DELIVERABLE REPORT

WP9 – JRA4 - Research on Time-resolved ultrafast probes on nanosystems

D9.1
Pump-probe setup with tunable pump beam

31-08-2017

M24

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DELIVERABLE DETAILS

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Pump-probe setup with tunable pump beam open to users

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DISSEMINATION LEVEL

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☐ PP - Restricted to other programme participants & EC: (Specify)
☐ RE - Restricted to a group (Specify)
☐ CO - Confidential, only for members of the consortium

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1. Executive Summary

To understand the physics of nanomaterials and nanosystems is cornerstone for contemporary Science and Technology. To this end, the macroscopic properties of materials can be traced down to their structural and electronic properties at the nanoscopic level. Knowledge of the electronic and lattice dynamics sets the basis for efficient modelling of the material in time and highlights properties useful for promoting technological applications. The time-resolved differential pump probe technique, with use of ultrafast laser sources, is ideal for such studies. By monitoring the differential transmissivity / reflectivity of nanosystems we provide insight into their ultrafast optical, optoelectronic and structural-phononic response. Deliverable 9.1 refers to building a pump-probe workstations for VIS near-VIS, near-IR and THz spectroscopy of nanosystems that will be readily available to NFFA-Europe users.

2. Pump probe workstations

2.1 Pump-probe visible and nIR setups

The first objective of this deliverable is the building a pump-probe workstations for VIS near-VIS, near-IR with tunable pump beam line and white light supercontinuum probe line (sub-ps resolution).

The basic laser system used is a Ti:Sapph laser oscillator producing broadband laser pulses with a band width of > 100 nm centered at ~ 785 nm. The minimum pulse duration is around 10 fs. This is streched and is passed through a multipass amplifier. There, the pulse energy is amplified to ~ 1 mJ per pulse. The amplified pulses are then subjected to temporal compression and the final amplified laser pulses have band width of around 40 nm and can be temporally compressed to a minimum pulse duration of < 30 fs. Temporal characterization of the output laser pulses is performed by means of a Michelson-type interferometer which is specifically designed and set up for this purpose and is shown in *Figure 1*.

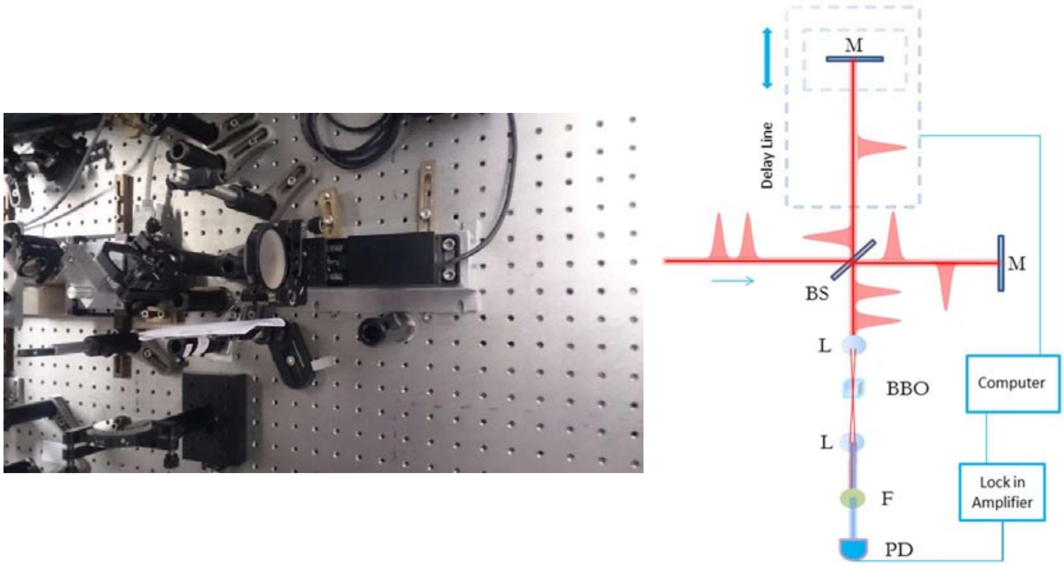

Figure 1: Left side: Picture showing the Michelson interferometer in the laboratory. Right side: schematic Michelson-type interferometer. BS: Beam Splitter, M: mirror, L: lens, F: low-pass filter, PD: photodiode.

