OSCAT: Novel Technique for Time-Resolved Experiments Without Moveable Optical Delay Lines

Rafał Wilk · Thomas Hochrein · Martin Koch · Michael Mei · Ronald Holzwarth

Abstract Here, we present a novel technique for time resolved experiments without any moveable external delay line. Optical Sampling by Cavity Tuning (OSCAT) method is based on a femtosecond laser with a tunable repetition rate and a passive delay line. The time delay between the pump and probe pulses is determined by the repetition rate of the femtosecond laser.

1 Introduction

The invention of modelocked femtosecond lasers has enabled a large variety of time resolved experiments in femtochemistry and femtobiology [1, 2], semiconductor physics [3, 4], terahertz time-domain spectroscopy [5, 6] or optical coherence tomography [7]. In most of the above mentioned pump and probe experiments an optical pulse is split into two parts which are subsequently superimposed on the sample. Mostly, mechanical delay lines are employed to temporally delay one pulse with regard to the other [8–11]. These include linearly moving stages, rotating mirrors or fiber-stretchers [12]. They comprise free-space optics and moveable components and are demanding in terms of mechanical stability and optical adjustment. Typically, the scanning speed of these mechanical delay lines is limited to several ten Hz with a range of a few picoseconds. Alternatively, an asynchronous optical sampling (ASOPS) technique overcomes these drawbacks as it does not require any mechanical delay lines. The technique relies on two synchronized pulsed femtosecond lasers which are slightly detuned in the repetition rate [13]. However, such systems need a

R. Wilk (✉) · M. Mei · R. Holzwarth
Menlo Systems GmbH, Am Klopferspitz 19, 82152 Martinsried, Germany
e-mail: r.wilk@menlosystems.com

T. Hochrein
Sueddeutsches Kunststoff-Zentrum, Friedrich-Bergius-Ring 22, 97076 Wuerzburg, Germany

M. Koch
Experimentelle Halbleiterphysik, Philipps-Universitaet, Renthof 5, 35032 Marburg, Germany

more complex stabilization of the pulse repetition rates and usually increase the price of the system.

In the following, we present a novel method [14] which allows a direct control of the time delay between two pulses by varying the pulse repetition rate. The pioneer work of Yamaoka *et al.* [15] and Ye [16] was presented in the field of unbalanced Michelson interferometry. Contrary to the ASOPS scheme only one femtosecond laser source is required. The new technique allows for very robust, compact and cost-efficient experimental setups in the above mentioned fields. We call this technique Optical Sampling by Cavity Tuning (OSCAT).

2 Theory

In conventional time-resolved experiments the pump and probe pulses result from the same optical pulse which is split into two portions by an optical beam splitter. The temporal shift between the pump and probe pulses is set using mechanical delay lines.

However, in principle it is also possible to employ pump and probe pulses that originate from sequenced optical pulses if the timing jitter of the femtosecond source is negligibly small. Thus, a change in the lasers repetition rate f_{rep} modifies the time shift between the pump and probe pulses. Let us assume that a beam from a femtosecond laser is split up into two beams, as shown in Fig. 1. The first beam A is guided directly to the target and travels along a path with the length l_p. The second beam B is guided through an additional passive delay line with the length l_d. In this configuration, a reference pulse i from the beam A will reach the target at the same time as one of the persecutive pulse i-a from the beam B. The temporal shift $\Delta\tau$ between the pulse i and i-a, as shown in Fig. 1, is determined by the repetition rate of femtosecond laser. A sweep of repetition rate from f_{rep} to $f_{rep} + \Delta f$ results in a continuous change of the temporal offset, as given by:

$$\Delta\tau = a \left(\frac{1}{f_{rep}} - \frac{1}{f_{rep} + \Delta f} \right). \tag{1}$$

Therefore, a simple frequency scan with the OSCAT technique allows time resolved experiments in general. Hence, the executable scanning range of the pulse delay scales with the factor a. The necessary optical path length l_d for a given a is:

