High-speed asynchronous optical sampling with sub-50fs time resolution

R. Gebs,* G. Klatt, C. Janke, T. Dekorsy, and A. Bartels

Department of Physics and Center for Applied Photonics, University of Konstanz, D-78457, Germany
*Raphael.Gebs@uni-konstanz.de

Abstract: We report an ultrafast time-domain spectroscopy system based on high-speed asynchronous optical sampling operating without mechanical scanner. The system uses two 1 GHz femtosecond oscillators that are offset-stabilized using high-bandwidth feedback electronics operating at the tenth repetition rate harmonics. Definition of the offset frequency, i.e. the time-delay scan rate, in the range of a few kilohertz is accomplished using direct-digital-synthesis electronics for the first time. The time-resolution of the system over the full available 1 ns time-delay window is determined by the laser pulse duration and is 45 fs. This represents a three-fold improvement compared to previous approaches where timing jitter was the limiting factor. Two showcase experiments are presented to verify the high time-resolution and sensitivity of the system.

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References and links

1. Introduction

Ultrafast time-domain spectroscopy (TDS) with femtosecond lasers is one of the pivotal techniques to elucidate dynamic processes in the natural sciences occurring on time scales between a few tens of femtoseconds to a few nanoseconds. Examples are studies of charge carrier dynamics [1], energy relaxation and spin dynamics in semiconductors, metals and their nanostructures [2–4], phonon dynamics in solid state materials [5,6], detection of picosecond ultrasound [7,8], or the phase-sensitive detection of THz electric fields in time-domain THz spectroscopy [9–11]. The functional principle of ultrafast TDS systems is to drive a sample into a defined non-equilibrium state using an optical pump pulse and use a time-delayed probe pulse to monitor the sample’s response as function of the time delay between pump and probe pulses. In a conventional experiment, the time delay between pump and probe pulses originating from the same pulsed femtosecond laser is scanned via a variation of the travel distance of one pulse train versus the other. This is mostly accomplished with a retro-reflector mounted onto a linear mechanical scanner. Typical measurement time windows have durations between a few tens of picoseconds and a few nanoseconds and require a retro-reflector motion between several millimeters up to a few tens of centimeters. Scan rates can be a few tens of Hertz for short time-delays (picoseconds) accomplished with vibrating membranes and are well below 1 Hz for long time-delays (nanoseconds) accomplished with stepper motors. The time resolution with such setups is usually given by the laser pulse duration. Other approaches using rotating mirrors have been demonstrated with scan rates of up to 400 Hz and up to 1 ns time delay [12,13]. While these present an important advance with respect to linear stages, mechanical masses rotating at >10,000 rpm on an optical table are a significant noise source. A key disadvantage of all mechanical scanning approaches is that the scan rates are lower than the frequencies of technical noise with significant Fourier content typically up to 1 kHz present on ultrafast TDS lasers. Thus, this noise will inherently be present on the time-domain signals and prevents measurements directly at the shot-noise limit. Another disadvantage of mechanical time-delay scanning is that the change of physical path length causes spot size variations on a sample due to the inherent divergence of the laser beams. Furthermore, slight misalignments of the scanner can lead to position changes of the laser beam on the sample as the time delay is scanned and lead to signal artifacts, in particular if long time-delays are used. Other advanced techniques are based on encoding the pump induced temporal dynamics on a chirped optical probe beam. This technique has been employed for the single-shot detection of THz field transients without using a mechanical...
delay line [14]. The limit of this technique is the accessible time delay given by the length to which a broadband pulse can be temporally stretched.

