NIREOS

GEMINI 2D

INTERFEROMETER

for time-resolved measurements

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The GEMINI 2D is the advanced model of the GEMINI interferometer.
It is a compact and ultra-stable interferometer, capable of generating a pair of **collinear and phase-locked replicas of ultra-short pulses**, with unrivaled stability and robustness.

The GEMINI 2D is specially designed to:

- keep constant the **dispersion** introduced during the scan of the delay between the two replicas
- keep fixed the **absolute arrival time** of one of the two replicas (with attosecond stability)

- High throughput
- 1 attosecond stability
- Scan range selectable by the user
- Compact and insensitive to vibrations

Main Applications

The GEMINI 2D is the ideal device for **time-resolved measurements**, where it is crucial to preserve the pulse duration and the synchronization with other light pulses.

Perfectly suitable for **two-dimensional (2D) spectroscopy experiments**, to generate a pair of collinear and phase-locked pump pulses

Two-dimensional electronic spectroscopy

Two-dimensional electronic spectroscopy: an ultrafast laser spectroscopy technique that can probe the electronic, energetic, and spatial landscapes of a sample.

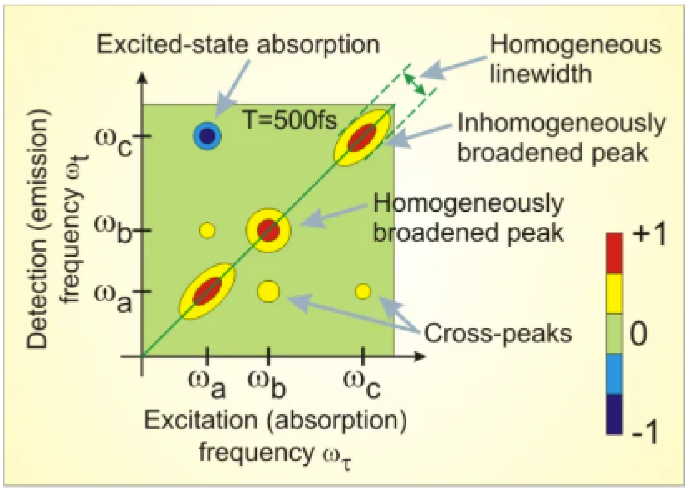


Figure 1: Model 2D spectrum for a three-component system. The elongation of the peaks along the diagonal displays the instantaneous distribution of frequencies in the transition, whereas the minor axes of the elliptically shaped diagonal peaks are measures of the homogeneous linewidths.

Two-dimensional electronic spectroscopy (2DES) is the optical analogue of Nuclear Magnetic Resonance, a spectroscopic technique that has revolutionized structural biology, enabling the determination of complex molecular structures with high spatial resolution.

2DES is the **"ultimate" time-resolved nonlinear optical experiment**, since it provides the maximum amount of information about the third-order nonlinear response of a system, and any other third-order nonlinear spectroscopy (such as pump-probe) is contained in the 2D spectrum.

2DES **displays its full power in systems with multiple interacting components.** 2DES provides two dimensional spectra that shows correlations between excitation and detection frequencies, with simultaneously high spectral and temporal resolution.

2DES can dissect congested spectra and reveal the molecular connections between transitions, thus providing a means to elucidate the overall functionality of a sample.

Two-dimensional electronic spectroscopy requires the interaction of the sample with a sequence of 3 laser pulses. Two of them must be phase-locked replicas, with variable delay, and can be easily generated with NIREOS' **GEMINI-2D interferometer** by placing it in the pump beam before the sample, thus turning a pump-probe setup into a state-of-the-art 2DES spectrometer.

