Fiber-coupled high-speed asynchronous optical sampling with sub-50 fs time resolution

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Abstract: We present a fiber-coupled pump-probe system with a sub-50 fs time resolution and a nanosecond time window, based on high-speed asynchronous optical sampling. By use of a transmission grism pulse compressor, we achieve pump pulses with a pulse duration of 42 fs, an average power of 300 mW and a peak power exceeding 5 kW at a pulse repetition rate of 1 GHz after 6 m of optical fiber. With this system we demonstrate thickness mapping of soft X-ray mirrors at a sub-nm thickness resolution on a cm² scan area. In addition, terahertz field generation with resolved spectral components of up to 3.5 THz at a GHz frequency resolution is demonstrated.

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References and links
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1. Introduction

A wide range of scientific experiments and industrial applications nowadays involve ultra-short optical pulses. In many fields like microscopy [1–4], coherent anti-Stokes Raman spectroscopy [5] or terahertz spectroscopy [6–9] fiber-coupled pulse delivery is advantageous compared to free-space setups. For broadband pulses this is challenging, because the pulses experience spectral and temporal distortions during propagation through the fiber. These effects can be divided into dispersive and nonlinear effects. Dispersion leads to temporal stretching of the pulse, whereas nonlinear effects influence the optical spectrum in dependency on the pulse intensity [10]. At peak powers below 10 kW and mode field diameters above 5 μm the dominating nonlinear effect is self-phase modulation. At much higher powers or smaller fiber cores intrapulse Raman-scattering and self-steepening also need to be taken into account.

Because many applications are limited to the available pulse duration after the fiber, a lot of efforts in fiber production have been made to reduce either the dispersion (microstructured fibers) or the nonlinearity (large-mode-area fibers, LMA fibers) of the fiber. With hollow core photonic crystal fibers (PCFs) the dispersion around 750 nm could be reduced to below 1 fs²/mm by introducing a radial chirp in the cladding [11, 12]. Using this fiber, the shortest pulses achieved so far with sub-20 fs duration were produced at peak powers of about 7 kW by adjusting the laser wavelength to the PCF guiding window. Commercial available PCFs for Ti:Sa laser light focus on white light generation in the anomalous dispersion regime with a zero dispersion wavelength at around 750 nm [13, 14]. Recently, Kagomé-style hollow core PCFs with high transmission and low dispersion for ultrafast applications were demonstrated [15, 16]. Another approach employing soliton formation in comparatively long hollow core fibers (8 m) by using regenerative amplifiers with pulse energies above 50 nJ resulted in pulse durations below 100 fs [17, 18]. For industrial applications incorporating long-range optical fibers, the advantage of a tunable dispersion is outbalanced by the large losses (>5 dB/m) and the costs of PCFs. That is why fused silica single mode (FSSM) fibers are still most frequently utilized, despite their high chromatic dispersion at around 800 nm.

The most common way for flexible femtosecond pulse delivery without amplification is using one or more dispersion compensating devices like prisms, gratings and chirped mirrors in front of a FSSM fiber [19]. The dispersive optics need to compensate second- and third-order chromatic fiber dispersion (GDD and TOD), while showing small and frequency-independent transmission loss. These conditions can be fulfilled by using a compact combination of prisms and gratings, called grism [20, 21]. The typical grism design is based on two reflection gratings engraved or placed close to two prisms [22, 23]. If used near the Littrow angle of the gratings, the efficiency of this device is limited by the efficiency of the gratings and the acceptance angle.
of the anti-reflection coating of the prisms (since the beam travels through every prism interface twice under a different angle) [24]. Although efficiency scaling of the reflection grisms is mentioned often, experimental results for pulse delivery with efficiencies above 40% using the combination of a grism and FSSM fibers have not been reported. Using a broadband laser source, a reflection grism was shown to support sub-35 fs and sub-30 fs pulses with nJ pulse energy after 3 m of FSSM and LMA fibers, respectively [25]. Very recently, the same group demonstrated pulses with a record peak power of 50 kW at a sub-30 fs pulse duration after 2 m of polarization maintaining fused silica single mode (PMFSSM) fiber incorporating fiber-based spectral broadening of the input pulse [26].

