Single Quantum Emitter Spectroscopy with Visible Sub-10 fs Pulses

Alexander Weigel, Aleksandar Sebesta, Philipp Kukura
Physical and Theoretical Chemistry Laboratory, South Parks Road, Oxford OX1 3QZ, UK
alexander.weigel@chem.ox.ac.uk

Abstract: We present a novel white light source providing sub-10 fs visible pulses with MHz repetition rates and apply it in femtosecond pulse pair experiments on single quantum emitters.
OCIS codes: (320.6629) Supercontinuum generation; (320.7150) Ultrafast spectroscopy**;** (140.7090) Ultrafast Lasers

1. Introduction

Femtosecond spectroscopy is key to understanding or even controlling ultrafast molecular motion and relaxation after optical excitation, but experiments have up to now been largely restricted to ensemble measurements. Ultimately, the current techniques have to be advanced beyond the ensemble average to study individual particles in their micro or nano-environment. Recent results [1,2] have been interpreted in terms of electronic coherence and wavepacket interference, and it was suggested that quantum control is possible on the single-molecule level. The experiments, however, were limited to the near-infrared region, because high-repetition rate femtosecond lasers for the visible are currently unavailable. On the other hand, visible absorption bands are found in many synthetic chromophores and biological systems like light harvesting complex II or green fluorescent protein. In order to target such systems we have developed a femtosecond white light source with 8 MHz repetition rate. We demonstrate visible femtosecond experiments on single quantum emitters.

2. Generating femtosecond visible pulses with 8 MHz repetition rate

The near-infrared output of MHz-repetition rate oscillators is conventionally converted to the visible region by producing white light in photonic crystal fibers.[3] However, propagation in the fiber leads to such strong phase distortions that the pulses can no longer be compressed to femtosecond duration. With high-power kHz amplifier systems, on the other hand, compressible white light is routinely generated by focusing into bulk material like sapphire or YAG. It has been shown that this approach is feasible also at higher repetition rates, if the pulse energy is raised to the 100 nJ level.[4] Following this route, we have reduced the repetition rate of a chirped-pulse Ti:Sa oscillator to 8 MHz by extending the cavity with a Herriott cell (Fig. 1), and thereby raised the pulse energy to 180 nJ. The output pulses, centered around 780 nm, are compressed with SF11 prisms to 100 fs. The peak intensity is sufficient to generate stable white light continuum directly by focusing into a 3 mm YAG crystal. By avoiding extensive nonlinear propagation in a photonic crystal fiber we reach exceptional stability with short-time fluctuations on the order of only 0.1 %. The spectrum, also shown in Fig. 1, covers the full visible and near infrared region, starting from 450 nm. With chirped mirrors and a spatial light modulator (SLM) we can routinely compress the visible part (magenta colored area) to below 10 fs duration. The repetition rate of 8 MHz provides an optimum duty cycle for the excitation of single molecules, and the pulse shaper allows performing a variety of femtosecond experiments without additional optical elements.

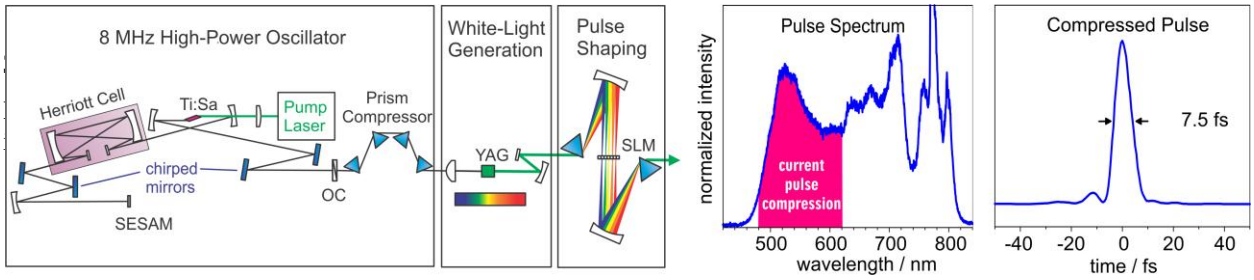

Fig. 1. Setup of the femtosecond white light source, output spectrum, and reconstructed temporal profile of the compressed output pulses. The current spectral range for compression is marked in magenta. Abbreviations: SESAM: Semiconductor saturable absorber mirror. OC: Output coupler. SLM: Spatial Light Modulator.

