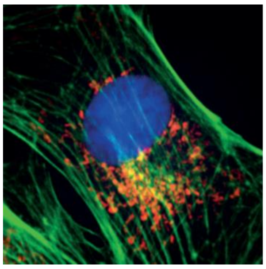
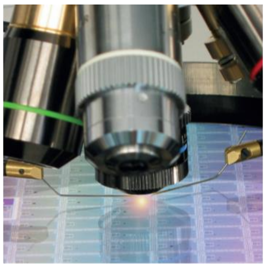



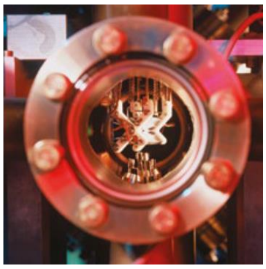

Time-resolved microscopy and spectroscopy using asynchronously synchronized fiber lasers

Synchronized modular laser systems support time-resolved measurements, as well as complex pump-probe methods like ASOPS. The presented laser system is used to measure Kerr-rotations by inducing a precession motion of the spin magnetization in ferromagnetic materials, which in turn leads to emission of microwave radiation that rotates the polarization of temporally delayed probe pulses. This way, the temporal evolution of the spin precession motion is detected.

Keywords: femtosecond lasers, ultrafast lasers, fiber lasers, ASOPS, laser synchronization, microscopy, spectroscopy

Introduction

Microscopy and spectroscopy allow to discover and study new phenomena in chemistry, biology, medicine and physics by using coherent light as source for excitation and detection of the (electro-) optical response of the scanned sample. Using pulsed lasers, dynamic processes can be visualized with temporal resolutions of several femtoseconds and of microscopic spatial scale.

Whereas many time-resolved or nonlinear spectroscopy and microscopy techniques have used titanium-sapphire lasers (Ti:sapph) in the past, recent advancements in laser technology have made fiber lasers equal alternatives. Their flexibility and modularity, combined with robust and reliable operation make them powerful tools. For multicolor or pump-probe measurements, several pulsed fiber laser outputs can be driven using only one mode-locked oscillator. This results in synchronized, phase-stable pulses between the individual laser outputs.

Furthermore, commercial fiber lasers reliably generate pulses with durations below 30 fs, excellent beam quality and best stability between consecutive pulses. Due to the large surface of the amplifier fibers, very effective heat dissipation and high-energy conversion efficiency can be reached. Saturable Absorber for passive mode-locking lead to extremely low intensity noise. This enables to build cost-effective and highly efficient laser systems that are ideally suited for time-resolved and nonlinear microscopy.

Fiber lasers for spectroscopy and microscopy

TOPTICA Photonics developed a system of several femtosecond fiber lasers that are synchronized asynchronously especially for time-resolved microscopy and spectroscopy. The presented laser system is a sophisticated pump-probe excitation source based on asynchronous optical sampling (ASOPS). The synchronization electronics was provided by Laser Quantum.

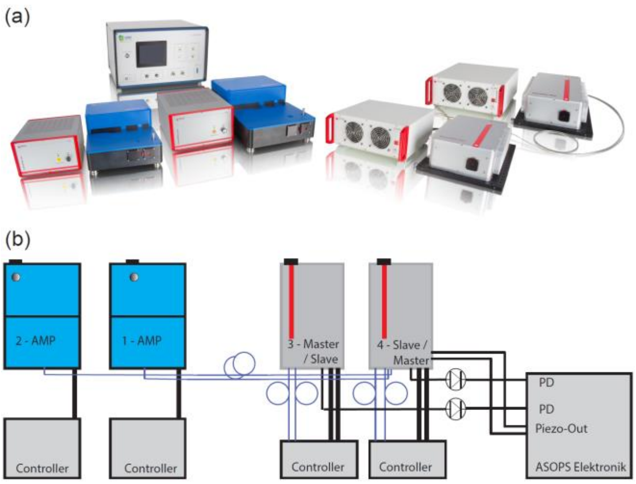

Figure 1: The full laser system (a and b) with its electrical (black) and optical (blue) connections: Each of the oscillators acts as Master or Slave for asynchronous optical pump-probe experiments. Both amplifiers (AMP) can be optically seeded by the Maser/Slave-system. They are spectrally tunable from 485-640 nm and from 830-1100 nm. The pump diodes for the main amplifiers are located in an external control unit for each laser for thermal decoupling. The ASOPS unit is connected to the Master/Slave lasers to enable synchronization of the pulse trains of both lasers.