High-speed asynchronous optical sampling (ASOPS) is an ultrafast TDS method that operates without mechanical scanner, permits multi-kHz scan rates, allows fast measurements directly at the shot-noise limit, and additionally offers the straightforward possibility to perform two-color pump-probe measurements [15–17]. Instead of a mechanical scanner, a high-speed ASOPS system employs two pulsed lasers with a small repetition rate offset \( \Delta f \) to accomplish time-delay scanning without motion of mechanical parts. One laser with repetition rate \( f + \Delta f \) serves as the pump laser, the other with repetition rate \( f \) serves as the probe laser in the experiment. As a result of the offset \( \Delta f \), the time delay \( t \) between pump- and probe-pulse-pairs is linearly ramped between zero and the inverse of \( f + \Delta f \) as a function of real-time \( t \): \( \tau(t) = \frac{(\Delta f \times t)}{(f + \Delta f)} \). This transformation from a time-delay axis \( t \) to a real-time axis \( \tau \) is inherent to high-speed ASOPS. It linearly stretches ultrafast processes by a factor \( (f + \Delta f)/\Delta f \) and makes them accessible to fast data acquisition electronics. The scan rate \( \Delta f \) can be arbitrarily chosen and is only limited by the desired time resolution \( \Delta \tau \). \( \Delta \tau \) is theoretically given by the maximum of the following three quantities: the laser pulse duration; the pulse-pair-to-pulse-pair time-delay increment \( \Delta f/(f \times (f + \Delta f)) \); and the time resolution given by the available bandwidth \( BW \) of the signal detection \( \Delta f/(BW \times (f + \Delta f)) \). Here, we use femtosecond lasers with 45 fs pulse duration and repetition rates of 1 GHz. Available analog-to-digital (A/D) converters with >14 Bit resolution (typically required to measure at shot-noise limit) offer bandwidths of \( BW \approx 100 \text{ MHz} \). Thus, at 1 GHz repetition rate a time resolution below 50 fs can theoretically be achieved at scan rates of up to 5 kHz. At such scan rates, a measurement is completed before typical technical noise has an effect and data acquisition directly at the shot-noise limit is possible. For comparison, the same scheme with more common femtosecond lasers with \( f = 80 \text{ MHz} \) would require \( \Delta f = 320 \text{ Hz} \) to achieve a theoretical resolution of 50 fs and would thus not permit avoiding influence of technical laser noise. This highlights the benefits of repetition rates in the GHz range for ASOPS. In our previous demonstrations of high-speed ASOPS, however, we have not been able to achieve the theoretical time resolution but were limited to 160 fs due to residual timing jitter between the offset-locked lasers that resulted in significant deviations from the ideal time-delay function \( \tau(t) = \frac{(\Delta f \times t)}{(f + \Delta f)} \) [16]. Here, we demonstrate for the first time a repetition rate offset-locking method operating at the tenth harmonics of the repetition rates combined with an offset-frequency definition based on direct-digital-synthesis (DDS) electronics. This method permits a laser-pulse-duration-limited time resolution of 45 fs over the full 1 ns measurement window, i.e. an improvement by a factor of three compared to our previous approach [16]. The high time resolution is verified by resolving optically excited coherent 13.2 THz optical phonons in ZnO. In a more application-oriented example, we use our system for rapid and sensitive mapping of the layer period and total stack thickness of soft-X-ray multilayer mirrors via detection of acoustic phonons and acoustic echoes following optical excitation.