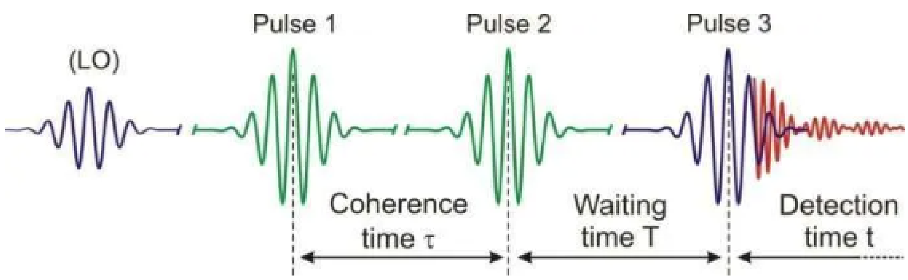


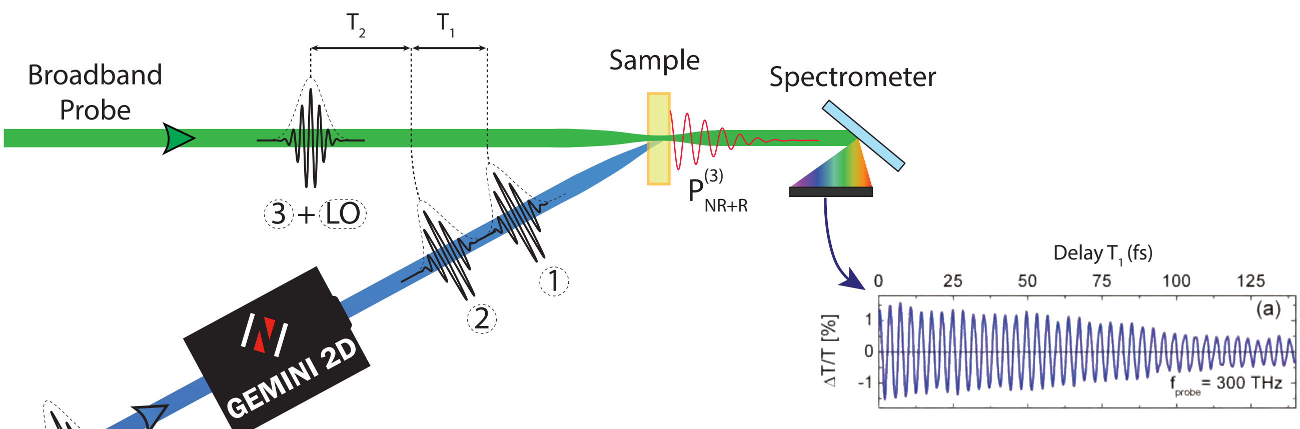
Figure 2: Pulse sequence in a 2DES experiment. In a 2-colour experiment pulses 1-2 (interferometrically stable) can have a different color with respect to pulses 3/LO (also interferometrically stable).

NIREOS' GEMINI-2D is employed to generate pulses 1 and 2 and control their coherence time τ.

Advantages of Two-dimensional electronic spectroscopy (2DES)

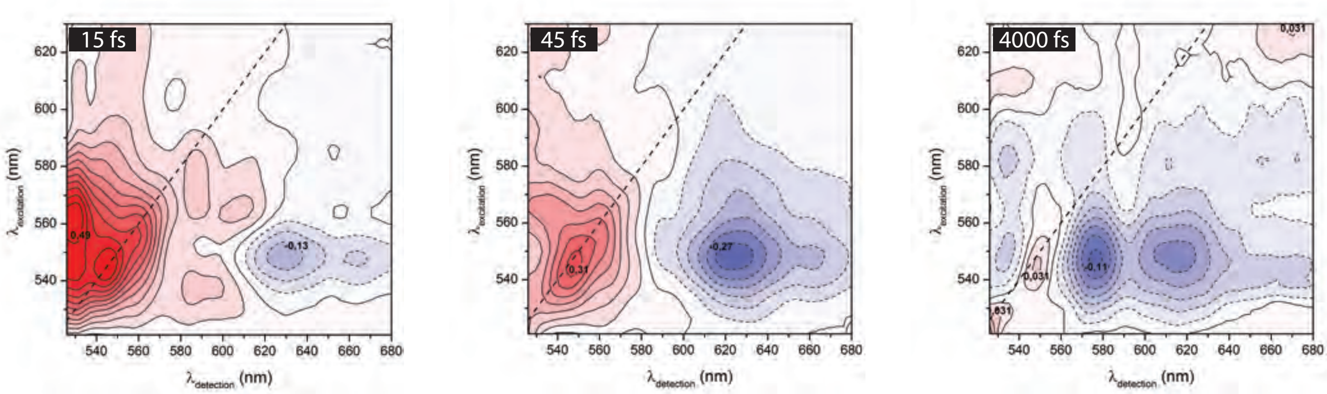
- It is possible to separate, and thus distinguish, contributions to the nonlinear signal that are spectrally overlapped in one-dimensional experiments. Analysis of cross-peaks reveals whether the different transitions seen in the sample absorption spectrum arise from the same or different molecular species and can quantify couplings and correlations between different excited states.
- 2DES removes inhomogeneous broadening and thus measures the homogeneous linewidth of optical transitions, enabling the individual levels to be singled out in strongly congested spectra.
- Two-dimensional electronic spectroscopy can follow the parallel pathways by which the coupled electronic dynamics evolve after photoexcitation in real time. This makes the 2D technique a particularly powerful tool for tracking excitation energy transfer processes from origin to terminus through multiple channels simultaneously.
- 2DES overcomes the Fourier limit, obtaining simultaneously high temporal resolution (to follow dynamics occurring on the femtosecond timescale) and spectral resolution (to resolve excitation and emission energies over significant bandwidths with high spectral accuracy).
- It is possible to retrieve structural information on the relative spatial arrangement of the chromophores, by either probing the intermolecular electronic couplings through the analysis of the cross-peaks or manipulating the pulse polarizations. 2DES can connect the molecular structure (determined by X-ray crystallography) with the electronic energy levels and, in favorable cases, it can provide insight into the molecular structure when unknown.

