Application Note

Zurich Instruments

Compressed Sensing in Field-Resolved Spectroscopy

Applications: Optics, Pump-Probe Spectroscopy

Products: UHFLI, UHF-BOX

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Introduction

The transient response of a sample to an external field provides detailed information about its properties. The time-dependent electric field of light can be measured by various spectroscopy techniques such as pump-probe spectroscopy, Fourier transform spectroscopy, dual-comb spectroscopy, and field-resolved spectroscopy. However, these techniques have limited measurement speeds due to the constraints of the required sampling points dictated by the Nyquist criterion. This application note describes the implementation of compressed sensing in field-resolved spectroscopy allowing to speed up measurement times by decreasing the number of required sampling points beyond the Nyquist criterion and then reconstructing the response with a dedicated algorithm. As this method requires measuring uncorrelated information from individual signal pulses, the Zurich Instruments 600 MHz UHFLI Lock-in Amplifier with the UHF-BOX Boxcar Averager Option is employed.

In field-resolved spectroscopy, the time-dependent electric field of light is measured after its interaction with matter. During this interaction, the molecular information of the matter's constituents becomes encoded onto the electric field of light, exhibiting a response that can last from hundreds of femtoseconds up to several tens of nanoseconds. When using a short excitation pulse, the molecular response is temporally separated from the excitation pulse. Temporal gating of the response with subsequent Fourier transformation allows for complex molecular fingerprinting with new orders of sensitivity and dynamic range. The electric field can be measured via electro-optic sampling in which a short probe pulse is spatially and temporally overlapped with the electric field in a nonlinear crystal. Upon nonlinear interaction inside the crystal, the probe pulse obtains information about the electric field strength at the temporal overlap. By subsequently changing the relative temporal delay between the probe pulse and the beam containing the molecular information, the entire waveform of the molecular response can be acquired.

In conventional field-resolved spectroscopy techniques, the measurement speed is limited by the number of sampling points which is required to fulfill the Nyquist criterion ($f_{sampling} \geq 2 \cdot f_{signal}$). Compressed sensing (or sparse sampling) is able to break the Nyquist barrier by determining the number of necessary measurement points according to the amount of information contained in a signal rather than its maximal signal frequency. Consequently, for signals possessing a sparse domain in which only a few signal amplitudes are non-zero, the original signal can be reconstructed with a reduced number of sampling points. In spectroscopy, the amount of sampling points can be reduced, since most samples exhibit only a few discrete spectral absorption lines. For improved signal reconstruction, the signal needs to be sampled randomly with randomly varying step sizes between sample points. Since compressed sensing does not have a clear boundary for the required number of sampling points, they need to be acquired uniformly across the entire signal length. Consequently, the convergence of subsequent reconstructions allows for an exit condition enabling real-time compressed sensing measurements.

Experimental Methods

A schematic of the experimental setup is depicted in Figure 1. A 1 kHz pulse train containing 54 fs laser pulses centered at 810 nm is split into two paths via a beam splitter: the reflected beam is used to generate the THz pulses for the sample excitation, while the transmitted beam is used to probe the effect. Before generating

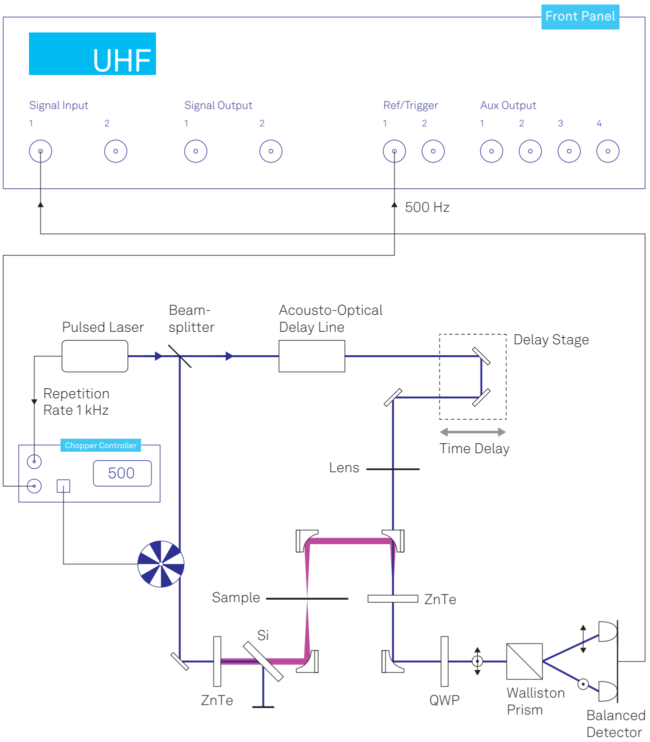

Figure 1. Simplified schematic of the experimental setup, adapted from Ref. [1]. A pulsed laser with a repetition rate of 1 kHz is split into a pump beam and a probe beam. The pump beam is modulated by a chopper before generating THz pulses which are then used to excite the sample. The probe beam passes through an acousto-optical delay line to introduce rapid changes of the temporal overlap between pump and probe.

