence-Free Detection Down to the Single Molecule Level

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Since its first development and applications in the early 1990s, single molecule detection demonstrated to be a powerful tool for a wide range of studies, ranging from biophysics to quantum optics. However, this approach has been limited so far to species with high fluorescence quantum yield. Over the past few years, we have developed extinction detection and spectroscopy as an alternative to fluorescence for investigating single nano-objects such as metallic nanoparticles, viruses, dye molecules, and quantum dots [1-5]. The contrast mechanism at work in this coherent scheme can be understood as the interference between the incident beam and the light scattered by the nano-object. We showed that the sensitivity of this technique can be pushed to detect single semiconductor nanocrystals at room temperature and measured the absorption cross section of a single quantum dot before and after photo-bleaching as well as during the on and off photo-blinking times [4]. Here, I will concentrate on the recent improvements of such technique which allowed the direct detection and imaging of a single molecule by optical absorption at room temperature [6, 7]. Through a transmission microscopy setup in an index-matched configuration we were able to suppress the background scattering and shot-noise fluctuations below the part per million. Additionally, by means of three distinct approaches, the residual background fluctuations produced by the environment could be suppressed to about $1 \times 10^{-6}$ RMS. This allowed us to recover the extinction/absorption dip produced by a single molecule (Terrylene Diimide, TDI) on an impinging laser beam which can be evaluated to be about $3 \times 10^{-6}$. Thanks to these background-subtraction methods, which allowed to extract the pure absorption signal, we could also exactly determine exactly the absorption cross section of TDI [7], which turned to be in good agreement with an evaluation made upon the ensemble cross-section. In this framework, we also demonstrated that spectroscopy at the single molecule level is within reach [7]. This simple contrast technique will allow interrogating a large class of nano-object and studying the photophysics of all the species which do not fluoresce or do so with very low quantum efficiency.

References