Here we make use of a highly transmittive grism design (>60% efficiency in double pass configuration) in combination with GHz repetition rate lasers, resulting in sub-50 fs pulses after 6 m of PMFSSM fiber with an average power of up to 300 mW (corresponding to a pulse energy of 0.3 nJ and a peak power of more than 5 kW). We apply these pulses in laser-based high-frequency ultrasonic and THz time-domain experiments. Both experiments are based on the asynchronous optical sampling technique, in which sub-ms scanning times for a nanosecond time window and thus high signal-to-noise can be achieved as a direct result of the high pulse repetition rate.

2. Fiber pulse delivery

The transmission grism device used here was proposed and demonstrated by the group of Tournois in a kHz chirped pulse amplification system [27, 28], where the ratio of second- and third-order material dispersion is $\phi_3/\phi_2 \approx 0.45$ fs. We adapted this configuration and show dispersion compensation of second- and third-order in 6 m long fused silica fibers, where the TOD/GDD-ratio is almost double this value. The fiber dispersion was assumed to be the one of pure fused silica with $\beta_3/\beta_2 = 28/35$ fs and $\beta_4 = 13$ fs$^4$/mm at around 810 nm. According to dispersion data of the manufacturer this assumption is valid, although FSSM fibers tend to show stronger dispersion than bulk fused silica. Numerical pulse propagation simulations reveal, that a slight modification of the fiber dispersion parameters has negligible effects on the resulting pulse parameters after the fiber when readjusting the grism compressor accordingly.

The layout of the compressor is shown in Fig. 1. We use two anti-reflection coated SF10 prisms with an apex angle of 45°, placed between two pre-existing volume phase holographic transmission gratings optimized for high efficiency (>94%, Kaiser Optical Systems, Inc.) with a grating period of 1200 lines/mm. The angles (i.e. incident angle $\theta_i$, refracted angle $\theta_d$ and $\varepsilon$) are connected via the grating equation and Snell’s formula. The choice of the prisms was based on group delay calculations of the grism compressor with different prism materials and apex angles [27]: A strong negative third-order dispersion contribution is achieved when using highly dispersive prism materials while keeping $\varepsilon$ large and reducing the distance H. This way, the third-order dispersion of the fiber can be compensated. With $G_1 + G_2 = 6$ mm, $H = 5$ mm, $L = 25$ mm, an incident angle of 30.1° and 6 m of FSSM fiber, zero net second- and third-order dispersion and a remaining net fourth-order dispersion (FOD) of about $-16 \times 10^5$ fs$^4$ was calculated. The design allows for easy alignment while working at the Littrow angle. The efficiency of the compressor in double-pass configuration was 62% in comparison to about 72% without the prisms. The grism did not introduce any measurable change in the optical spectrum. The chirped pulses were launched into 6 m of polarization maintaining FSSM with a core diameter of 4.5 μm (Thorlabs, PM780-HP) by an aspheric focusing lens with a focal length of 7.5 mm. Including the fiber coupling, the overall transmission efficiency amounted to 42%. A loss in spectral width of about 1-2 nm during injection into the fiber core was measured, possibly due to imperfect alignment of the compressor that results in a remaining spatial chirp. Alignment was done in a step-by-step manner: First, the two gratings were aligned.
Fig. 1. Layout of the compact grism compressor. VPH: volume phase holographic transmission grating, P: prism. The whole compressor has a $50 \times 40 \text{mm}^2$ footprint.

To simulate the pulse propagation in the fibers, we numerically solved the Nonlinear Schrödinger Equation (NLSE) with a split-step-Fourier procedure [10, 29], taking into account the full Raman integral and dispersion up to fourth-order. As simulation parameters we used a nonlinearity of $\gamma = 10 \text{ W}^{-1}/\text{km}$ and the dispersion values of bulk fused silica. Based on the optical spectrum of the laser, we assumed a 30 nm bandwidth at a center wavelength of 812 nm and a sech$^2$ shape. It should be mentioned, that the simulation includes 5 mm of bulk lens material after the fiber to account for the focusing lens that was used in the experiments. Pulse characterization was carried out using a home-made second harmonic interferometric autocorrelator and a spectrometer, placed after the fiber.