2. High-speed ASOPS setup

The high-speed ASOPS setup is shown in Fig. 1. Core elements are two 1 GHz repetition rate Ti:sapphire lasers sharing a common housing milled from a solid aluminum block (Gigajet TWIN, Gigaoptics GmbH) which are operated in a master-slave configuration. The repetition rate of the master laser is free-running and the slave laser repetition rate is stabilized with an offset \( \Delta f \) using a phase-locked loop (PLL). Both lasers deliver \( \approx 800 \text{ mW} \) of average output power at center wavelengths independently tunable between 750 nm and 850 nm. The pulse durations are \( \approx 45 \text{ fs} \). A small portion of the output power of each laser \( \approx 5 \text{ mW} \) is split off with beam-splitters BS3 and BS4 and focused onto 10-GHz bandwidth photodiodes PD1 and PD2 to detect the repetition rates and their harmonics. The photodiode signals are used to phase-coherently generate an error signal representing deviations of the repetition rate-offset \( \Delta f \) from the desired value \( \Delta f_{\text{set}} \). The electronics to generate this error signal are summarized in the gray box in Fig. 1 and are referred to as the error signal unit (ESU) in the following. The
ESU selects the 10th harmonics of the repetition rates of the master $10 \times f$ and slave lasers $10 \times (f + \Delta f)$ with bandpass filters BP1 and BP2 (10 GHz center frequency, 1 GHz bandwidth) and adds those signals to the outputs of two DDS integrated circuits (ICs) using double-balanced mixers M1 and M2. The DDS elements operate with a 48-bit tuning word, permitting a practically arbitrary choice of the output frequency (tuning step size $\sim 4 \mu$Hz), and with a maximum clock frequency of 1 GHz. The DDS-ICs use the master repetition rate $f$, that has been split off before BP1 using a power splitter (PS), as clock frequency. The output frequency of DDS-IC DDS1 is set to $f/2$ and that of DDS-IC DDS2 is set to $f/2 - (10 \times \Delta f_{\text{set}})$. The outputs of mixers M1 and M2 are bandpass filtered (BP3 and BP4, 10.5 GHz center frequency, 100 MHz bandwidth) to isolate the sum-frequency products. Finally, mixer M3 is used as analog phase-detector to generate an error signal that is proportional to the phase between the outputs of BP3 at $(10 \times f) + f/2$ and BP4 at $10 \times (f + \Delta f) + f/2 - (10 \times \Delta f_{\text{set}})$. The error signal is amplified and low-pass filtered with a loop filter and further amplified by use of a high voltage amplifier (HVA) to control the slave repetition rate via a weight reduced cavity mirror mounted on a high-bandwidth piezoelectric transducer (disc with 2 mm thickness, 10 mm diameter, resonant frequency $>100$ kHz). The highest possible feedback-loop bandwidth is $\sim 12$ kHz, determined by the combined response function of the PLL components. When the PLL is closed, i.e. the error signal is forced to a DC voltage, $\Delta f$ equals $\Delta f_{\text{set}}$ and the lasers are offset-locked in a phase-coherent manner. The ESU contains two major improvements compared to our previous approach [16]. Firstly, using the 10th harmonics of the laser repetition rates allows an enhancement of the feedback loop sensitivity towards timing jitter by a factor of 10 compared to using the first harmonics and thus enables a tighter PLL. Secondly, employing a DDS-IC-based definition of $\Delta f$ instead of using analog single-sideband generators (SSBG) eliminates problems with sidebands separated by $\Delta f$ and higher harmonics of $\Delta f$ from the carrier, as typically created by SSBGs [16]. These sidebands previously caused a significant miscalibration of the time-axis if the feedback loop bandwidth approached $\Delta f$ (typically 2-10 kHz) and thus limited the applicable feedback loop bandwidth to values well below $\Delta f$. In contrast, the system presented here permits to exploit the full available PLL feedback bandwidth of $\sim 12$ kHz, an essential ingredient to achieve a tighter PLL.

Approximately 700 mW of average power from each laser is reflected by BS3 and BS4 and is used for ultrafast time-domain spectroscopy as described previously [15,16].
laser induced transient reflectivity changes of a sample are probed with the probe laser that is
detected with a 125-MHz bandwidth photoreceiver with a Si-pin photodiode as the
photoactive element. The photoreceiver output is digitized with a 100-MS/s 14-Bit A/D
converter. The A/D converter is triggered by a cross-correlation signal between master- and
slave laser generated via two-photon absorption in a GaP diode (PD3) using split off beams
from both lasers (BS1 and BS2) with ≈100 mW average power. It should be pointed out that
the photoreceiver has an AC-coupled signal output with 40 V/mA and a DC monitor output
with 1 V/mA transimpedance gain. This allows to amplify the signal to a level at which the
detection shot-noise exceeds the A/D converter noise (~0.3 mV peak-to-peak) enabling
measurements directly at shot-noise limit and at the same time avoiding saturation of the
transimpedance amplifier and A/D converter with the DC contribution. The low-frequency 3-
dB cutoff of the signal output is at 25 kHz. Thus, if slow signal contributions with Fourier
content below \((f \times 25 \text{ kHz})/\Delta f\) (e.g. 5 GHz in the case of \(\Delta f = 5 \text{ kHz}\) are to be faithfully
resolved (e.g. slow electronic signal decay), \(\Delta f\) must be set to values greater than 25 kHz. The
AC-coupling has no effect on experiments focussing on fast signal dynamics as discussed in
the following.

