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BOXCARS-geometry 2DES setup in the 300-340nm range with pulse-to-pulse phase correction at 50kHz

Thomas Roland, *Vincent* Kemlin, *Julien* Nillon, *Jean-Sébastien* Pellé, *Olivier* Crégut, *Johanna* Brazard, *Jérémie* Léonard, *Stefan* Haacke*

Université de Strasbourg - CNRS UMR 7504, IPCMS, Strasbourg 67034, France

Abstract. A 40-nm broad pulse centred at 320nm is produced from an amplified Yb-doped fiber laser operated at 50kHz, and used in a BOXCARS geometry setup for 2DES, with shot-to-shot monitoring of the relative optical phase stability.

Two-dimensional electronic spectroscopy (2DES) is a method of choice for the investigation of the ultrafast photo-reactivity of complex molecular system to disentangle electronic from vibrational couplings, competing ultrafast photochemical processes, and structural or chemical heterogeneities. We aim at developing such an experiment, targeting the challenging UV range, [1-3] in particular the 300-345 nm window of high interest for biomolecule-chromophore complexes, such as DNA sequences labelled with long wavelength-absorbing nucleic base analogues [4,5].

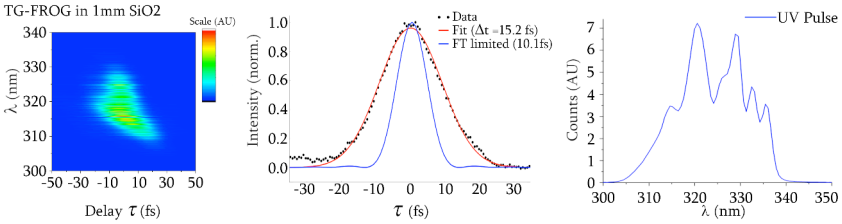

Figure 1: UV-pulse characterisation. Left: TG-FROG map. Middle: spectrally-average TG-FROG trace with its fit and the Fourier-transform limited pulse width. Right: UV pulse spectrum.

We generate a sub-15fs UV pulse starting from a Yb-doped fiber amplifier delivering high-energy (100 uJ) NIR pulses at high and tuneable repetition rates up to 2MHz. A 6-fs red-NIR pulse is produced with a Non-collinear Optical Parametric Amplifier (NOPA) pumped with the second harmonic of the fiber amplifier as previously described [6]. We adapt the Achromatic Sum-Frequency Generation (ASFG) scheme proposed by Nabekawa et al. [7] to convert a large fraction of this Red-NIR spectrum into the 300-345 nm spectral

* Corresponding author: Stefan.Haacke@ipcms.unistra.fr

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range at a repetition rate of up to 200 kHz. ASFG is achieved by engineering the required angular dispersion of the red-NIR pulse in order to fulfil the phase matching condition for SFG in BBO of all its spectral components with the 515-nm, nearly monochromatic (300-fs long, Fourier Limited - FL) second harmonic of the fundamental pulse. More precisely this angular dispersion is computed to be $3.38*10^{-4}$ rad/nm, to ensure relative internal angles with the crystal axis ranging from 67.5° to 73.9° for wavelengths spanning from 700 to 1030 nm, respectively. The red-NIR pulse is thus dispersed at the apex of a 60° SF10 prism, magnified by a 1/8 telescope and overlapped with the 515-nm pulse coming at an internal angle of 44.6° with respect to the crystal axis. The resulting UV pulse is shown in fig. 1, supporting 10-fs FL pulse duration. Accurate optimization of the angular dispersion of the incoming Red-NIR pulse is critical to optimize both the spectral width and the spatial profile of the UV pulse.

Two pulse pairs are produced in two interferometers and overlapped again in the BOXCARS geometry (see Fig. 2), where the UV pulse duration is characterized by a transient grating - FROG measurement. The reconstructed pulse duration is observed to be 15-fs (see Fig. 1).

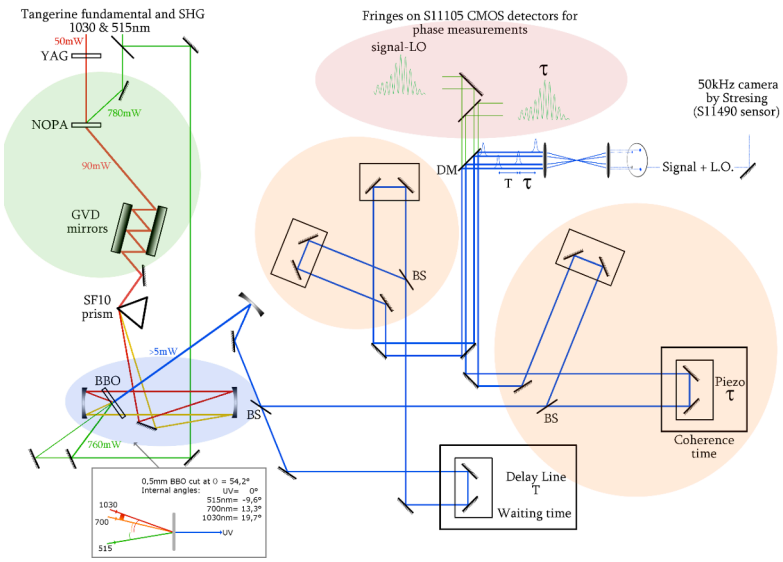

Figure 2: Scheme of the optical set-up, highlighting the ASFG stage (bottom left) and the interferometric tracking of phase fluctuations (right).

To overcome the optical phase stability issues expected in such a 2DES setup, we chose to monitor the pulse-to-pulse relative phase fluctuations in order to correct for those by post-processing. To this end, a fraction of the 515-nm beam is used as a tracer beam overlapped with the UV beam at the ASFG stage (see Fig. 2) and separated again, after travelling through both interferometers, with a dichroic mirror. The two resulting green pulse pairs are used to generate 2 interferometric patterns, reporting on the fluctuations of the coherence time and of the probe-LO time. Producing the same interferometric patterns with the UV pulse and comparing the phase fluctuations recorded simultaneously with the

UV and green beams allows us to demonstrate that the optical path fluctuations in the UV pulse pairs are measured to better that 3nm (<lambda/100), for each laser shot at 50kHz.

In conclusion, we report on the generation of 15-fs UV pulses in the 300-340nm range, which is used to implement a 2DES set-up in a BOXCARS geometry. The optical phase fluctuations within pulse pairs are monitored to better than λ/100 for each individual laser shot at 50kHz repetition rate, which should allow us to perform accurate Fourier-transformed spectroscopy via phase fluctuation post-processing, a prerequisite for applying this set-up to 2DES spectroscopy.

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