It consists of two mirrors at an equal distance from a beam splitter. One of the mirrors is mounted on a motorized stage which acts as a delay line. The incoming beam is split and recombined at the beam splitter after being backreflected by the mirrors. The recombined beam is focused on a BBO crystal for Second Harmonic Generation (SHG). Using the delay line to change the optical path of one arm of the interferometer the relative delay time between pulses can be set. By recording the intensity of SHG versus time the duration of the pulse can be estimated using the interferometric autocorrelation function. The laser output is typically attenuated and guided into the entrance of the Michelson interferometer. The temporal autocorrelations of a fully compressed (and thus of minimum pulse duration) pulses vs a not optimally compressed (and thus chirped laser pulse) are shown for comparison in *Figure 2.*

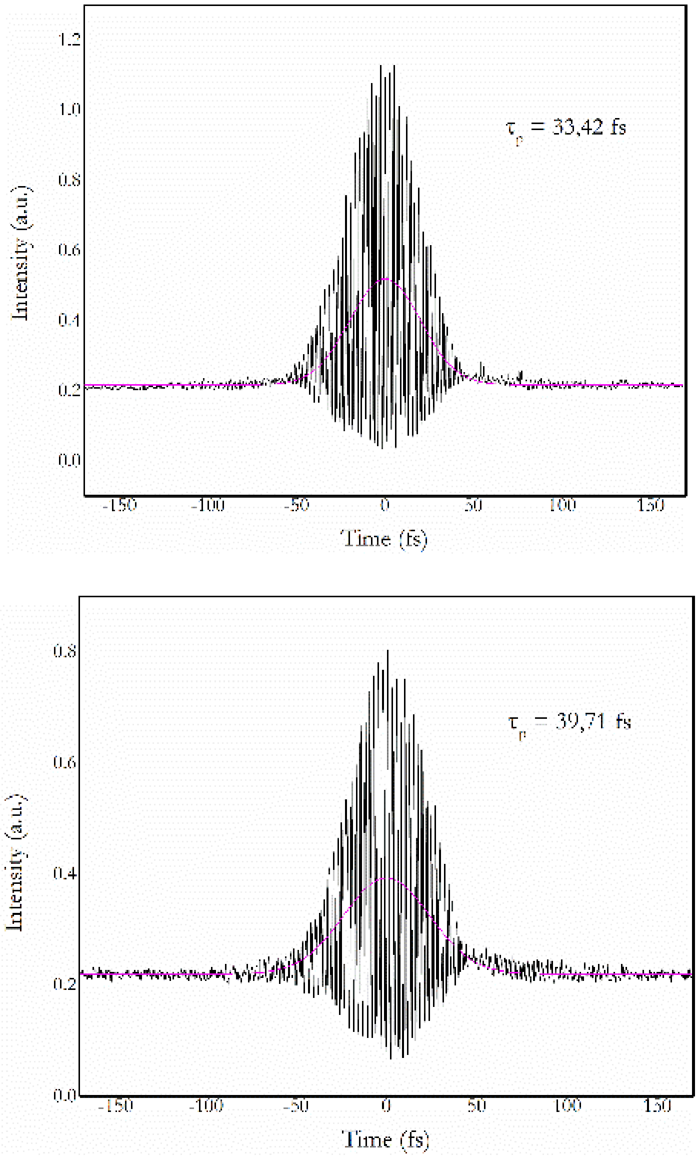

Figure 2*:* Temporal autocorrelation. above: almost fully compressed pulse, below: not optimally compressed (chirped laser pulse).

A tabletop workstation for differential pump probe spectroscopy with tunable pump (VIS - near-VIS & near-IR) and white light supercontinuum probe lines is realized. To build such a tabletop workstation one needs an interferometer set up in a configuration to use with the pump probe technique. This needs to be combined with Optical Parametric Amplifiers (OPAs) to achieve tunablity of the excitation beam. Our setup is readily available and provides enhanced capability of differential pump probe measurements. This is achieved by splitting of the probe beam in two parts and their use as Signal Reference functions. Moreover, in order to monitor main experimental parameters, as are pulse duration and dispersion characterization of the light sources involved in the experiments, appropriate tools are built. Those include (apart from the optics constituting experimental setups of such kind), the development of the software which synchronizes the equipment, records the results and analyses them for interpretation.

Use of the pump probe technique nessecitates an interferometer (Mach Zehnder) (Figure 3) featuring one arm of controllable length. The setup we have built has secondary light sources generated at each interferometer arm, namely, frequency doubling at the pump arm by use of a BBO crystal and white light supercontinuum generation at the probe arm. Additionally, the option exists for the pump arm to consist of the output bam from the home-built Noncollinear Optical Parametric Amplifier (NOPA).