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The ASOPS electronics synchronizes four individual fiber lasers (see Figure 1). Two of these lasers are fixed-frequency high-performance fiber lasers of TOPTICA's product line FemtoFiber ultra. They provide laser pulses at a 780 nm central wavelength with 0.5 W average power and 150 fs, respectively 1050 nm with 5 W and < 120 fs. The pulse duration can be tuned by a motorized compressor via the graphical user interface which allows compensation of the dispersion of the optical system (i. e. microscope objective). The lasers are equipped with specialized oscillators that modulate the individual repetition rate by varying the resonator lengths using piezo elements. This way they can be synchronized to one fixed repetition rate to realize ASOPS. The offset-frequency (ASOPS frequency) can be set using Laser Quantum's TL-1000-ASOPS unit for pulse synchronization.

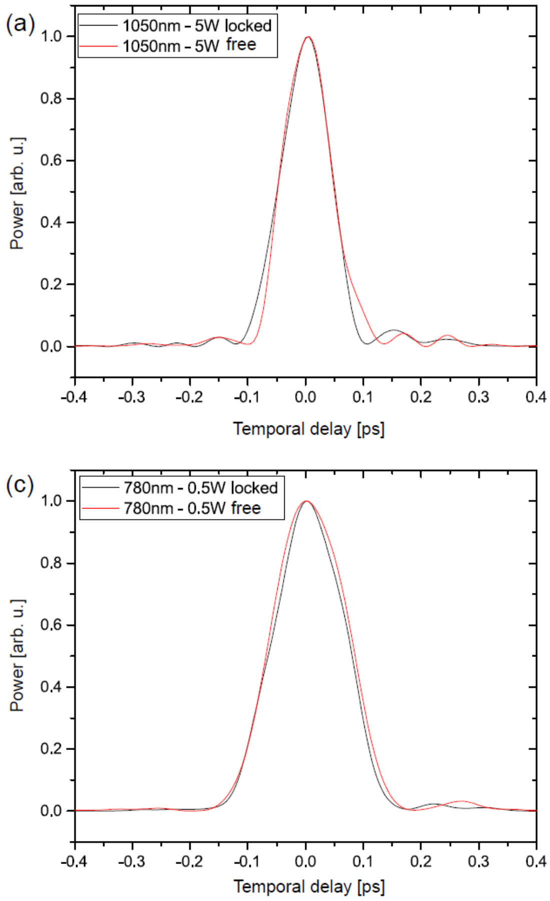

Figure 2: Neither the ASOPS-synchronization, nor output power scaling have an effect on the pulse duration of the Master/Slave lasers. The pulse shape was reconstructed using a numerical fit of the interferometric autocorrelation of the respective pulses.

The described variations of the resonator length (and therefore the repetition rate) have no influence on the pulse duration or on the intensity of the laser pulses (see Figure 2). In addition, the pulse duration of the FemtoFiber ultra lasers stays constant over the full power range, i.e. 0.5 W at 780 nm, as well as 5 W at 1050 nm.

High-resolution pump-probe experiments using synchronized lasers require very low intensity noise in the observed frequency band. This is enabled by the patented SESAM-based oscillator technology of TOPTICA's FemtoFiber lasers, c.f. Figure 3.

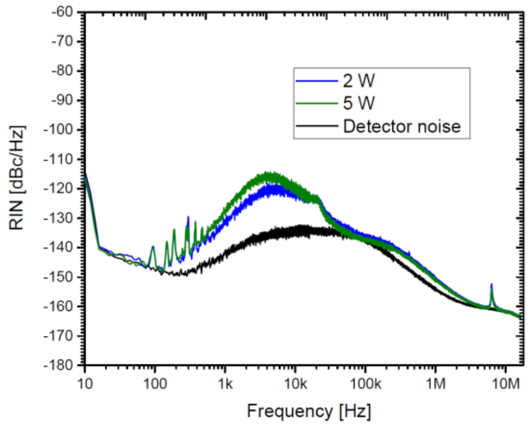

Figure 3: The relative intensity noise (RIN) of the FemtoFiber ultra 1050 fiber laser in the frequency region between 10 Hz and 10 MHz is compared to the detector noise (black curve).

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The oscillator pulses either of the Femto Fiber ultra 780 or 1050 seed up two additional, tunable laser amplifier systems. These lasers are designed to cover a broad wavelength tuning range in the visible spectral range (FemtoFiber pro TVIS, 485 – 640 nm) and in the near-infrared spectral range (FemtoFiber pro TNIR, 930 – 1100 nm), c.f. Figure 4.

