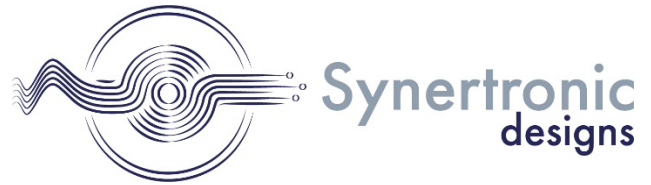


APN-006

Optical pump - THz probe (OPTP) spectroscopy



Introduction

Optical pump–terahertz probe (OPTP) spectroscopy is a time-resolved technique used to investigate ultrafast carrier dynamics and transient photoconductivity in materials following optical excitation, which provides a highly time-resolved and repeatable trigger for molecular processes. In this method, an ultrafast optical pulse (the pump) excites the sample, creating nonequilibrium states such as photoexcited carriers or excitons. A delayed terahertz (THz) pulse (the probe) then interrogates the sample, allowing for measurement of its transient conductivity, and dielectric response as well as detecting transient states with sub-picosecond resolution. The delay is mediated by a mechanical delay line that adjusts the optical path length for either pulse, and a series of adjustments in relative pathlength allow the observation of dynamic processes. Due to the spectral content of a THz pulse, it shows sensitivity to low-energy excitations, charge mobility, and free carrier absorption. OPTP is particularly valuable for studying semiconductors, nanomaterials, perovskites, and correlated electron systems. It provides critical insights into carrier lifetimes, recombination dynamics, and scattering processes, making it a powerful tool in both fundamental research and materials development for photovoltaics, optoelectronics, and quantum materials.

OPTP is analogous to Transient Absorption Spectroscopy (TAS) in that it monitors changes in the absorption strength of a sample volume over time following excitation by a short light pulse, but the absorption changes are in the THz-domain. Ultrafast THz pulses are typically detected indirectly, by tracking the optical birefringence induced in a suitable medium by the THz- electric field, so the need for **high-repetition rate detection with low noise and high dynamic range** in the optical frequency range remains.

High-repetition rate CMOS-based LineScan cameras offer unique capabilities that can significantly enhance OPTP measurements, particularly in terms of **throughput, sensitivity, data quality, and long-term stability**.

Role of Spectral Detection in THz-domain spectroscopy

As mentioned in the introduction, the THz frequency range (typically 0.1 – 10 THz) is ideal for probing low-energy excitations in materials. The types of resonances commonly detected, consider charge, lattice and spin properties of materials and may be used to detect phonons, polaritons, magnons etc. A widely deployed method is Time-Domain THz Spectroscopy (TDS) which measures the time evolution of a THz-regime electro-magnetic signature. This is enabled by Electro-optic sampling (EOS) which works by exploiting the electro-optic effect (Pockels effect). Specifically, the refractive index of certain nonlinear crystals (like ZnTe or GaP) changes in response to an external electric field provided in this case by an intense THz pulse.

A rough outline of the methodology is outlined below in a step-by-step fashion:

1. **Generation of THz Pulse:** A femtosecond laser pulse is split into two parts—one generates the THz pulse (e.g., via a photoconductive antenna or nonlinear crystal).
2. **Probe Pulse:** The second part of the femtosecond laser pulse (called the probe) is delayed in time and directed through the same volume of electro-optic crystal that is subjected to the THz-beam.
3. **Sampling:** When the THz pulse and the time-delayed optical probe pulse overlap in the crystal, the transient THz electric field induces birefringences which alter the polarization state of the optical probe pulse in proportion to the instantaneous THz field strength.
4. **Detection:** A polarization-sensitive optical detection setup (typically using a quarter-wave plate, polarizer, and balanced photodiodes) measures this change, sampling the THz field at a single moment in time defined by the ultrafast probe pulse duration.
5. **Time Scan:** By scanning the time delay between the THz pulse and the probe pulse, the full time-domain waveform of the THz electric field can be reconstructed with sub-picosecond resolution.

EOS therefore enables precise, time-domain measurements of the electric field evolution of a THz pulses by using an optical ultrafast laser probe. TDS then involves Fourier transforming the detected THz waveform to access amplitude and phase information across a broad frequency range. The THz absorption of a sample can therefore be studied using TDS, by allowing the THz pulse to pass through the sample and comparing the resulting waveform to a reference measurement. This frequency-resolved approach allows researchers to disentangle different physical processes—such as free carrier absorption, phonon resonances, and excitonic features—based on their distinct spectral signatures.

Of course, this would describe the ground state THz absorption and does not yet allow for the study of the evolution of THz resonances. But this can be enabled by incorporating EOS into another pump-probe scheme much like conventional transient absorption spectroscopy, where the sample is first excited by a laser pulse and transient non-equilibrium states can be interrogated.