3. High-speed ASOPS characterization

Key performance parameters of a high-speed ASOPS system are time resolution and residual
error of the time-axis calibration. In contrast to conventional TDS systems, these can depend
on the time delay, i.e. on the position within the measurement window. A characterization is
performed using a cross-correlation setup as shown in Fig. 2(a). The beam from the master
laser is split into two arms and recombined using beam-splitters BS5 and BS6. One arm of the
master laser path has a retro reflector (RR) on a variable delay-stage in order to form a double
pulse with well-defined pulse spacing. The delay-stage with 20 cm travel range has been
calibrated independently with \(\approx 10^{-6}\) relative precision by measuring the pulse-to-pulse
distance of a single laser with well-known repetition rate in an autocorrelation setup. The
pulse train from the slave laser is non-collinearly overlapped with the double pulses from the
master laser in a BBO crystal to create a sum-frequency signal that is detected with an
amplified photodiode (PD). Figure 2(b) shows a typical transient, 4000-times averaged, for a
fixed translation stage position. The signal at 0-ps, labelled “trigger”, corresponds to the
master pulse train transmitting both beam-splitters BS5 and BS6. This signal is used to trigger
the data acquisition. The position of the second peak, labelled “cross-correlation”, changes
with the retro reflector position and defines the position within the 1 ns measurement window
at which the characterization is performed. The full width at half maximum (FWHM) of the
averaged “cross-correlation” signal divided by a deconvolution factor 1.54 for the squared-
hyperbolic secant pulses from the lasers is a measure of the time resolution of the high-speed
ASOPS system at a given time-delay position. Figure 2(c) shows the time resolution evaluated
at 150 equidistant positions within the 1 ns window for \(\Delta f = 2 \text{ kHz}, 3 \text{ kHz} \) and 5 kHz. A time
resolution of 50 fs is obtained for \(\Delta f = 5 \text{ kHz}\) throughout the full measurement window. Here,
Fig. 2. (a) Cross-correlation based characterization setup of the high-speed ASOPS system. BS5 and BS6: optical beam-splitters, RR: retro reflector, F: focusing lens, P: slit to filter out second harmonics, PD: amplified photodiode. (b) Exemplary detected transient of the cross-correlation based characterization. Inset: zoom into cross-correlation peak. (c) & (d) Extracted time resolution- and residual error of time-axis calibration data from a measurement series for $\Delta f = 5$ kHz, $3$ kHz and $2$ kHz.

the time resolution limitation is given by the available signal detection bandwidth $BW$ to $\frac{\Delta f}{BW \times (f + \Delta f)} = 50$ fs. For $\Delta f = 3$ kHz and $\Delta f = 2$ kHz the time resolution reaches the pulse duration limit of 45 fs. This shows that timing jitter is not the limiting factor of the time resolution as has been the case in previous experiments [16]. The slight increase of the time resolution by $\approx 4$ fs at time delays $<100$ ps is currently not understood but has negligible effect on the system performance. In addition to the time resolution, it is also important to verify the time-axis calibration $\tau(t)$ as a function of position in the measurement window. Systematic deviations from the ideal linear behaviour $\tau(t) = \frac{(\Delta f \times t)}{(f + \Delta f)}$ may be imposed by the PLL electronics. For instance, sidebands on the SSBG output previously used [16] are modulated into the error signal and can cause errors of the time-axis calibration by several hundreds of femtoseconds if too high feedback bandwidth is employed. In such a case, the recorded signals will be significantly distorted. The residual calibration-error of the time-axis is obtained by comparing the time delay of the cross-correlation peak measured with the ASOPS system to the master laser time delay preset with the retro reflector on the calibrated translation stage. As shown in Fig. 2(d), the residual calibration-error of the time-axis is below 12 fs for all values of $\Delta f$ and is thus insignificant compared to the time resolution. A system characterization at $\Delta f = 1$ kHz yields the same performance as obtained with $\Delta f = 2$ kHz but can only be performed for the first 500 ps of the measurement window due to the limited memory of the A/D converter.
4. High-speed ASOPS experiments