Two-dimensional Spectroscopy (in pump-probe geometry)



Oscillating transient signal as a function of the relative delay T_1 between the two pump pulses at a selected probe wavelength and at a fixed population time T_2.
A **Fourier Transform** as a function of T_1 for each probe wavelength allows one to retrieve the 2D maps as a function of detection and excitation wavelengths.

Bidimensional maps of 2D Electronic Spectroscopy measurements obtained on a Light Harvesting (LH1) complex of a sample of Rhodospirillum Rubrum for three different population times T_2 (15 fs, 45 fs, 4000 fs).



- The GEMINI 2D interferometer is placed in the pump beam before the sample, allowing one to generate two collinear and phase-locked pump pulses (1 and 2) with a relative variable delay T_1 (coherence time).
- The absolute arrival time of pulse 2 is kept fixed in order not to change the delay T_2 (population time) during the scan of T_1.
- In the pump-probe geometry, the rephasing and non-rephasing signals - P_{NR+R} - are emitted in the same direction as the probe (3), which also acts as a local oscillator (LO) that heterodynes them on the detector.

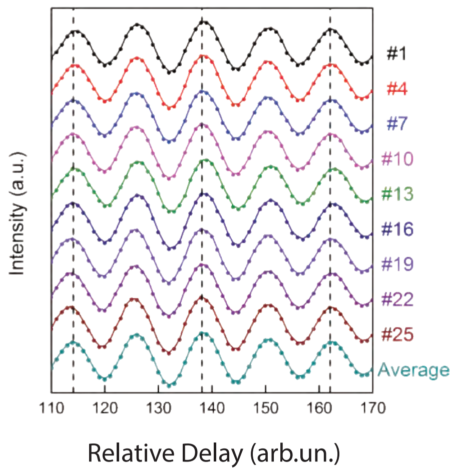
D. Brida et al., Opt. Lett. 37, 3027-3029 (2012).

J. Réhault et al., Rev. Sci. Instrum. 85, 123107 (2014).

R. Borrego-Varillas et al., Opt. Express 24, 28491-28499 (2016).

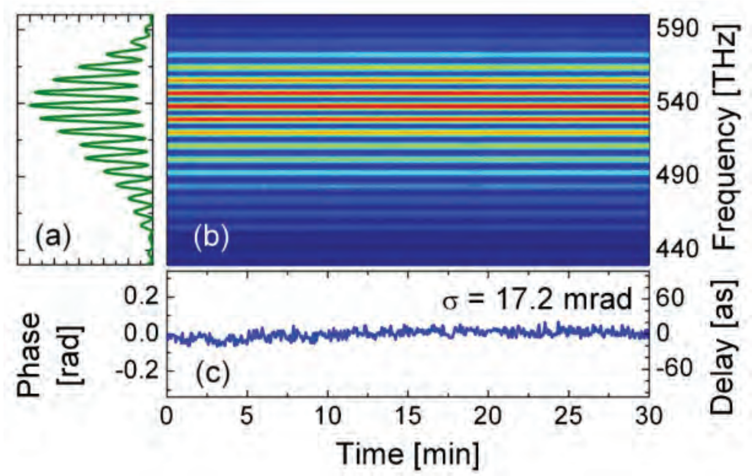
Performances

REPEATABILITY TEST



Interference patterns between the two replicas generated by the GEMINI 2D Interferometer, measured for different scan number indicated on the right. The lower one is the average of 40 scans. The measurements show an exceptional repeatibility!

STABILITY TEST



(a) Fringe pattern generated by the interference of the two replicas created by the GEMINI 2D Interferomter; (b) sequence of fringe patterns acquired by keeping the relative delay fixed for 30 min; and (c) corresponding phase fluctuations. The phase jitter is less than 1 / 360th of the optical cycle.

Technical Specifications

VERSION		S	L
Spectral range [nm]		400 - 2300 (Standard) 250 - 3500 (Ultra-broadband)	
Max. Delay τ [fs @ λ=600 nm]	SYM	± 400	± 1050
	ASY	-100 → 700	-100 → 2000
Delay τ Stability		< 1 attosecond	
Dimensions [mm]		180 x 180 x 90	
Weight [kg]		2	

Spectral Resolution