White light suercontinuum is generated via optical rectification and plasma. This is done by tightly focusing the ultrashort pulses of the probe beam in a quartz cuvette filled with distilled water, while propagating it through a telescope to maintain beam collimation, shown in *Figure 3*. The supercontinum pulses are guided to a delay line which enables control of the length of the probe arm, which sets the relative time delay between two pulses. The delay line consists of a motorized stage with a mounted retroreflector, set on the optical path, thus, providing control over the travel distance of laser pulses along the path.

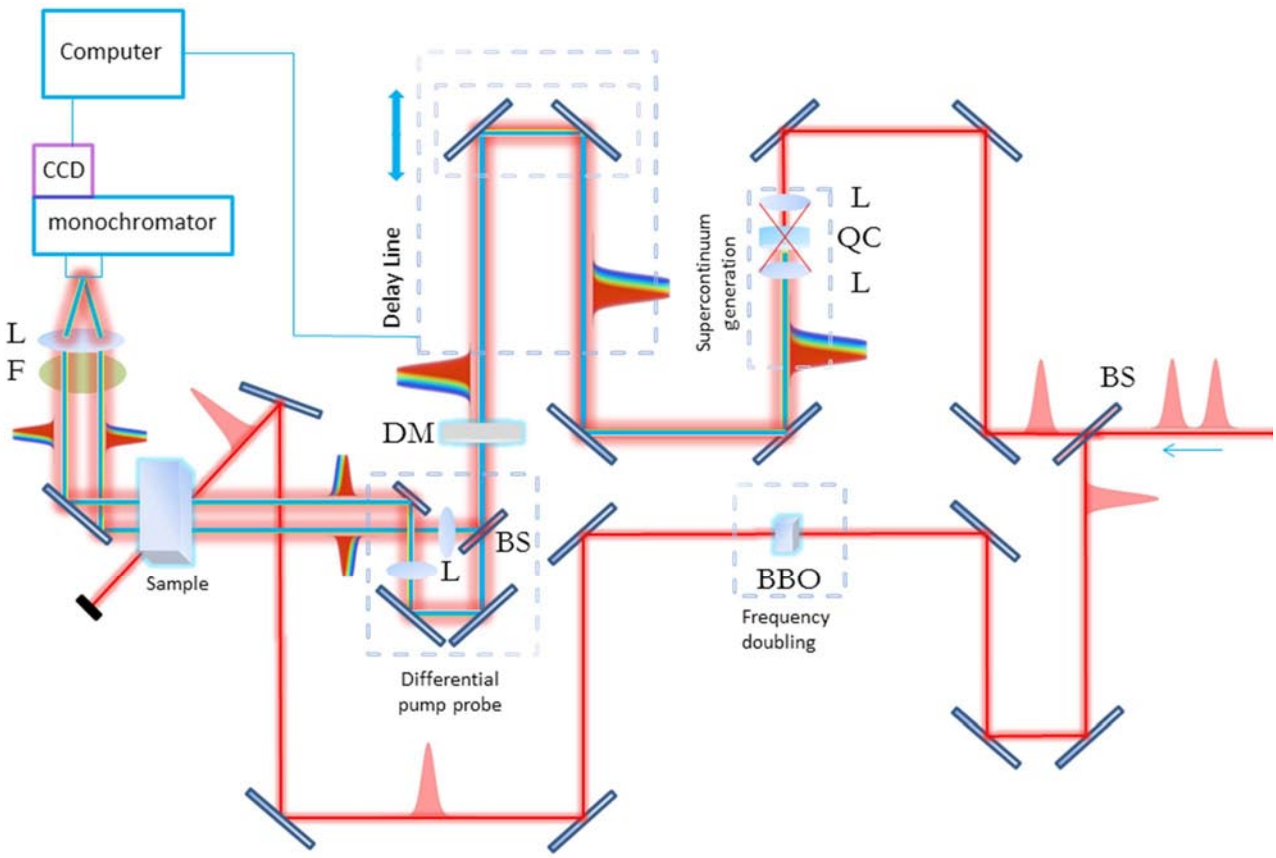

Figure 3: Differential pump probe setup. BS: Beam Splitter, L: lens, DM: dichroic mirror, F:band-pass & absorbance filters. The initial pulse pair and the generated supercontinuum pulses appear spatially separated for representation purposes.

For provision of *differential* pump probe measurements, our probe beam is split in two. The resulting beams are used as Signal and Reference probe beams. The former, overlaps with the pump beam at the sample plane probing the changes induced by the excitation. The latter, is vertically displaced and is directed to an unperturbed area of the sample. Both beams are directed at the detection system which consists from a monochromator with a CCD camera mounted at its exit. By concurently recording the signal and reference beams we are able to obtain differential pump probe measurements. A workstation consisting of the above provides a powerfull tool for pump probe measurements with broad probing capability.