For this modular laser system, the Master and Slave oscillators are freely selectable. In addition, for each of the two amplifiers the Master oscillator, as well as the Slave oscillator can be used as seed independently. With this flexible setup, experiments with up to four synchronized excitation pulses are feasible. Alternatively, two completely independent pump-probe experiments can be carried out.

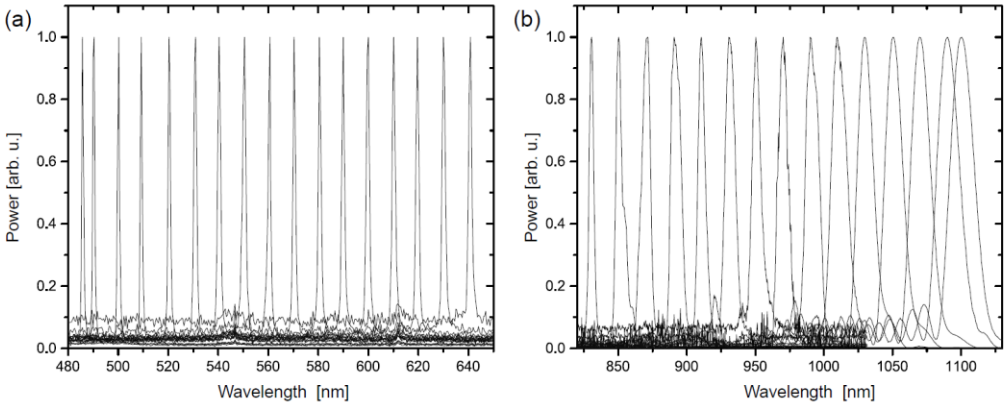

Figure 4: The amplifier systems support a relatively broad wavelength tuning from 485 to 640 nm (a), and 830 to 1100 nm (b), respectively.

ASOPS

Laser Quantums synchronization electronics is based on patented DDS-technology (direct digital synthesis). It reaches temporal resolutions of less than 100 fs in a measurement interval of 1 ns, which was achieved using two 1 GHz Ti:sapph oscillators. The temporal resolution using ASOPS is limited by the repetition rate offset of pump and probe lasers following:

$$\Delta\tau = \Delta f_R / f_R^{\ 2} \text{ (Eq. 1)}$$

The temporal scan range is therefore only limited by the interpulse distance (i.e 10 ns for 100 MHz repetition rate) of the emitted pulse trains without the use of an additional mechanical delay. The probing of the pulse is done at the sampling frequency Δf_R. For data acquisition the intensity of the probe beam is digitally monitored and rescaled by multiplying the data with the factor $\Delta f_R / f_R$ (c.f. Figure 5). As a consequence the ASOPS technology is not influenced by electronic noise below Δf_R. Therefore lock-in detection is not required in most cases.

Eq. 1 shows that high repetition rates are beneficial for the temporal resolution and the sampling frequency using ASOPS. However, at repetition rates in the 100 MHz range and below the temporal resolution is on the order of slightly more than 100 fs (180-320 fs for 80 MHz system over the whole temporal delay of 12.5 ns, c.f. Figure 6). This relatively low repetition rates featuring interpulse distances of 10-20 ns are required for the visualization of longer decay times.

To fully characterize the utilized ASOPS system it is essential to know the temporal resolution as function of pulse delay leading to best performance over the full scan range allowing for the detection of fast decay times far off the time zero (pulse overlap of pump and probe). This is only feasible using the DDS technology employed by Laser Quantum within their TL – 1000 - ASOPS unit which outperforms other technologies like using two independent reference signals to stabilize two laser. This concept suffers from random phase noise while it was shown that for the use of single-sideband signal as reference the temporal resolution is significantly increased with increasing temporal delay.

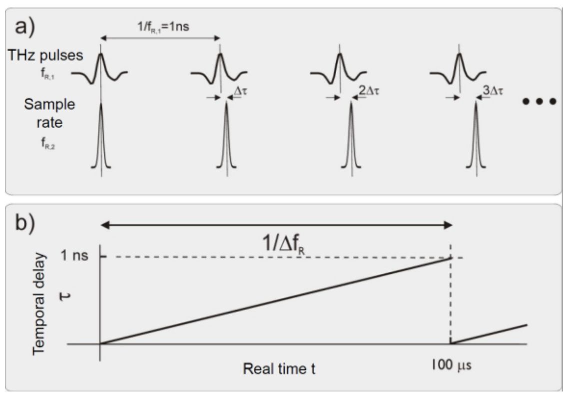

Figure 5: Asynchronous optical sampling is performed using a combination of two lasers with an optically generated terahertz-pulse (1 GHz repetition rate, 10 kHz difference frequency). The temporal resolution τ increases linearly with the time. This way, the complete measurement region is sampled between two consecutive laser pulses.

