Ultrafast stimulated emission microscopy of single quantum emitters

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The majority of spectroscopic techniques used to study nano-samples and single quantum emitters rely on spontaneous emission (PL). Spontaneous emission, however, is typically slow, occurring on a nanosecond time scale. Consequently it does not allow for ultrafast time-resolved probing of the excited state dynamics, which occur on femto to picosecond time scales. In my talk I will present a recently developed microscopy technique based on the time-resolved stimulated emission detection. First I will discuss some of the experimental challenges underlying femtosecond microscopy. I will then move on to the key experimental results and demonstrate that femtosecond detection of stimulated emission (SE) can be successfully used for imaging and probing the excited state dynamics in individual CdSe/CdS quantum dots. In our experiments simultaneous detection of the time-dependent PL and SE signals give direct access to the exciton relaxation time, which for the studied quantum dots amounts to 500 fs. Our spectrally resolved excitation-detection scheme allows us to directly disentangle the ground state depletion and stimulated emission processes.

Based on stimulated emission rather than spontaneous emission this microscopy approach holds a great potential for imaging and studying ultrafast processes in non-fluorescent or weakly fluorescent systems. The presented ultrafast detection scheme is a coherent variant of the commonly used transient absorption and as such it opens up a range of interesting experiments on charge dynamics, coherent effects and nanolasing.