The best agreement between the numerical results and the measurements is achieved when using zero net second- and third-order dispersion and a net fourth-order dispersion of $\approx -9 \times 10^5 \text{ fs}^4$ in the simulations, which is smaller than expected from the group delay calculations. Although ambiguities in interferometric autocorrelations prohibit the extraction of higher-order dispersion values, the remaining net fourth-order dispersion could be less than expected. This could result from the angle adjustment in the alignment procedure that leads to a deviation from the layout symmetry depicted in Fig. 1. The results of the numerical simulations in comparison to the measured autocorrelations at an average output power of 300 mW after the fiber are displayed in Fig. 2: Figures 2(a) and 2(b) show the time-dependent power and the optical spectrum of the output pulse, having a flat spectral phase and clean shape with only little sidewings originating from net fourth-order dispersion. As can be seen by the spectral phase, the minimum pulse duration is achieved when little positive net GDD and negative FOD cancel each other to a certain degree. Since nonlinear effects introduce an additional spectral phase, the amount of net GDD for minimum pulse duration depends on the pulse energy and needs to be evaluated numerically. In this case, the net GDD is around 1500 fs$^2$. The width (full width at half maximum) of the intensity autocorrelation measurement yields 65 fs. Assuming a sech$^2$ shape, this corresponds to a pulse duration of 42 fs with a peak power of more than 5 kW. The peak power of the bandwidth-limited input pulse is roughly twice this value. Figures 2(c) and 2(d) compare the measured autocorrelations (interferometric and intensity) with the results of the numerical calculations, showing excellent agreement. The pulse duration was limited by remaining FOD and nonlinearities in the fiber. The presence of nonlinearities can be seen by comparing input and output optical spectra, as shown in Fig. 2(e). Spectral narrowing [30]
amounts to about 50% at maximum output power. To investigate the scaling capabilities of the presented system, we also measured the minimum pulse duration together with the bandwidth for different average powers. For each measurement point, slight readjustments of the grism compressor need to be done to achieve minimum pulse duration after the fiber due to the change in the spectral phase by SPM. Results can be seen in Fig. 2(f). Over the whole accessible power range the pulse duration (dots) shows a linear increase of only 10 fs, whereas the spectral width (crosses) decreases in a more radical fashion. Both behaviours are in agreement with the prediction of the numerical simulations (dotted lines).

The strong influence of third-order dispersion compensation on the pulse profile is revealed in Fig. 2(a). If the two gratings of the grism are applied in a grating compressor configuration, the third-order dispersion of the fiber and the compressor add to about $6 \times 10^5 \text{ fs}^3$. This amount of dispersion will highly disturb the pulse profile after the fiber, which is shown in the inset of Fig. 2(a). In fact, 50% of the whole pulse energy is shifted into emerging pulse echos, with the peak of the first echo being located at a time delay of about 150 fs after the main peak. Even after 500 fs the amplitude of the echos still reaches 10% of the central intensity. The peak
intensity is lowered by a factor of more than 5 compared to the grism configuration (below 1 kW instead of more than 5 kW). For pump-probe experiments like broadband THz time-domain spectroscopy and laser-induced high-frequency acoustics such distorted pulses will cause a tremendous reduce in measurement bandwidth and signal-to-noise, making these applications unattractive due to inevitable huge measurement times.

