NIREOS

Broadband Pump–Probe Spectroscopy at MHz Modulation Frequency

Advantages of this Configuration

- Broad Spectral Range (from UV to IR)
- Single-Pixel Detector
- Single-Channel Lock-in Amplifier
- MHz Modulation Frequency
- Extremely high sensitivity

The GEMINI interferometer enables an innovative approach to broadband pump-probe spectroscopy. Thanks to the employed time-domain Fourier Transform (FT) detection system, this configuration allows one to measure the broadband pump-probe signal with a single-pixel detector and a single-channel lock-in amplifier. In this way, it is possible to combine an ultra-broad spectral coverage with an extremely high sensitivity, due to high frequency modulation (in the MHz regime) and detection.

This approach is applied, as an example, to a high-repetition rate laser system, for which no single-shot multi-channel detectors are available. An Erbium-fiber oscillator (FemtoFiber Pro from Toptica Photonics) is employed, providing pulses at 1.55-μm central wavelength and 40-MHz repetition rate. The output beam is divided into two branches: the first branch is sent to a highly non-linear fiber, which broadens the spectrum of the incoming beam, producing a broadband (λ_{probe}=950-1450 nm) probe beam (in red in Figure 1). The pump beam (in blue in Figure 1) is generated by second harmonic generation (λ_{pump}=780 nm) with a duration of ~300 fs; the pump beam is then sent to an acousto-optic modulator working at 20-MHz frequency (synchronized with the laser clock). As shown in Figure 1, the pump and probe beams are focused on the sample in a non-collinear geometry. After the sample, the broadband probe is sent to the GEMINI interferomter, that creates two collinear replicas delayed with a variable delay t. The two replicas are sent to a single-pixel detector, where they interfere giving rise to an interferogram as a function of their relative delay t.

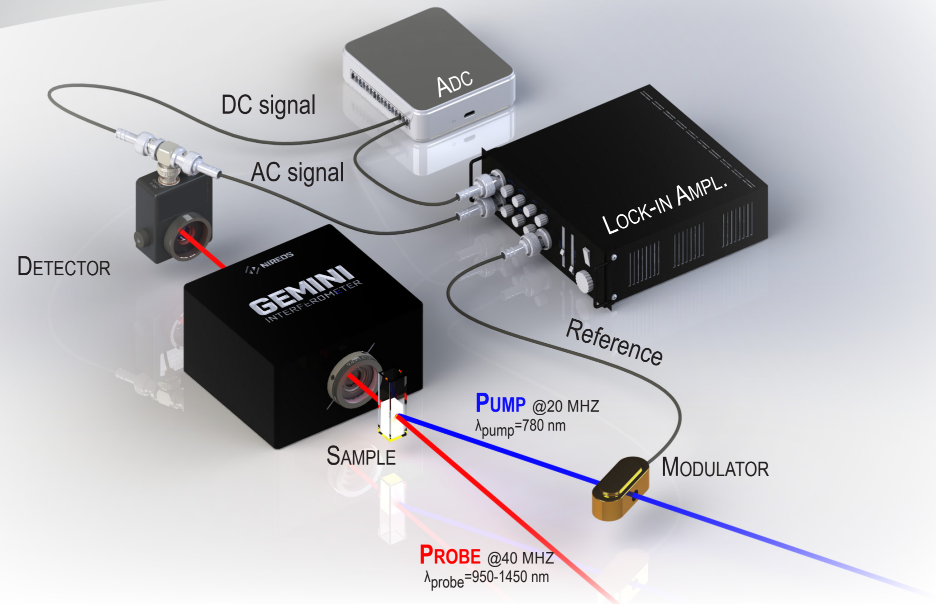

***FIGURE 1**: Experimental Setup for Pump-Probe Spectroscopy using the GEMINI Interferometer.*

The AC output of the single-pixel detector is sent to a high-frequency lock-in amplifier. An analog-to-digital conversion card (ADC) simultaneously records the demodulated signal from the lock-in and the DC output of the detector. These two waveforms, shown in Figure 2, are called the differential interferogram ΔT and the linear probe interferogram T, respectively. By computng their FTs one obtains the differential transmission spectrum ΔT and the linear transmission spectrum T of the sample as a function of wavelength λ (shown in Figure 3(B)). The main advantage of this procedure is that, thanks to the linearity of the FT operator, to retrieve the ΔT spectrum there is no need to record two interferograms of the perturbed and unperturbed sample and compute the difference of their FTs (as in a typical pump-probe measurement), but it is sufficient to record the interferogram of the ΔT signal, measured by the lock-in, and compute its FT. The pump-probe signal is then retrieved by computing the ratio ΔT/T.

