Whitepaper

An introduction into high-speed ASOPS

***Abstract**: High-speed asynchronous optical sampling (ASOPS) using 1GHz source demonstrates many advantages over conventional time-domain spectroscopy.*

Conventional ultrafast time-domain spectroscopy (TDS) is based on pump-probe schemes in which a single femtosecond (fs)-laser provides the pump and the probe pulses. A pump pulse excites a sample under investigation and creates a non-equilibrium state. The evolution of that state is interrogated using a time-delayed pulse that probes the sample response as function of time-delay versus the pump pulse. The time-delay is usually adjusted by varying the distances that the pulses travel and thus their relative time-of-flight before arriving at the sample. Many data points are acquired at different time-delays to complete a full measurement. In most cases one pulse travels over a retro-reflecting mirror whose position can be modified using a motorised translation stage or a vibrating membrane.

In contrast to a conventional ultrafast TDS system the time-delay in a high-speed ASOPS system is realised without any mechanical delay scanning devices. To this end two femtosecond lasers with repetition rates f_R=1 GHz are employed that are stabilised at an offset of Δf_R=2 kHz (offsets up to 20 kHz are possible). The faster laser serves as the pump laser, the slower one as the probe laser. As result of the detuning, successive pairs of pump and probe pulses arrive at the sample with a delay that incrementally increases by 2fs with each pulse pair. Thus, the delay between pump and probe pulses is linearly ramped from 0 to 1ns. The ramp is reset to zero whenever the faster pump laser 'overtakes' the probe laser after exactly 500µs (the inverse of Δf_R) and a new measurement cycle starts. See Fig. 1 for an illustration of the high-speed ASOPS time-delay principle.

In a high-speed ASOPS system, the pump and probe pulses are applied to the sample in exactly the same way as in standard setups, except that they originate from two separate femtosecond lasers. The probe laser is detected using a fast photoreceiver and digitised with a fast A/D converter as a function of real-time. The real-time scale is then simply converted to a time-delay scale by applying a scaling factor $\Delta f_R / f_R$.

High-speed ASOPS permits a boost to ultrafast measurement techniques that is probably comparable to the step from an analog data plotter to a digital sampling oscilloscope. With a high-speed ASOPS system a 1ns time-delay window can be scanned at a rate of 2kHz with a time-delay resolution of 45fs. The 1ns time-delay - given by the inverse repetition rate (1GHz) and

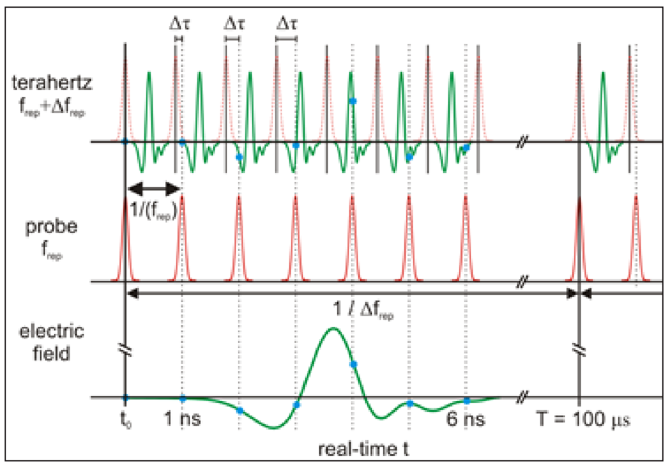

Figure 1: ASOPS time-delay principle. For a better understanding the principle is shown with single cycle THz pulses generated by the pump pulses (red dotted curves in the upper part labelled "terahertz").