So far, the results apply to pulse trains with gigahertz repetition rate. Although these lasers limit the output pulse energy and the pulse peak power, the GHz repetition rate directly yields a high signal-to-noise ratio in high-speed asynchronous optical sampling experiments. In some cases, using common 80 MHz oscillators can be favourable. Ultrashort fiber pulse delivery is more challenging for these systems, due to the higher pulse energy and the correspondingly stronger nonlinear interaction in the fiber. Thus, PCFs and LMA fibers are often employed to reduce pulse distortions by self-phase modulation. We numerically investigated the applicability of the presented grism compressor for fiber delivery of pulses with several nJ energy. While the pulse energy and the nonlinear parameter are varied in the simulation, the remaining parameters are the same as above. The results are summarized in Fig. 3. Figure 3(a) shows the peak power in dependency of the injected pulse energy for different $\gamma$ values. For $\gamma = 10 \ W^{-1}/\text{km}$, which corresponds to the investigated single mode fiber, the peak power strongly saturates at about 20 kW due to SPM. Large mode area fibers exhibit values of about $\gamma \approx 1 \ W^{-1}/\text{km}$, thus allowing for higher peak powers and shorter pulse durations. The simulations reveal record peak powers of more than 60 kW at 4 nJ pulse energy when using a LMA fiber. In the linear regime ($\gamma = 0 \ W^{-1}/\text{km}$) a 4 nJ pulse energy corresponds to more than 90 kW peak power. Figure 3(b) compares two resulting pulse shapes. Whereas SPM in FSSM fiber results in a complex and broad pulse structure, the pulse duration in a LMA fiber is below 50 fs and the shape is almost free of nonlinear distortion. These results demonstrate the scaling capabilities of the presented system to deliver highly energetic ultrashort pulses after 6 m of optical fiber. Due to the high efficiency of the presented grism compressor, an experimental demonstration of delivered pulse energies up to 4 nJ and peak powers exceeding 60 kW should be straightforward. This would represent the highest peak power from a fiber-coupled 80 MHz laser system without any additional amplification so far and meet the increasing demands of many applications like multiphoton endoscopy and THz spectroscopy for high-power and flexible ultrashort
pulse delivery.

3. Fiber-coupled ASOPS experiments

To demonstrate the use of the flexible ultrashort pulse delivery system, we show pump-probe measurements with the compressor-fiber setup described in section 2. In typical pump-probe schemes a beam splitter is used to split the beam into pump and probe beam. A mechanical delay line is then implemented for generating the time delay between pump and probe pulses. The delay line limits the measurement in terms of speed and mechanical noise and requires the use of a lock-in amplifier. Especially for the sensitive fiber-coupling it is beneficial to circumvent any mechanical moving device like a delay stage. In our experiment we make use of a high-speed pump-probe system, based on two gigahertz repetition rate Ti:Sapphire lasers in a master-slave configuration with a stabilized repetition rate difference $\Delta f$ of some kHz [31]. In this so-called asynchronous optical sampling (ASOPS) scheme, the time delay between successive pulse pairs from both lasers increases linearly with time without any moving optomechanics. With an appropriate $\Delta f$-stabilization, the whole time window of 1 ns can be used for pump-probe measurements at a time resolution below 50 fs, limited only by the pulse duration if the timing jitter is below this value [32]. The layout of the setup is shown in Fig. 4. Each of the two oscillators was fiber-coupled with the transmission grism and 6 m PMFSSM fiber. The fiber facets were equipped with FC/APC connectors with an $8^\circ$ cleave to prevent back reflection. This way, the system could be operated without the use of optical isolators. Half wave plates were used to optimize the efficiency of the compressor. The polarization maintaining fibers were adjusted for propagation along a single axis of the fiber. Pump and probe pulses are centered at a wavelength of around 810 nm and 790 nm, respectively. The dispersion compensation was adjusted independently for minimum pulse duration. First, we measured the time resolution via cross-correlation of pump and probe beam in a 200 $\mu$m thick gallium phosphide
crystal. The deconvoluted width of the two-photon absorption signal at a repetition rate offset of $\Delta f = 2 \text{ kHz}$ was determined to be roughly 45 fs. Based on earlier results [32], residual timing jitter can be neglected. Therefore the time resolution can be assumed to be constant over the whole 1 ns time window.

As an application example, we demonstrate the thickness inspection of dielectric Bragg mirrors for the extreme ultraviolet (EUV) range. The potential of the EUV lithography to further scale the integration density of silicon chips relies on the availability of highly-reflective silicon/molybdenum (Si/Mo) multilayer mirrors for radiation at a wavelength $\lambda$ of 13.5 nm [33, 34]. The thickness of the individual mirror layers must be around $\lambda/4$, resulting in a superlattice period of $\lambda/2$. For use as highly reflective mirrors in a lithography system, the layers must therefore meet sub-nm thickness tolerances on several 100 cm$^2$ areas, thus calling for reliable and fast in-situ growth control systems. Common inspection tools are based on X-ray reflectometry [35, 36]. As an alternative we demonstrate thickness mapping of EUV mirrors via laser-based generation and detection of picosecond ultrasonic. This technique has been used intensively over the last years to extract essential material parameters of a variety of materials and structures [37, 38]. The absorption of an ultrashort laser pump pulse induces a picosecond stress pulse that propagates into the material with longitudinal sound velocity. Interfaces and inhomogeneities partly reflect the ultrasonic pulse. The back traveled part induces a reflectivity change at the surface via the elasto-optic effect and can be detected with a laser probe pulse [39]. By analyzing the acoustic response of the multilayer structure to optical excitation, the homogeneity of the structure can be revealed.