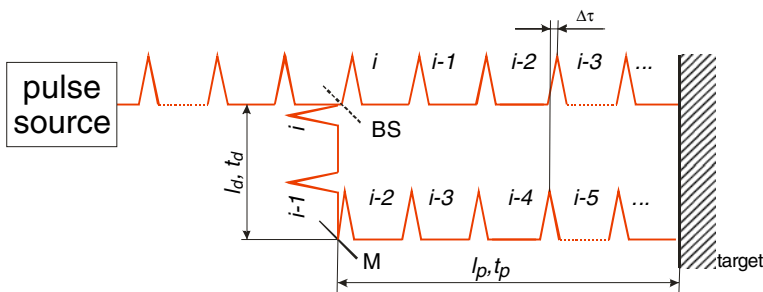$$l_d = \frac{a \cdot c_0}{f_{rep} \cdot n}, \tag{2}$$

Fig. 1 Principle of the OSCAT technique.

where n is the refractive index of the delay line medium and c_0 is the speed of light in vacuum. Thus, the necessary optical length of passive delay line for a given temporal scanning range $\Delta\tau$, repetition rate f_{rep} and tuning range Δf equals to:

$$l_d = \frac{\Delta\tau \cdot c_0 \cdot (f_{rep} + \Delta f)}{\Delta f \cdot n}. \tag{3}$$

While the necessary length of delay line depends on $f_{rep}/\Delta f$, lasers with high repetition rate are preferable for OSCAT technique, in which a small changes in the oscillator length results in a large sweep range Δf. On the other hand, for a large value of the index a the timing jitter could be an issue. Thus, lasers sources used in this technique have to have an excellent stability.

Another limitation of the OSCAT method could arise from the phase jitter and the pulse-to-pulse carrier-envelope phase slippage. This phenomenon can be measured as a carrier envelope offset frequency f_{CEO} [17]. Time-resolved experiments, where the pulse-to-pulse carrier phase relation is important, such as pulse interferometry, would require the carrier phase stabilization in addition to the stabilization of repetition rate. The f_{CEO} has to be set to zero to archive the same pulse-to-pulse carrier phase. Alternatively, f_{CEO} can be stabilized to a fraction of the repetition rate f_{rep}/k. In this case every k-th pulse will have the same carrier phase.

3 Experimental setup

We have chosen cross-correlation measurements for the experimental verification of the presented technique. The experimental setup is shown schematically in Fig. 2 and consists of four parts: tunable femtosecond source, passive delay line, conventional delay stage and BBO crystal with a photodiode detector.

3.1 Femtosecond laser source

The core component is a sub-90 fs fiber laser with a repetition rate of 250 MHz (Model M-Comb, Menlo Systems GmbH) [18]. Its repetition rate can be tuned over 2.5 MHz with a precise stepper motor or over 3.5 kHz with a much faster internal piezo-stack, respectively. The laser output is connected to a fiber amplifier with an output power up to 600 mW (Model P-250, Menlo Systems GmbH). The amplifier output is split into two fiber ports A

Fig. 2 Experimental setup based on 1560 nm fiber laser and passive optical delay line. Further components: half wave plate (HWP), quarter wave plate (QWP), and polarization beam splitter (PBS).
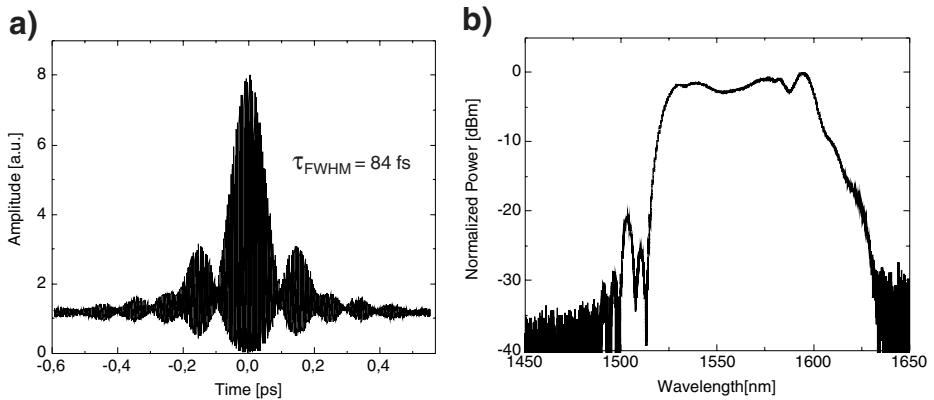
a)

b)
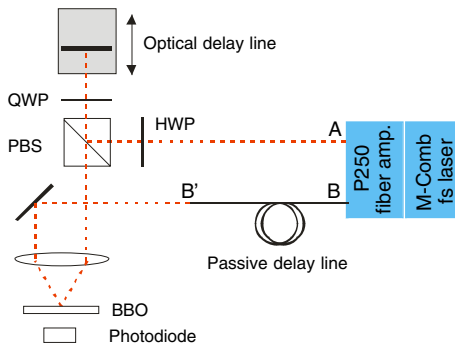
Fig. 3 Autocorrelation trace (**a**) and optical spectrum (**b**) of pulses form Port A.

