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Energy stability in multi-timescale pump-probe spectroscopy with free-running lasers (ADASOPS)

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Abstract. ADASOPS pump-probe spectroscopy is a multi-timescale technique that is spreading rapidly especially in the field of biomolecule dynamics. Based on slight variations of the laser repetition rate, it is simple to implement and can cover a time range extending from a hundred femtoseconds to a millisecond or more. We have studied the energy fluctuations associated with this approach and have proposed a method for overcoming any instabilities.

The study of the dynamics of complex systems, particularly biological molecules, is of great interest, but can be extremely difficult to implement experimentally. The main challenge is due to the fact that the response to stimulation is in general a complex combination of different factors and therefore can evolve over very large timescales. For this reason, it is necessary to access the various temporal ranges, either by different techniques specific to each interval, or by a single multi-scale technique. This latter has the advantage of strictly identical experimental conditions for each timescale, but the experimental setup can be rather complex.

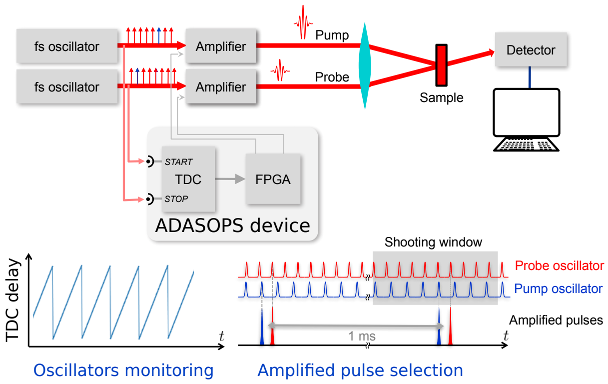

Figure 1. Multi-timescale pump-probe experiment: the ADASOPS device receives the two oscillator pulse trains as input and generates two trigger signals for managing the amplifiers.

ADASOPS (Arbitrary Detuning Asynchronous Optical Sampling) is a spectroscopy technique that enables multi-timescale experiments to be carried out with relative experimental simplicity, since the complexity of the technique is confined to the design of the electronic device that drives the experiment [1–3]. ADASOPS applications are expanding rapidly, particularly in the field of biomolecule dynamics [4, 5], thanks to the fact that the method is easily implemented on pre-existing pump-probe experiments with two lasers.

Fig. 1 shows a typical ADASOPS pump-probe experiment with 1 kHz amplified laser systems. Two trains of pulses generated by independent femtosecond oscillators have in general a difference in repetition rate, resulting in an asynchronous scan of the time delay. The ADASOPS device uses a TDC (time-to-digital converter) combined with an FPGA (field-programmable gate array) to measure the delay variation at 10 MHz and, by averaging, track this temporal phase in real time with sub-picosecond resolution. The device uses this information for repeatedly select the best pulse to be amplified, according to the user-requested delay: after each amplified pulse it identifies a shooting window centered 1 ms later and including N oscillator pulses (N being limited by the laser's tolerance to frequency variation, typically 1-2 %). The amplified pulse is the one in the window with the delay closest to the target delay.

Since ADASOPS generates an instantaneous variation of the amplification period $\mathscr{T}$ that could affect the amplifier efficiency, we have analysed its effect on the stability of the laser energy E. The experimental setup is as shown in Fig. 1 but without the sample, so that the absorption measurements (ΔA) give a direct estimation of the laser noise. The pump beam is chopped at 500 Hz in order to record a reference spectrum (pump-OFF) in between each signal acquisition (pump-ON). For each amplified pulse, the ADASOPS device communicates the delay that had actually been obtained and the counter of the oscillator pulse that had been amplified, which can be used to calculate $\mathscr{T}$. The energy information is obtained by integrating the spectra recorded by the detector. The graphs of Fig. 2 clearly show the correlation between $\mathscr{T}$ and ΔE on the top,