However, combining two delay stages, one for scanning the THz waveform and another for varying the time delay between sample excitation and THz probe, implies rather imposing data acquisition times. If the desired experiment requires the study of an additional parameter, a varying external magnetic field is typical for this field, the total acquisition time may become unfeasible.

A workaround is to replace the photodiodes typically deployed in EOS with an array of photodetectors (such as a LineScan camera) and deploying a sophisticated scheme that overlaps different parts of the optical probe beam with different parts of the THz wavefield, which is described in section 4. As with TAS, efficient and accurate detection will require:

- **High temporal resolution** to resolve fast processes (femtoseconds to nanoseconds)
- **Fast frame rates** for averaging and capturing low-signal events and matching laser repetition rates
- **High signal-to-noise ratio (SNR)** to detect subtle changes
- **High dynamic range** to accommodate both weak and strong signals.

Application of CMOS-Based Linescan Cameras in OPTP

High Repetition Rate and Frame Rate

Modern CMOS LineScan cameras can operate at tens to hundreds of kHz line rates, allowing rapid acquisition of data with minimal dead time. This is well-matched with high-repetition-rate pump-probe laser systems, enabling high-throughput data collection.

Single shot Monitoring

The high speed of CMOS LineScan cameras allows capture of **every laser pulse** or **alternating pulse sequences**, enabling **shot-to-shot noise rejection**, **excitation correction**, **reference probe spectral characterization** and **real-time signal tracking**, which is critical for experiments with low signal levels or fluctuating laser sources.

Compact Form Factor and Integration

CMOS sensors are compact and easier to integrate into experimental setups. Their low power consumption and digital output interfaces simplify data handling and processing.

Multi-Region of Interest (ROI) and High Dynamic Range

Advanced CMOS cameras allow programmable ROIs and exposure control, enabling dynamic adjustment based on signal strength on a per pixel basis. Combined with on-chip digitization, this results in improved **dynamic range and SNR**.

Application in OPTP Workflows

Signal Detection

A basic description of an effective OPTP experimental setup requires 3 beampaths: an optical pump to populate transient states of the sample, a THz pulse to detect absorption of low frequency resonances and an optical gate pulse that is split into orthogonal polarization states and detects the THz waveform by EOS as described in section 2. In contrast to conventional EOS, the photodiodes can be replaced by a CMOS LineScan camera to enable single-shot detection of the THz waveform. This is achieved by dispersing the optical gate pulse by a stair-step echelon that separates the optical pulse into a sequence of beamlets with fixed temporal delays between them. The array of beamlets are then focused together with the THz pulse, after the latter has passed through the sample, into the birefringent EOS crystal. The beamlets are split into orthogonal polarization states and imaged as a line using cylindrical lenses onto one of two LineScan cameras that monitor the transmitted intensity of orthogonal polarization states. Separate beamlets represent separate time windows and the THz waveform effect on the birefringent crystal is encoded onto the relative polarization state of each beamlet, effectively allowing for detection of the THz waveform with a single pulse. The dispersed optical probe beam is thereby imaged onto the CMOS LineScan sensor, with each pixel calibrated to a corresponding time delay. High line rates ensure rapid acquisition, ideal for resolving transient absorption features in the THz spectral domain.

Change Monitoring

By synchronizing the camera to the laser system, one can continuously monitor changes in transmission through the sample as a function of time after photo-excitation and, if required, due to a varying external magnetic field.

Observation of Transient States

Due to its high-speed capability, the camera can capture transient states that exist for only nanoseconds to femtoseconds. This is particularly important for:

- Charge carrier dynamics and recombination in optoelectronics
- Plasmons in structured materials like graphene
- Cyclotron resonances in semiconductors

Synchronization and Data Acquisition

The camera's clock can be externally triggered by the laser's control system, a programmable delay generator or a light detector such as a photo-diode. Typical acquisition schemes include:

- **Pump-on / pump-off cycles** for differential measurements, both for optical pump pulses and THz pulses, enabled by a 4-shot measurement cycle
- **Real-time averaging** across thousands of pulses or measurement cycles
- **Single-shot mode** for unstable or single-event measurements

Advanced software frameworks can be used to control the data pipeline, applying real-time corrections such as dark subtraction, reference measurements, and ΔA computation.

Conclusion

High-repetition rate CMOS-based LineScan cameras offer a powerful, flexible, and cost-effective solution for sensitive spectral detection and change monitoring in the context of time-resolved THz spectroscopy. Their high speed enables high temporal resolution and integration capabilities make them well-suited for modern digital OPTP setups, enabling detailed insights into ultrafast phenomena with high fidelity and throughput.