and B. The linearly polarized pulses from port A with a duration of 84 fs and bandwidth of 74 nm (Fig. 3) are collimated directly after the amplifier. As mention in the previous section, in order to generate long delays, the laser source has to be characterized by a low timing jitter. Typical timing jitter measurements for the M-comb laser (Fig. 4) show values around 10 fs for integration times from several MHz down to 10 kHz. This implies that the OSCAT method can be used without any problem for indices a up to several 10000. Therefore, mainly the length of the passive delay line and the associated nonlinearities will limit the maximum scanning range using the OSCAT technique.

3.1.1 Passive delay line

The piezo stack integrated inside the laser ring can tune the laser repetition rate over 3.5 kHz. In order to generate a time delay of 1 ps according to Eq. 1 the pulse train in the beam B has to be delayed by $a=18$ pulses with respect to the beam A. The resulting optical

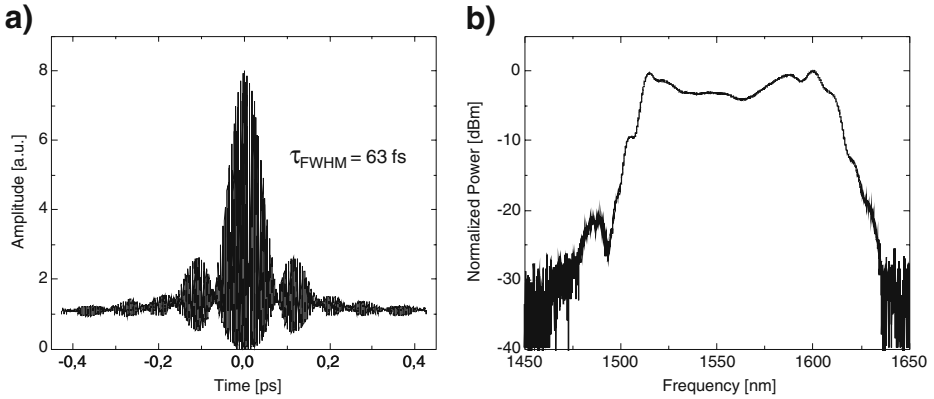Fig. 4 Integrated timing jitter of the free-running M-Comb fiber laser.
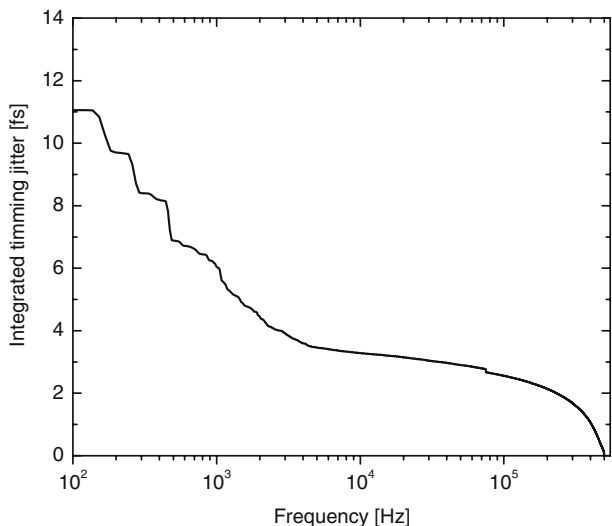
Fig. 5 Autocorrelation trace (**a**) and optical spectrum (**b**) of pulses form Port B after propagation through the fiber delay line.

length of 21.6 m is impractical for free space delay lines. Therefore, we have designed an optical fiber based passive delay line. The 14.4 m long fiber link consist of two fiber types in order to achieve zero group-velocity dispersion operation: a standard single mode fiber and a telecom dispersion shifted fiber. The fiber delay line is connected to the port B of the P-250 amplifier. Figure 5 shows the autocorrelation trace and a spectrum of the pulse train after propagation through the fiber link. The optical pulses with a length of 63 fs have a 3 dB bandwidth of 97 nm. The current design of the passive fiber delay line allows a nearly distortion-free propagation of femtosecond pulses over approx. one hundred meter fiber links.