The setup for the inspection of soft X-ray mirrors is depicted in Fig. 4(b). Pump and probe beams are coupled out of the fiber and collinearly overlapped in an enclosed measurement head. A microscope objective is used to focus the beams onto the sample. The back reflected probe light is coupled into a multimode fiber for detection. This part of the setup highly benefits from the well-defined state of polarization of the light at the fiber exit, since the collinear overlay and the separation of the pump- and the probe light before and after reflection at the sample is based on polarization sensitive elements. Utilizing standard single mode fibers would lead to a reduced maximum pump power at the sample and power instabilities when moving the fiber, since the polarization state of the light transmitted through the FSSM fiber is sensitive to mechanical disturbance. Pump and probe beams are aligned in a way to prevent back coupling into the fibers, which would influence the laser operation. The whole detection unit has a footprint of $12 \times 12 \text{ cm}^2$ and can be scanned along the sample for laterally resolved measurements. The lateral resolution of the measurement is given by the laser spot size, which can have values below 1 $\mu$m when using microscope objectives with high numerical aperture. At the full available power of 300 mW this corresponds to a pump fluence of more than 25 mJ/cm$^2$, giving access to a broad range of applications.

Our sample consists of 60 Si/Mo layers sputter-deposited on a mono-crystalline Si wafer. The superlattice period is 6.8 nm, leading to a total stack thickness of 408 nm. Pump and probe powers were adjusted to 150 mW and 15 mW, respectively. The measured beam diameters at the sample were below 10 $\mu$m. By introducing more complex microscope objectives, the spot sizes can be further reduced to below 1 $\mu$m. $\Delta f$ was set to 8 kHz. An exemplary ASOPS ultrasonic measurement result is shown in Fig. 5(a). At zero time delay a strong reflectivity change, followed by an exponential decay with a few ps time constant due to electron relaxation in the Mo layers, is visible. On top of that, a smaller signal contribution can be seen at short time delays in form of a fast damped oscillation with a frequency $f_{\text{ph}}$ in the terahertz range and an acoustic echo at around $\tau_{\text{echo}} = 130 \text{ ps}$. Both result from the generation of coherent acoustic phonons in the multilayer structure. The first acoustic signal part is generated by localized surface modes [40]. The latter one is the reflection of the stress pulse at the substrate. In a lin-
ear approximation of the dispersion relation of the Si/Mo superlattice they can be connected to the superlattice period \( d_{sl} \) and the total stack thickness \( d_{tot} \) with the longitudinal sound velocity \( v_l \) in Si/Mo via \( d_{sl} = v_l/\tau_{ph} \) and \( d_{tot} = v_l \times \tau_{echo}/2 \). We estimate the sound velocity \( v_l \) with the position of the acoustic echo and the total thickness provided by the manufacturer to be 6180 m/s. By moving the measurement head along the sample, we can laterally map the structure point-by-point. Reducing the number of averages to \( 5 \times 10^4 \) lead to a measurement time of roughly 6 seconds per pixel. Figure 5(b) shows the extracted superlattice period of \( 100 \times 100 \) ASOPS measurements along a square grid with a pixel-to-pixel distance of 100 \( \mu \)m. Statistics with the 5000 neighboring pixels close to the sample center lead to a superlattice period of 6.161\( \pm \)0.006 nm. The deviation of this value to the expected value of about 6.8 nm can be explained by the linear approximation of the dispersion relation and the uncertainty of the sound velocity in Si/Mo multilayers. Towards the wafer edge the superlattice period shrinks due to inhomogeneities in the growth process. Given the results above, fiber-coupled ultrasonic measurements can be used for non-invasive inspection of soft X-ray mirrors on larger scales with sub-nm thickness resolution, few-\( \mu \)m lateral resolution and high speed.

