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 \( f_{\text{p}} = 1 \text{GHz} \) are employed that are stabilised at an offset of \( \Delta f_{\text{p}} = 2 \text{kHz} \) (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 1 ns. The ramp is reset to zero whenever the faster pump laser `overtakes' the probe laser after exactly 500µs (the inverse of \( \Delta f_{\text{p}} \)) 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 axis is converted to a time-delay axis \( \mathcal{U} \) by applying the scaling factor \( \Delta f_{\text{p}} / f_{\text{p}} \). Notably, the stability of the absolute repetition rate \( f_{\text{p}} \) has no practical effect, the stability of the repetition rate difference \( \Delta f_{\text{p}} \) crucially determines the precision of the time-delay axis calibration. Even small drifts in \( \Delta f_{\text{p}} \) 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].

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 \( f_{\text{p}} \) is linked to the time-delay resolution \( \Delta \mathcal{U} \) by the formula \( \Delta f_{\text{p}} = \Delta \mathcal{U} \times B / f_{\text{p}} \), where \( B \) is the bandwidth of the measurement system (detector + A/D converter). If \( f_{\text{p}} \) is decreased by a factor of 10 either \( \Delta \mathcal{U} \) 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 \( \mathcal{U} \) by applying the scaling factor \( \Delta f_{\text{p}} / f_{\text{p}} \). While the stability of the absolute repetition rate \( f_{\text{p}} \) has no practical effect, the stability of the repetition rate difference \( \Delta f_{\text{p}} \) crucially determines the precision of the time-delay axis calibration. Even small drifts in \( \Delta f_{\text{p}} \) 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.

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References:

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