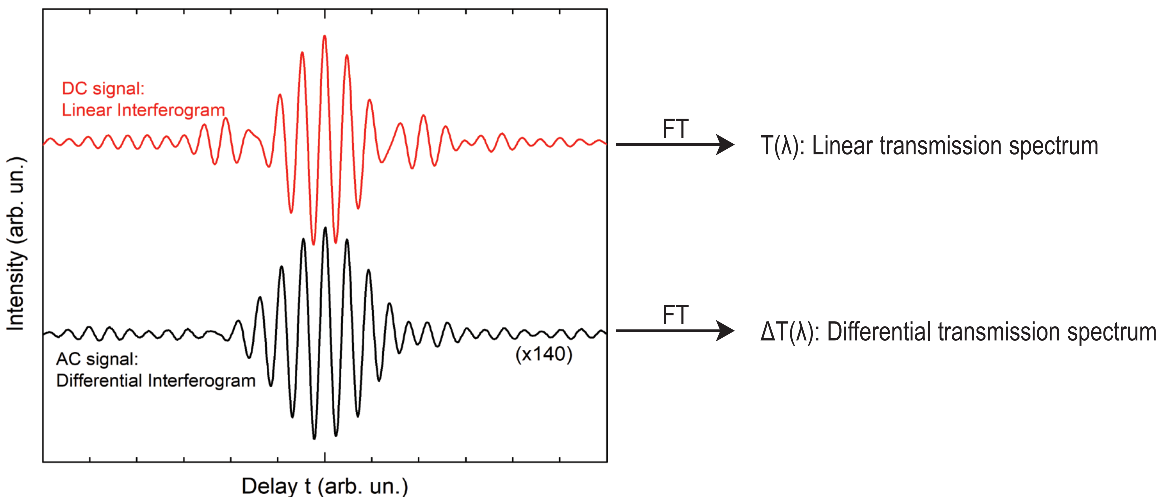

***FIGURE 2**: Examples of the linear and differential interferograms, which are measured simultaneously.*

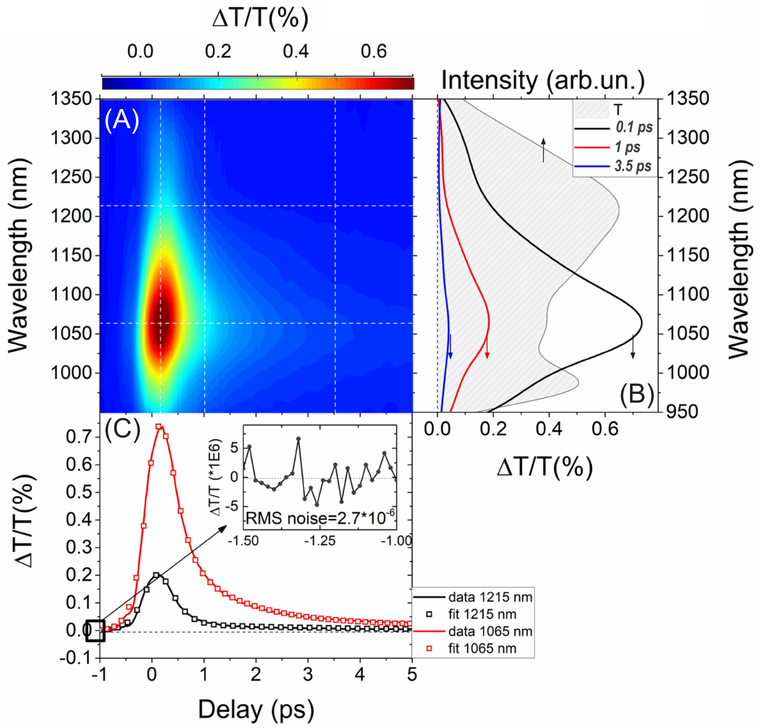

***FIGURE 3**: ΔT/T map (A) for a SWNT sample, together with linear transmission spectrum and pump-probe spectra at different pump-probe delays (B) and dynamics at selected wavelengths (C).*

By scanning the pump-probe delay, ΔT/T dynamics on a spin-coated sample of semiconducting single-walled carbon nanotubes (SWNTs) are reported in Figure 3. The integration time for measuring the signal ΔT/T at each pump-probe delay was 1.5 seconds. The two-dimensional ΔT/T map is reported in Fig.3(A) as a function of probe wavelength and pump-probe delay. ΔT/T spectra at selected delays (100 fs, 1 ps and 3.5 ps) are plotted in Fig. 3(B). After photoexcitation, a strong positive signal arises, peaking at λ=1065 nm, due to ground-state photo-bleaching of the first excitonic transition of the SWNTs, which decays on the ps-timescale. To highlight the population dynamics in the SWNT sample, Fig. 3(C) reports ΔT/T time traces and fits to the data at the peak of the signal (λ=1065 nm, red) and on its red-shifted shoulder (λ=1215 nm, black).

The real advantage of our detection scheme consists in running at high modulation frequencies, where the laser relative intensity noise is typically the lowest. This is clear by taking a closer look at the dynamics at λ=1215 nm and negative delays (see inset of Fig. 3(C)). The RMS noise is 2.7×10^{-6}.

In conclusion, this innovative detection scheme for pump-probe spectroscopy combines a broad spectral coverage with very high modulation frequencies. This enables one to perform broadband measurements with an excellent signal-to-noise ratio in a short time. Moreover, being based on a single detector and lock-in amplifier, this system is less expensive and complex than other implementations using a detector array connected to a multichannel lock-in.