

Work Plan

Pump-probe experiments are a powerful tool for accessing molecular dynamics and photochemical responses of biological compounds, where ultrafast processes can be directly tracked in the time domain. The state of the art of these experiments has reached the timescale of about 4 femtoseconds, which has enabled electron motion to be directly observed [1]. However, in order to gain a deeper understanding of such processes and particularly how electronic and nuclear motion is coupled, it will be necessary to use more selective laser pulses. This necessitates the development of ultrashort UV pulses which can directly access specific valence orbitals in the molecule. However, the generation of sub-10 fs UV pulses poses serious challenges in terms of: (i) broad bandwidth generation, (ii) pulse energy, (iii) spectral phase handling, (iv) pulse measurement. To address these issues, at IFN-CNR they will be developing schemes for generating ultrashort UV pulses based on 3rd harmonic generation and collinear sum-frequency generation (SFG).

Through this STSM I intend to be fully involved in this development from the outset and hopefully be leading some of the work by the end of my visit. There will be two stages of the development work:

Ultrashort Third Harmonic Pulse Generation: 2 Nov – 4 Dec 2015 (5 weeks)

As first step, UV laser pulses will be produced by third harmonic generation through non-linear interaction with a gas medium. Using an annular ring beam it is possible for the phase matching to produce an output with the fundamental component on the outside and the 3rd harmonic component in the middle of the beam [3]. This allows the two beams to be separated using a mirror with a hole which reflects the fundamental but allows through the 3rd harmonics. This scheme has the advantage that neither beam encounters additional optical material which may stretch the pulses due to their large bandwidth.

Ultrashort Tuneable UV Pulse Generation and Characterisation: 17 Jan – 14 March 2016 (8 weeks)

The second part of the project is dedicated to an enhancement of the beamline to produce ultrashort tuneable UV pulses. This source could provide more information on the electronic energy levels and the relative ultrafast processes in biological molecules. The tuneable UV pulses will be produced by a non-linear up-conversion process by combining fundamental pulses with broadband visible pulses [4].

An essential part of any ultrashort pulse generation process is the pulse characterisation. Figure 1 shows a Mach-Zehnder interferometer which will be constructed for autocorrelation measurements of the pulse length. In this case the non-linear medium needs to be a dilute gas (to avoid pulse stretching) with the measured signal being the yield of ions detected in a time of flight mass spectrometer. This setup will then be used for a formative experiment of ultrafast dynamics in DNA nucleobases.

It is the ultimate goal of this project to have a two colour UV(fixed)-UV(tuneable) pump probe experiment with which to study a range of biomolecular systems with high temporal resolution. By the end of this STSM, I hope to have gained hands-on experience of ultrashort pulse non-linear optics and will be in a position to participate in and lead experiments in Belfast, Milan, and elsewhere in the future.

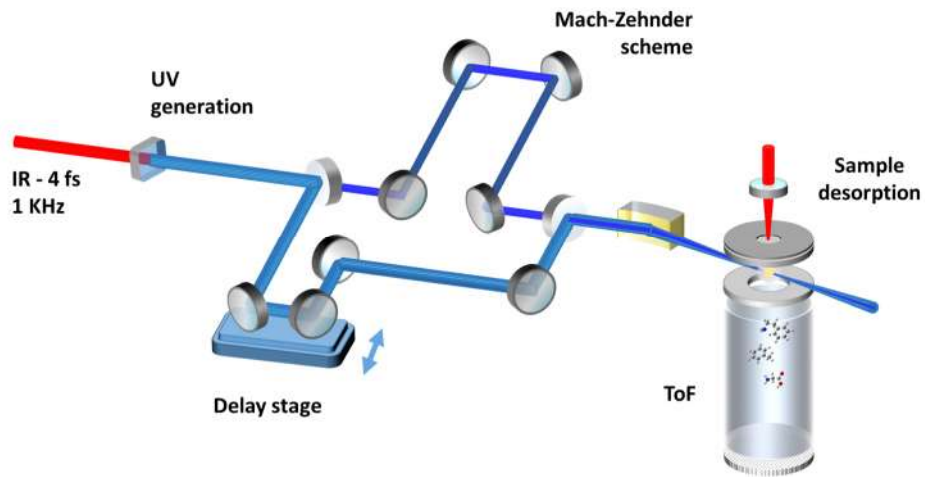


Figure 1: Schematic representation of the UV beamline

[1] F. Calegari et al., *Science*, **346**, 336 (2014)

[2] F. Calegari et al., *IEEE Journal of Selected Topics in Quantum Electronics*, **21**, 1-12 (2015)

[3] B. Glushko et al., *Phys. Rev. Lett.*, **71**, 243 (1993)

[4] A. Candeo, P. Farinello, C. Manzoni, and G. Cerullo The European Conference on Lasers and Electro-Optics 2013. Munich Germany (2013)