To test the time resolution of the high-speed ASOPS system, optically excited coherent \( E_{2}^{\text{low}} \) phonons at 3 THz and \( E_{2}^{\text{high}} \) phonons at 13.2 THz in ZnO are measured in reflection geometry at room temperature \([18,19]\). The ZnO sample was hydrothermally grown in a wurtzite structure with a (0001) orientation and a thickness of 0.33 mm. The pump- and probe powers are set to 680 mW and 160 mW, respectively, and both lasers have a center-wavelength of 825 nm. The offset frequency is set to \( \Delta f = 1 \text{ kHz} \) to obtain one data point per 10 fs, which is sufficient to clearly resolve the 76 fs oscillation period of the 13.2 THz optical phonons. In ZnO, excitation and detection of the optical phonon modes below the optical band gap occurs via impulsive stimulated Raman scattering \([19]\). Thus the reflected probe light is spectrally modulated at the coherent phonon frequencies. To obtain a non-zero signal, the detection was performed using the short wavelength edge of the probe light spectrum isolated by means of an 815 nm short-wavelength-pass filter and an amplified photodiode \([20,21]\). Figure 3(a) shows the first 2.5 ps of the transient after subtraction of a slow exponential decay caused by optically excited electrons. Oscillations with a period of 337 fs are observable which are superimposed by faster oscillations with a 76 fs period. Figure 3(b) shows the fast Fourier transform (FFT) of the full transient (500 ps length for \( \Delta f = 1 \text{ kHz} \), due to the limited memory of the A/D converter). A large peak at 2.97 THz, corresponding to low-frequency optical phonons \( E_{2}^{\text{low}} \), and a small peak at 13.15 THz, corresponding to high-frequency optical phonons \( E_{2}^{\text{high}} \), are observed \([19]\). The inset in Fig. 3(b) shows a magnified view of the FFT and displays a small peak at 10.02 THz which has to our knowledge not yet been reported in TDS measurements. This peak has previously been observed by Raman spectroscopy \([18,22]\) and is assigned to the second order Raman scattering process \( E_{2}^{\text{high}}-E_{2}^{\text{low}} \) \([22]\). The visibility of this peak, which represents the combined density-of-states of the involved phonons, in a TDS experiment is a good demonstration of the high sensitivity of high-speed ASOPS. To verify the 45-fs time-resolution of the system at late delays after the trigger signal, we modified the optical trigger beam path such that the signal appears around 900 ps time-delay and find no changes to the signal.

![Fig. 3. High-speed ASOPS measured data of wurtzite ZnO. a) Zoom into first 2.5 ps of the recorded time domain transient revealing optical phonon oscillations. b) Fast Fourier transform of the ZnO transient showing peaks at 2.97 THz, 10.02 THz and 13.15 THz. Inset: Amplitude zoom into the 10.02 THz and 13.15 THz peak by a factor of 100.](image-url)