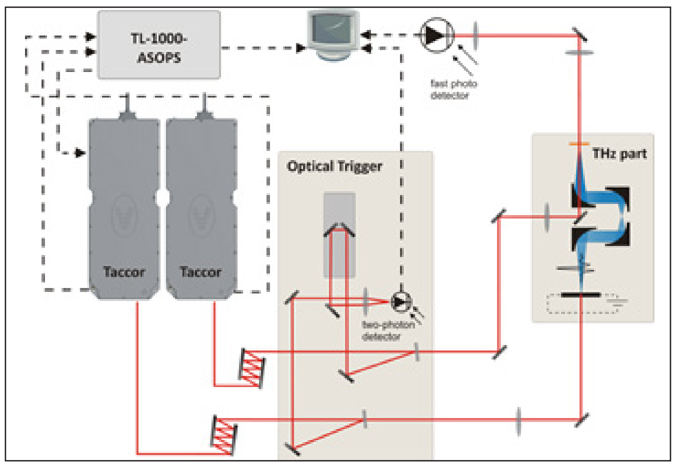

Figure 2: Overview of the HASSP-THz spectrometer containing 2 taccor lasers, one TL-1000-ASOPS stabilisation unit, the data acquisition computer and fast photo detector, the Optical Trigger unit and the THz part.

hence typically not tunable - corresponds to a frequency resolution of 1GHz in the spectral domain. The scan rate of 2kHz makes online signal monitoring and optimisation possible and permits the suppression of technical and environmental noise very efficiently. Typical noise sources are laser fluctuations, vibrational perturbations or motion of air and have significant contributions up to 1kHz limiting the scan rate to more than 1kHz. Compared to a conventional ultrafast TDS system the continuous data accumulation in a high-speed ASOPS system increases the signal-to-noise ratio within a given measurement time significantly. In a well-designed high-speed ASOPS experiment the signal-to-noise ratio is entirely determined by shot-noise meaning it has a square-root dependence on data acquisition time.

Apart from a much accelerated data acquisition, inherent advantages of high-speed ASOPS are the absence of changes in optical spot size at the sample location and the absence of pointing instabilities originating from the scan unit. In addition there is no need for searching the temporal zero point because the time-delay is scanned across a full pulse-to-pulse spacing of the pump laser each time. With independently tunable pump-and probe-lasers the realisation of two-color experiments is straightforward, providing additional versatility.

A high-speed ASOPS system based on fs oscillators with 1GHz repetition rate has several advantages compared to a low-speed ASOPS system with 100MHz fs oscillators. It permits a higher scan rate, better time-delay resolution and a higher signal-to-noise ratio at a given measurement time. Notably, the scan rate Δf_R is linked to the time-delay resolution $\Delta\tau$ by the formula $\Delta f_R = \Delta\tau \times B \times f_R$ where B is the bandwidth of the measurement system (detector + A/D converter). If f_R is decreased by a factor of 10 either $\Delta\tau$ is increased by a factor of 10 or the scan rate needs to be reduced by a factor of 10. While in the first case measurements in the sub-100fs regime become impossible the second route is not applicable as the scan rate then falls well within the range of technical noise sources and thus the ASOPS advantage is lost.

In addition, the lasers a high-speed ASOPS system require an active stabilisation of the repetition rate offset in order to maintain precise time axis calibration over long acquisition times. The real-time axis t is converted to a time-delay axis τ by applying the scaling factor $\Delta f_R / f_R$. While the stability of the absolute repetition rate f_R has no practical effect, the stability of the repetition rate difference Δf_R crucially determines the precision of the time-delay axis calibration. Even small drifts in Δf_R can cause severe effects on the precision of the calibration and thus destroy the time resolution, especially if multiple transients are to be averaged for noise reduction.

The TL-1000-ASOPS stabilisation unit from Laser Quantum performs the stabilisation of the repetition rate offset frequency and is a one box - one button solution. In combination with two taccor lasers, the turn-key femtosecond 1GHz laser from Laser Quantum, the ensemble forms the core of Laser Quantums high-speed ASOPS system. Figure 2 gives an overview over all components of Laser Quantums HASSP-THz spectrometer (also shown in figure 3) offering the unique combination of more than 6THz spectral coverage and 1GHz spectral resolution [1,2]. If THz is not of interest the THz part can be easily replaced by any other ultrafast spectroscopy setup [3].

Figure 3: Laser Quantum's HASSP-THz - high resolution THz spectrometer

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References:
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[2] Senna et al., Determination of Cellulose Crystallinity by Terahertz-Time Domain Spectroscopy, Analytical Chemistry Vol. 86, p. 3780 (2014).
[3] Gebs et al., High-Speed Asynchronous Optical Sampling with sub-50fs Time Resolution, Optics Express Vol. 18 p, 5974 (2010).