THz pulses via optical rectification in a ZnTe crystal, the reflected beam is chopped at 500 Hz, meaning that the THz pump pulse is present only for every second probe pulse. After the generation crystal, a silicon plate filters out the pump beam. The THz beam then interacts with the sample before being focused into a thin ZnTe detection crystal.

The probe pulses pass an acousto-optical delay line and a mechanical delay line to be able to scan the entire waveform of the THz electric field. The slow mechanical delay line extends the capable scanning range while the acousto-optical delay line allows for shot-to-shot changes of the temporal overlap between the THz waveform and the probe pulse every time the THz field is sampled. The probe pulse is then focused through a pierced parabola onto the detection crystal spatially overlapped with the THz beam. The polarization of the probe pulses changes hereby depending on the instantaneous electric field strength of the THz beam (E_{THz}) at the temporal overlap. This polarization change is detected via an ellipsometer consisting of a quarter wave plate, a Wollaston prism, and a balanced photodetector. The resulting voltage of the balanced photodetector is directly proportional to the electric field of the THz beam at the temporal overlap of the probe pulse $V \propto E_{THz}$. When no THz field is present, i.e. for every second probe pulse, the polarisation of the probe pulse is not changed in the detection crystal and the balanced detector yields 0 V.

In a real-time compressed sensing experiment, the rapid changes of the time delay induced by the acousto-optical delay line leads to rapid changes in the signal amplitude and there is no relation between the information of two consecutive pulse pairs. To capture such a rapid change in the signal, it is crucial that every pair of pulses is measured individually, without any memory from the previous pulse pair. This can be achieved using the UHF-BOX Boxcar Averager, where the voltage signal from the photodiode is filtered in the time domain by selecting a defined measurement window within the period of the repetition rate. The repetition rate of the boxcar averager is set to 500 Hz, such that every period contains one probe pulse with and one without interaction with the THz pulse. Figure 2(a) shows the measurement of the probe pulses for one boxcar period using the Periodic Waveform Analyzer (PWA) of the UHFLI-BOX. The amplitude of the unpumped pulse is used as a reference and thereby allows to correct for drifts and systematic noise. By choosing no averaging, the measurement result from every pair of pulses can be saved individually.

Experimental Results

Compressed sensing can be used for terahertz field-resolved spectroscopy of many types of samples. Here, we review the results from measuring water vapor in ambient air, that are published in Ref. [1]. As a reference, the experiment was first performed using a conventional measurement where the time delay was adjusted with the delay stage, and the signal amplitude was measured for each delay step. Figure 2(b) shows the electric field response of a THz pulse for two different samples: ambient air with a relative humidity of 50 % and a nitrogen-purged environment with less than 5 % humidity. In the first case, the molecular response of water vapor is imprinted onto the electric field trailing the main excitation pulse. The latter case shows solely the excitation pulse without any visible molecular response. The molecular response in the window between 7 ps and 35 ps is used for a comparison between a conventional sampled experiment with an undersampled compressed sensing experiment.

Figure 2(c) shows a blue curve that was obtained by performing a standard lock-in measurement at small steps of the delay line while the black sample points were obtained with the random sampling technique. The good agreement confirms that the detection scheme can follow the rapid amplitude changes when jumping between random electric field amplitudes. The red curve shows the reconstructed response from the black data points. The randomly acquired sample points are fed into a basis-pursuit denoising (BPD) compressed sensing algorithm to reconstruct the original waveform [2, 3]. The reconstructed waveform is shown in red.

Figure 2(d) shows the corresponding spectrum of the reconstruction. Furthermore, the spectrum of a normal undersampled waveform (green) with the same number of sampling points is illustrated where the Nyquist