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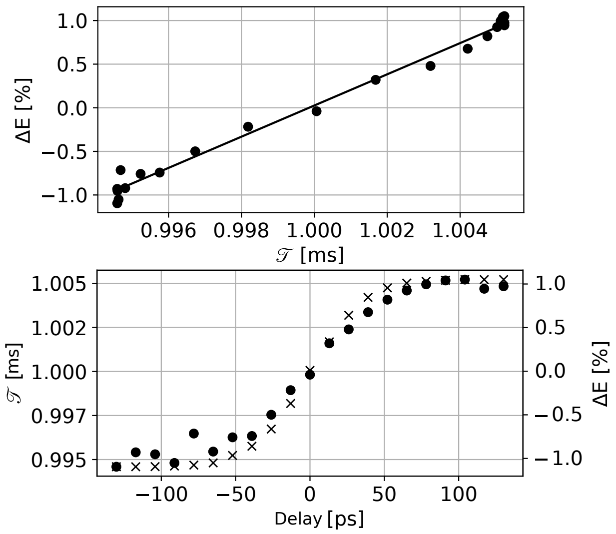

Figure 2. Top: correlation between laser energy variation and the instantaneous period of the amplifier. Bottom: correlations of laser energy variation (dots) and instantaneous period (crosses) with delay.

and the correlations between these two quantities and the obtained delay on the bottom. The dependency between $\mathscr{T}$ and the delay is due to the fact that the best delays approximating the target are at the beginning of the shooting window (i.e. small $\mathscr{T}$) if they are smaller, or at the end of the window (i.e. large $\mathscr{T}$) if they are greater than the target. The reason for this depends on the ratio between the oscillator repetition rates and is beyond the purpose of this abstract.

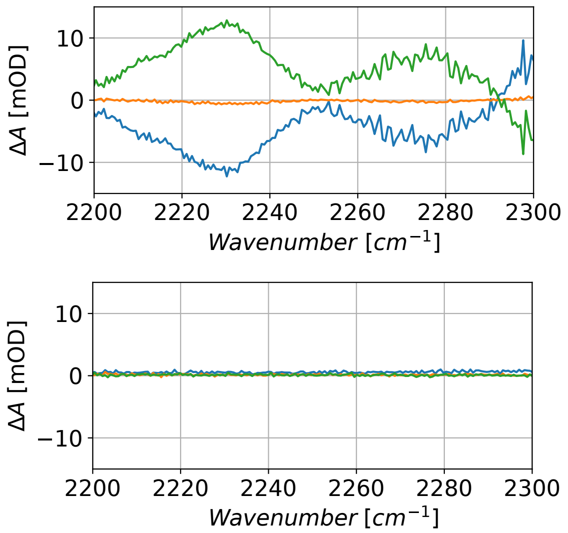

Figure 3. Top: ΔA measurements without compensation algorithm for delays below (blue), above (green) or centered (orange) with respect to the target delay. Bottom: same measurements with compensation algorithm.

In the upper plot of Fig. 3 are reported the ΔA measurements recorded by targeting a given delay. The curves blue, green and orange correspond to the ΔA calculated for achieved delays respectively below, above and centered with respect to the target delay. The difference between the blue and green curves suggests that there are symmetric modifications of the signal spectra for delays exceeding or staying below the target, which is clearly a direct consequence of the laser energy fluctuation. The flatness of the orange curve is due to the compensation of these effects for delays centered on the target delay. Therefore, ADASOPS can indeed increase laser instability, particularly if the amplifiers are poorly adjusted. However, these instabilities are easily counterbalanced by implementing a compensation algorithm in the ADASOPS device: since the probe pulse used to record a reference is coupled with a chopped pump pulse its delay has no meaning, hence it can be adjust in such a way that it always has the same instantaneous period $\mathscr{T}$ as the associated pulse used to measure the signal in the calculation of ΔA. The great efficiency of this approach is illustrated in the lower graph of Fig. 3.

In conclusion, although the ADASOPS technique can generate laser instabilities, these can be readily compensated by a convenient reprogramming of the algorithm used for amplified pulse selection, as was achieved e.g. in [5].

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