In a second experiment, we address the generation and field-resolved detection of terahertz radiation. The non-invasive nature of the terahertz radiation gave rise to a broad range of applications [41]. For implementation in an industrial environment, the flexibility of optical fibers is essential. This is especially true when probing large, heavy and sensitive samples (for example during production process), where moving the device under test is undesirable. Thickness monitoring in drying paint films and polymers or art conservation are just a few examples [42–44].

The schematic layout of the setup can be seen in Fig. 4(c). THz pulses were generated in a large-area photoconductive emitter and electro-optically detected in a 500 \( \mu \)m thick \( <110> \)-cut ZnTe crystal using a single photodiode [45, 46]. Whereas the generation of the terahertz radiation is not influenced by the the pump beam polarization, the electro optical detection relies on a well-defined state of polarization of the probe beam. Hence, an additional polarizer was used
Fig. 6. (a) Terahertz transient. Inset: Zoom into the transient (b) Dynamic range at a measurement time of 2 s (black), 200 s (red) and 200 s using only the 50 ps time window centered around the main peak with a Gaussian window for the Fourier transformation (blue). Data is normalized to the noise floor.

to suppress any DC contribution in the measured photocurrent from remaining non-polarized probe light. Replacing the PMFSSM fibers with standard FSSM fibers leads to a decreased transmission of the probe beam through the polarizer and therefore a smaller photocurrent. Beam guiding was carried out with gold-coated parabolic mirrors. Pump and probe fluence at the full available power of 300 mW of the pump and about 200 mW of the probe beam in the generation and detection material was around 5 μJ/cm² and 3 μJ/cm², respectively. Dry air was used to eliminate residual absorption lines from water vapour in air. The measurement results of these experiments are displayed in Fig. 6. The time-domain data shows a picosecond transient at around zero time delay with additional echos at 11 ps and 16 ps from internal reflection in the detection and the emitter material, respectively. In Fig. 6(b) the dynamic range of the transients for two different number of averages, obtained by fast Fourier transformation of the time-domain data, is displayed. The peak frequency is around 0.9 THz. The echos in the time-domain lead to Fabry-Perot oscillations on top of the spectrum. At 2 s measurement time (black curve), the maximum visible frequency is around 3 THz. With a factor 100 more averages (red curve), higher frequencies can be resolved. We also display the spectrum for the case of only using the time interval from -25 ps to +25 ps for the fast Fourier transformation and a Gaussian window function (blue curve). In this case, the frequency resolution is 20 GHz instead of 1 GHz. Therefore Fabry-Perot oscillations show smaller amplitude and some smaller absorption lines around 2.8 THz from remaining water vapour cannot be resolved anymore. On the other hand signal-to-noise is further improved, thus the dynamic range reaches more than 60 dB for frequencies of up to 2.5 THz and frequency components approaching 4.5 THz can be resolved.

4. Conclusion

In summary, we demonstrated a fiber-coupled pump-probe system, based on high-speed asynchronous optical sampling. Pump pulses as short as 42 fs at a center wavelength of 810 nm and a gigahertz repetition rate with peak powers exceeding 5 kW and an average power of 300 mW were realized after 6 m of fused silica single mode fiber. Dispersion compensation of second- and third-order was achieved with a compact and efficient transmission grism. Scaling to pulse energies up to 4 nJ and its limitations was discussed and supported by numerical pulse propagation simulations, showing the feasibility of peak powers of more than 60 kW when 80 MHz oscillators are being employed. We demonstrated the system capabilities by thickness mapping...
of silicon/molybdenum Bragg mirrors with sub-nm thickness resolution on a cm$^2$ lateral scale. In a second experiment, we implemented a terahertz time-domain scheme. Spectra with frequencies exceeding 4 THz and a dynamic range of more than 60 dB in the 0.2-2.5 THz region have been obtained at moderate measurement times. At the full frequency resolution of 1 GHz, the highest visible frequencies were around 3.5 THz.

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