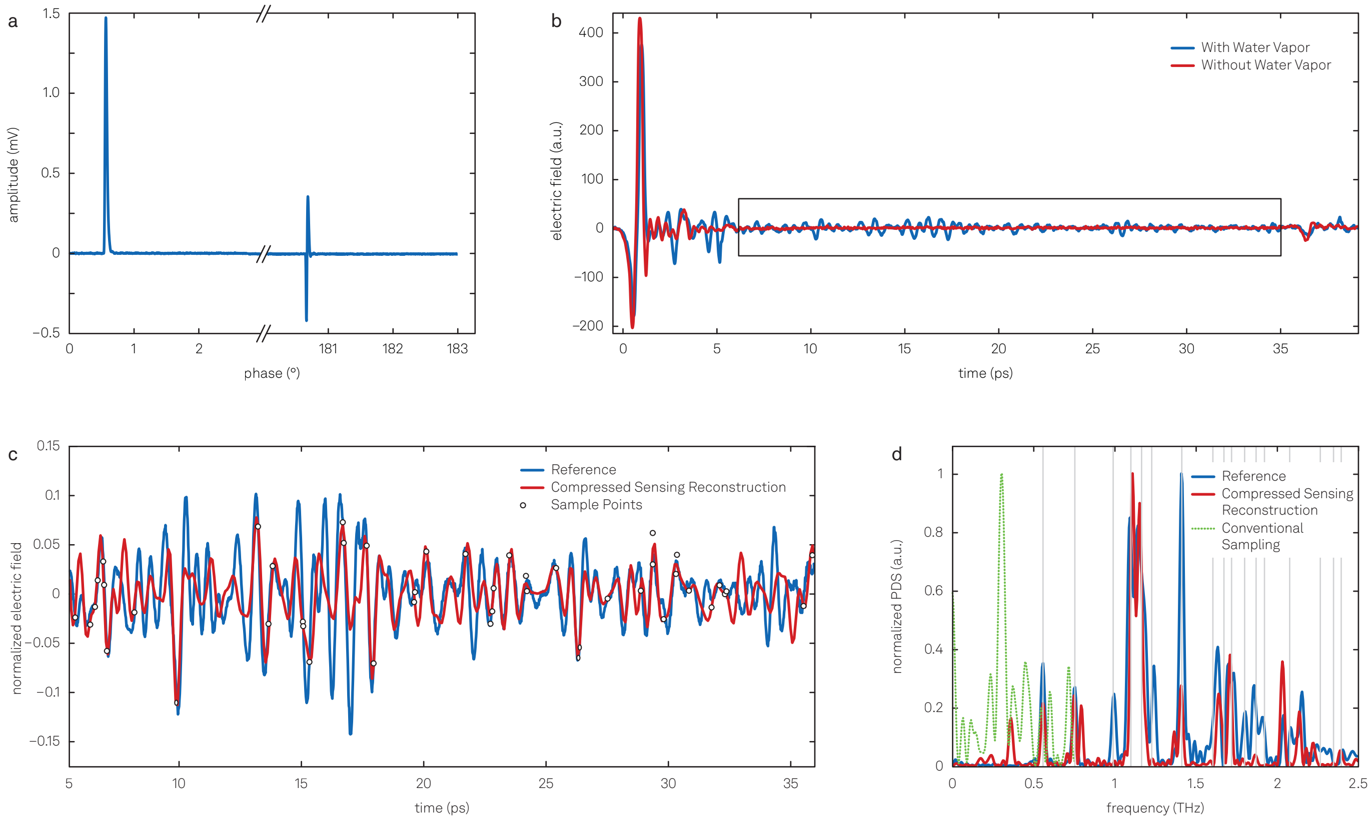

Figure 2. Experimental results, adapted from Ref. [1] (a) Signal amplitude as a function of phase recorded by the Periodic Waveform Analyzer of the UHFLI. The probe pulse with and without THz field interaction can be observed at the phase of 0.5° and 180.5 °, respectively. (b) Measurement of the electric field response of a THz pulse measured by sweeping the time delay with the delay line. The measurement was performed on two samples with and without water vapor. The signatures of the water vapor can be identified for delay times between 7 ps and 35 ps. (c) Time-domain comparison of the measured electrical field response of a conventional measurement (blue line) and the data points measured via rapid random scanning using the acousto-optical delay line (black circles). (d) Frequency-domain comparison of the conventional measurement (blue) and the compressed sensing method (red). Additionally, the green curve shows the spectrum which would be obtained by a conventional FFT using the compressed sensing data points.

criterion limits the spectrum of the normal undersampled waveform to 0.7 THz.

To evaluate the information from the spectra, the absorption frequencies of water are indicated in Figure 2(d) with gray vertical lines. It can be observed that the spectrum of the compressed sensing reconstruction shows peaks at the water absorptions up to a frequency of 2.2 THz while the spectrum from the normal undersampled waveform does not capture the information of the absorption lines and even shows some artifacts at low wavelengths due to aliasing effects.

Conclusion

In this application note, we have presented an experimental setup for field-resolved spectroscopy using compressed sensing. By using rapid random scanning of the delay capturing the signal with the UHF-BOX Boxcar Averager, it is possible to reconstruct the water absorption lines of a test sample based on 33% percent of the sampling points that would be required with a standard sampling technique.

Acknowledgments

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References

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