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The fiber lasers in this experiment are synchronized using the 125th harmonic of their repetition rate (80 MHz). The performance of the system is characterized by measuring a cross-correlation of a double-pulse with variable temporal delay of one of the two lasers. This way, the achievable temporal resolution is determined depending on the position in the temporal measurement interval.

At small temporal delay, the FWHM of the cross-correlation response is about 180 fs, which corresponds to two convoluted Gauss-shaped pulses with 150 fs and 120 fs. For delay settings between 50 and 100 ps the cross-correlation FWHM increases to about 320 fs. This is due to uncompensated phase noise between both lasers in the frequency interval between 1 Hz and 10 kHz.

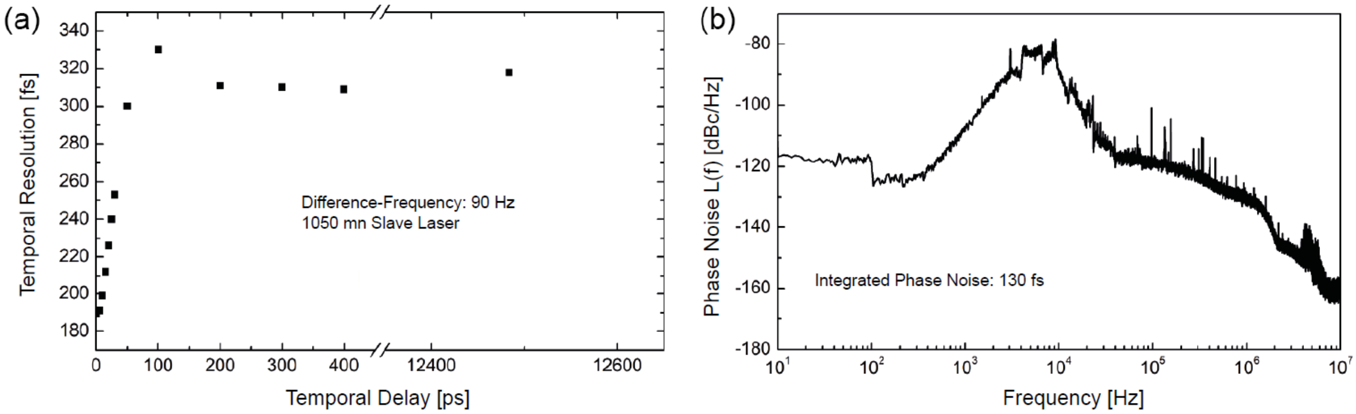

Figure 6: Full width at half maximum of the cross correlation of both fiber lasers as a function of the position in the measurement region during asynchronous optical sampling (a), and phase noise between both lasers during stabilized operation (b).

The presented laser system is used to measure Kerr-rotations. In these experiments, powerful pump pulses induce precession motion of the spin magnetization in ferromagnetic materials. This precession motion leads to the emission of intense microwave radiation, which in turn rotates the linear polarization of probe pulses. This polarization rotation is detected as a function of the temporal delay between the pump and probe pulses. With these methods, the temporal evolution of the spin precession motion is detected.

Previously, the temporal delay could be varied only using mechanical delay stages with a relatively low accuracy of about 1 ps. This method also requires long measurement intervals since the mirrors of the delay stage have to be displaced to change the temporal delay between pump and probe pulses.

With an ASOPS setup, no mechanical delay stages are necessary. The detected signal is Fourier transformed to obtain the temporal evolution of the spin precession motion. Since the Fourier transformation requires only little acquisition times, an excellent signal-to-noise ratio is achieved.

The presented laser system allows manipulating the microwave signal with additional laser pulses. This way, the setup enables a unique flexibility for pump-probe measurements or multicolor excitation. It shows the excellent variability and modularity of modern fiber lasers for applications in time-resolved microscopy. Synchronized modular systems not only support time-resolved measurements, they also enable complex pump-probe methods like ASOPS.

Fiber lasers provide the necessary flexibility and tunability, as well as the required pulse parameters. Furthermore, fiber lasers are very user-friendly: Their “push-button turnkey operation” guarantees a straightforward initialization of the lasers without the necessity for alignment. In addition, state-of-the-art fiber lasers operate maintenance-free and they do not require expensive laboratory infrastructure like water cooling or high-voltage current. This way, the user can fully concentrate in their experiment.

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