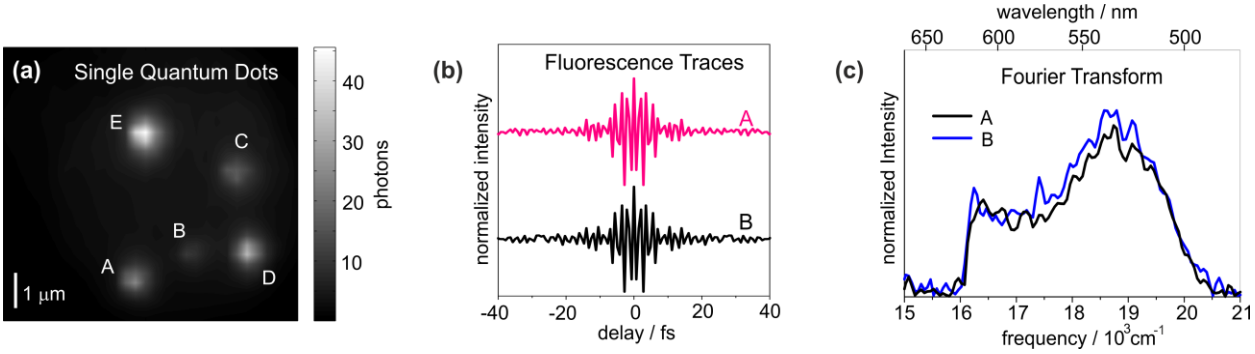

Fig. 2. Femtosecond experiments on single quantum dots. (a) Wide-field fluorescence image of single quantum dots (QDot 655 from Invitrogen) on a glass surface. (b) Delay-dependent fluorescence intensities of two individual dots illuminated with a pair of identical transform-limited white-light pulses. For Fourier transformation the traces were mirror-imaged around time zero. (c) Fourier transform spectra of the traces in (b).

3. Femtosecond pulse pair experiments on single quantum dots

The white light was used to perform femtosecond experiments on single nanoscopic particles. For a first experiment, single CdSe nanocrystals (QDot 655 from Invitrogen) deposited on a cover glass surface were chosen as the sample. The compressed white light pulses were focused with reflective optics onto the sample, and fluorescence was collected with a 1.42 NA microscope objective (Olympus PLAPON). Transmitted excitation light was rejected by a longpass filter, and wide field fluorescence images of the quantum dot sample like the one in Fig. 2 (a) were collected with an EMCCD camera. With the SLM, the excitation pulse was split into two identical transform-limited copies, and the inter-pulse delay was scanned. Following the emission intensity of individual quantum dots with varying delay leads to the traces in Fig. 2 (b). We ascribe the oscillatory modulation to delay-dependent interference of the excitation pulses. Electronic coherence phenomena and wavepacket interference, as proposed in the discussion of similar experiments on single molecules, were not required for the explanation of our results.[1,2] Fourier transformation of the interference traces (c) reveals the fluorescence excitation spectra of each quantum dot. This provides access to the absorption spectra of single quantum dots, which have not been directly measured, to date.

4. Conclusions and Outlook

We have developed a femtosecond white light source with 8 MHz repetition rate, for which the visible part of the output pulses is compressed to below 10 fs. In a first experiment the novel continuum source was used for pulse pair experiments on single quantum dots. The recorded fluorescence traces can be explained by optical interference only. We will present further femtosecond experiments on single quantum dots and fluorophores.

5. Acknowledgements

We thank Alexander Fuerbach from the Macquarie University in Sydney for providing the SESAM. A. Weigel was supported by the Royal Society with a Newton International Fellowship, and by the New College, Oxford with a Junior Research Fellowship.

4. References

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