In a second example, we focus on non-destructive inspection of a multilayer structure via laser induced picosecond ultrasound. We investigate Si/Mo superlattices sputter-deposited on a mono-crystalline Si wafer, which are promising candidates for future soft-X-ray or extreme ultraviolet mirrors \((\lambda=13 \text{ nm})\) \([23]\). Our sample is composed of 60 periods, each nominally consisting of 4.1 nm Si and 2.7 nm Mo \([24]\). The nominal period and total stack thickness are thus 6.8 nm and 408 nm, respectively. The Si- and Mo layers are amorphous \([24]\). The high-
speed ASOPS TDS system is employed for rapid measurements of the mirror’s acoustic response to optical excitation. Mapping of the acoustic signal permits extraction of information about the spatial distribution and homogeneity of the structure period and variations of the total stack thickness, which is essential information required to optimize the mirror performance. Measurements are performed in reflection geometry [16]. The pump- and probe powers are set to 200 mW and 20 mW, respectively. Both lasers are operated at a center wavelength of 825 nm and \( \Delta f \) is set to 5 kHz. Figure 4 shows transient reflectivity changes of the sample after optical excitation at a fixed position for different averaging times. The optical pump pulse excites hot electrons at the top of the superlattice which lead to ultrafast heating and expansion of the crystal lattice via electron-phonon-scattering. This causes the initial signal peak and launches a picosecond ultrasound pulse into the structure [23,25]. The exponential signal decay due to carrier relaxation and lattice cooling on a 10 ps time scale is superimposed by a coherent acoustic phonon signature at frequency \( f_{ph} \approx 1 \) THz that is detected in the transient reflectivity of the mirror surface via the elasto-optic effect [5,23,26–28]. The analysis of the Fourier spectra of these coherent phonons reveal a combination of zone-folded acoustic modes of the superlattice and a localized surface mode [23,27,28]. These oscillations are visible at short time delays (\( \approx 25 \) ps, see Fig. 4 “Zoom 1”) and are correlated to the period \( d_{SL} = v_l f_{ph} \) of the superlattice if the phonon dispersion is approximated linearly. \( v_l \) is the longitudinal sound velocity in the superlattice averaged over the constituting layers. In addition, the optically generated ultrasound pulse propagates through the superlattice, is reflected at the interface to the Si-substrate and propagates back to the surface causing an acoustic echo in the transient reflectivity at a time delay of \( \tau_{refl} \approx 132 \) ps (see “Zoom 2” of Fig. 4). The position of the echo maximum \( \tau_{refl} \) is correlated to the superlattice total stack thickness via \( d = (v_l \times \tau_{refl})/2 \). Thus, in principle, the Si/Mo-period \( d_{SL} \) and the multilayer total stack thickness \( d \) can be derived from \( f_{ph} \) and \( \tau_{refl} \), respectively. However, the sound velocities of the amorphous constituting thin films and thus the average sound velocity in the superlattice are largely unknown at present. Thus, we use the echo position \( \tau_{refl} = 132 \) ps measured at sufficient distance from the wafer edge (8 mm) and the nominal stack thickness

Fig. 4. Transient reflectivity changes of a Si/Mo-multilayer superlattice following optical excitation for different averaging times 0.2 s (10^3 averages), 2 s (10^4 averages) and 20 s (10^5 averages). “Zoom 1” shows \( \approx 1 \) THz phonon oscillations and “Zoom 2” shows an acoustic echo caused by the mirror/substrate interface.
to determine $v_l$ to 6180 m/s and use this value in the following. The error of this value for $v_l$ is below 5% and stems from the uncertainty in the determination of the exact position of $\tau_{\text{re}}$ and from the maximum possible error in the actual layer period and total stack thickness that would still yield the experimentally confirmed reflection band at the design wavelength of the soft-X-ray mirror at 13.4 nm [24]. Due to the requirement to extract $v_l$ from the measurement, our data do not yield absolute values for $d\text{SL}$ and $d$ without additional structural information from X-ray analysis. Yet, the technique is able to estimate variations of the superlattice period and total stack thickness when mapping a wafer and is thus well suited to analyze the structure homogeneity. Figure 5 shows the extracted superlattice period $d\text{SL}$ and total stack thickness $d$ over a 10 mm long radial line-scan from the Si/Mo-multilayer wafer edge towards the wafer center for different acquisition times per data point. Data were acquired at 200 equidistant positions separated by 50 µm with optical spot sizes of $\approx 30$ µm (FWHM). Transients with 0.2 s acquisition time were low-pass filtered before evaluation of $d\text{SL}$ and $d$ to eliminate high frequency noise. Owing to the 5 kHz scan rate and our measurement capability directly at the shot-noise limit, the line scan with 0.2 s acquisition time per data point (i.e. 5 Hz pixel acquisition frequency, 48 s per line-scan) readily permits clear identification of a reduction of the mirror period and total stack thickness when approaching the wafer edge. This effect is expected and is caused by the inhomogeneity of the sputtering process. Increasing the averaging time by factors of 10 and 100 reduces the statistical error but essentially yields the same qualitative result as a scan at 5 Hz pixel acquisition frequency. More than 6 mm away from the wafer edge the extracted mean multilayer period $<d\text{SL}>$ is 6.2 nm, with a standard deviation of 43 pm, 16 pm, and 8 pm for the data with 0.2 s, 2 s, and 20 s per pixel, respectively. The deviation of this value from the nominal 6.8 nm period can be explained by the fact that the linear approximation of the phonon dispersion used to extract $d\text{SL}$ ignores phonon bandgaps at the Brillouin-zone center and possible excitation of localized surface modes in the bandgaps [27,28]. The mean total stack thickness $<d>$ is 408 nm, with a standard deviation of 3.1 nm, 1.8 nm, and 0.8 nm for the data with 0.2 s, 2 s, and 20 s per pixel, respectively. This value for $<d>$ is expected since the nominal total stack thickness has been used to determine $v_l$. The presented data suggest that our technique is capable of detecting superlattice period variations on the order of 0.1 nm and total stack thickness variations on the