3.1.2 Cross-correlator

Optical pulses form Port A and Port B' of the passive delay line are guided to the coross-correlator via free space optics. Subsequently, both beams are focused on a 1 mm thick non-linear BBO crystal. The free space paths Port A-BBO and Port B'-BBO are of equal length. The optical power in port A and B is $P_A = P_B = 125$ mW. The frequency doubled signal is
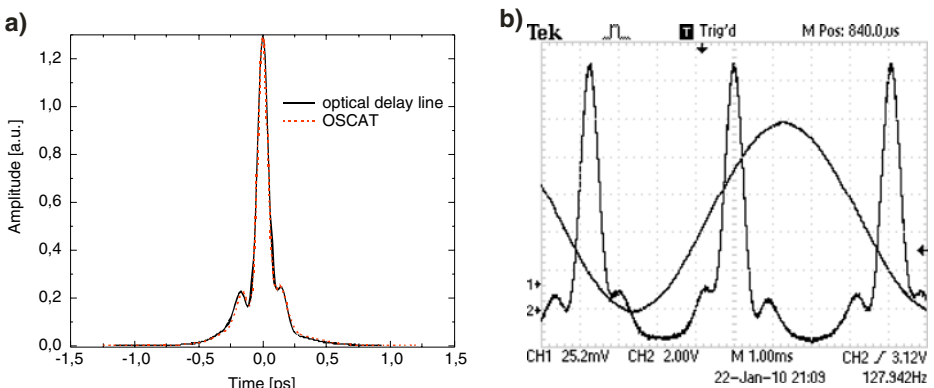

Fig. 6 Cross-correlation signal. Comparison between a conventional mechanical delay line scan and the OSCAT technique (**a**) and a rapid scan with the internal piezo-stack (**b**).

detected by a Si-photodiode. The quarter and half wave-plates (QWP and HWP) as well as the polarization beam splitter cube (PBS) are needed for the reference measurement with the conventional delay line.

4 Experimental results and discussion

For a quantitative comparison of the OSCAT technique, the cross-correlation signal is measured in two ways. In the first experiment the repetition rate is kept constant at 250.028918 MHz. A cross-correlation signal between the pulse i from beam A and the pulse i-18 from port B is measured with standard motorized delay line. The measurement with a scan range of 2.08 ps long was performed with a step size of 10.4 fs. Figure 6a shows the measured cross-correlation trace with a full width at half maximum of 98 fs.

In the second experiment the conventional delay line was in a fixed position and the repetition rate was tuned from $f_1=250.024416$ MHz to $f_2=250.032880$ MHz with an intracavity stepper motor with a repetition rate step width of approx. 28 Hz. The tuning range of 8464 Hz results in temporal delay of 2.438 ps. The measured signals with a conventional delay line (solid line) and the OSCAT technique (dashed line) are shown in Fig. 6a. As can be easily seen, both cross-correlation signals are nearly indistinguishable. The entire tuning range of 2.5 MHz corresponds in over 700 ps time delay. Such a scanning range with a conventional delay line would require high pointing stability of the laser and very careful adjustment of the optical setup. In the case of a fiber coupled OSCAT system the size of the free-space part can be minimized significantly. Moreover, the fiber coupled design guarantees a minimum of beam pointing fluctuations.

One of the most important advantages of OSCAT technique is a capability to perform long scans with fast rates. Figure 6b shows the measured cross-correlation signal using the intracavity piezo stack. The laser repetition rate is being tuned over 3.5 kHz with a rate of 128 scans per second. The resulting 1 ps long cross-correlation trace agrees very well with the slow scan obtained with an intracavity stepper motor or with a conventional delay line. With a larger piezo-stack and a longer passive delay line a scanning range of several tens picoseconds would be possible.

5 Conclusion

Here, we present the novel Optical Sampling by Cavity Tuning (OSCAT) technique for time resolved experiments without any moveable external delay line. The time delay is determined by the repetition rate of the femtosecond laser. Contrary to the ASOPS scheme only one femtosecond laser source is required. Experimental results show an excellent agreement between the measurements with a conventional optical delay line and the proposed OSCAT method. The intracavity piezo stack in the femtosecond laser allows for a scanning range of several picoseconds with a scanning rate in the range of some 100 Hz.

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