Fig. 5. Si/Mo-multilayer superlattice period (plotted versus left axis) and total stack thickness (plotted versus right axis) versus the radial distance from the wafer edge for different total line-scan acquisition times. The values of the total stack thickness are plotted upside-down to distinguish the curves from the period plots. The color-code (red, green, black) of these curves corresponds to the transients of Fig. 4 used for the evaluation at a fixed position.
order of 1 nm with a measurement time on the order of just 1 s per pixel. While we are currently limited to thickness variations, an exact determination of the sound velocities should be straightforward with an analysis of superlattices of different relative Si/Mo composition and comparing those data to absolute thickness measurements performed, e.g. with high resolution X-ray analysis. In this case, our technique would also be capable of yielding absolute thickness values. This example highlights the advantages of high-speed ASOPS for laser induced picosecond ultrasound applications, e.g. non-destructive wafer inspection at the nanometer thickness level. Fast oscillations at THz frequencies and small acoustic signatures ($\Delta R/R \approx 10^{-5}$) at long time delays of hundreds of picoseconds can simultaneously be resolved with a single measurement lasting only between a few hundreds of milliseconds to a few seconds, depending on the desired signal-to-noise ratio. This is particularly useful for rapid mapping of functional structure properties because imaging of significant areas can be performed in a few minutes.

5. Summary and conclusion

We have demonstrated a high-speed ASOPS TDS system with 45 fs time resolution over a 1 ns time-delay window based on two 1-GHz repetition rate femtosecond Ti:sapphire lasers. The three-fold improvement of the time resolution compared to our previous approach [16] is enabled by an improved feedback loop employing the 10th harmonics of the laser repetition rates and DDS-ICs to define the repetition rate-offset. The residual error of the time-axis calibration is below 12 fs. We have demonstrated the excellent time resolution by resolving optically excited 13.15 THz coherent optical $E_2^{\text{high}}$ phonons in ZnO. Due to the high signal-to-noise ratio, excitation of a coherent signature $E_2^{\text{high}} - E_2^{\text{low}}$ via second order Raman scattering has been resolved for the first time with a time domain spectrometer. In a second example, layer period and total stack thickness variations of soft X-ray Si/Mo-multilayer mirrors have been mapped via laser induced picosecond ultrasound. The resolution is on the order of 0.1 nm and 1 nm, respectively, with an acquisition time per pixel of $\approx 1$ s. We conclude that the presented high-speed ASOPS spectroscopy system matches the time resolution of most conventional time-domain spectroscopy setups. Additionally it offers uniquely high scan rates in the kilohertz range and signal detection directly at the shot-noise limit. It eliminates mechanical delay stages and provides the possibility to perform two-color pump-probe measurements. We believe that the presented high-speed ASOPS system thus has the potential to fully replace conventional time-domain spectrometers.

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