The motor stages comprising the delay stages, in both Michelson and Mach Zehnder type interferometers, need be electronically controlled and synchronized with the detection equipment (lock in amplifier, monichromator - CCD camera); for both interferometric autocorrelation and differential pump probe measurements. The build-up of the main part of the pump-probe workstation, i.e the Mach Zender interferometer is now complete and operational and shown in *Figure* 4.

Figure 4: Mach Zehnder interferometer.

An example of the measurement capability of the pump-probe setup for time- & spectrally resolved measurements is shown in *Figure 5*. The differential transmitted intensity is shown with a pseudo-colour representation. The y-axis is the wavelength in nm and the x-axis is the pump-probe delay time in ps.

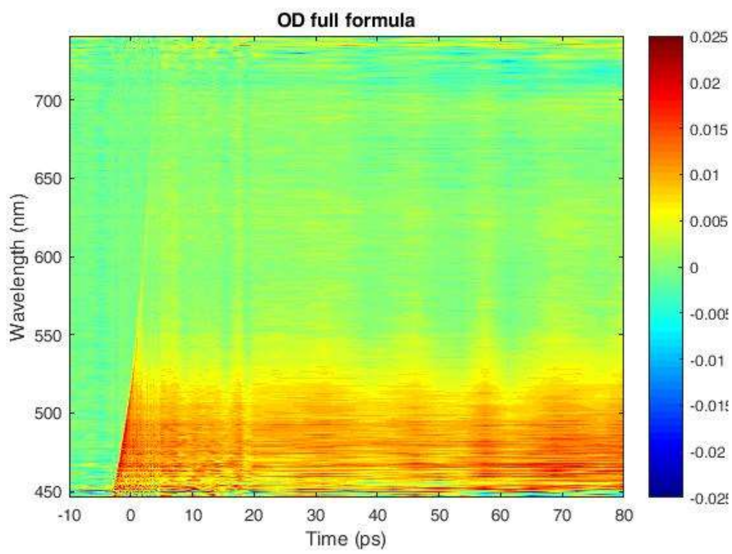

Figure 5. A time- & wavelength-resolved measurement of a thin film of a metal oxide as a result of the pump-probe workstation operation. Horizontal and vertical cross sections will reveal the ultrafast electron dynamics of the system.

For the secondary wavelength option of the pump arm of the pump-probe setup a home built visible Optical Parametric Amplifier is utilized. This optical setup takes advantage of nonlinear phenomena such as 2nh harmonic generation and White Light Supercontinuum generation and delivers a broad spectrum ranging from 430nm to 750 nm with parts f this spectrum being amplified, thus delivering an ultrashort intense laser beam with wavelength tuneable within this range. The broadband pulses are then temporally compressed using a prism pair and are sent to seed the pump arm of the pump probe workstation. The setup is shown in *Figure* 6.

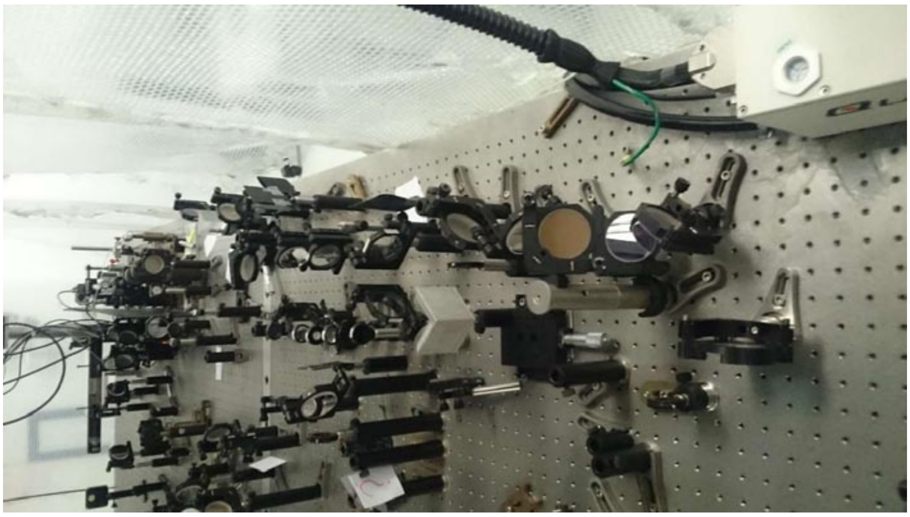

Figure 6. The optics comprising the Non Collinear Optical Parametric Amplifier.

2.2 Pump-probe THz setups

Two homemade tabletop THz time-domain spectroscopic systems (THz–TDS) are available at FORTH, providing the ability of measurements both in transmission and reflection mode (*Figure* 7).

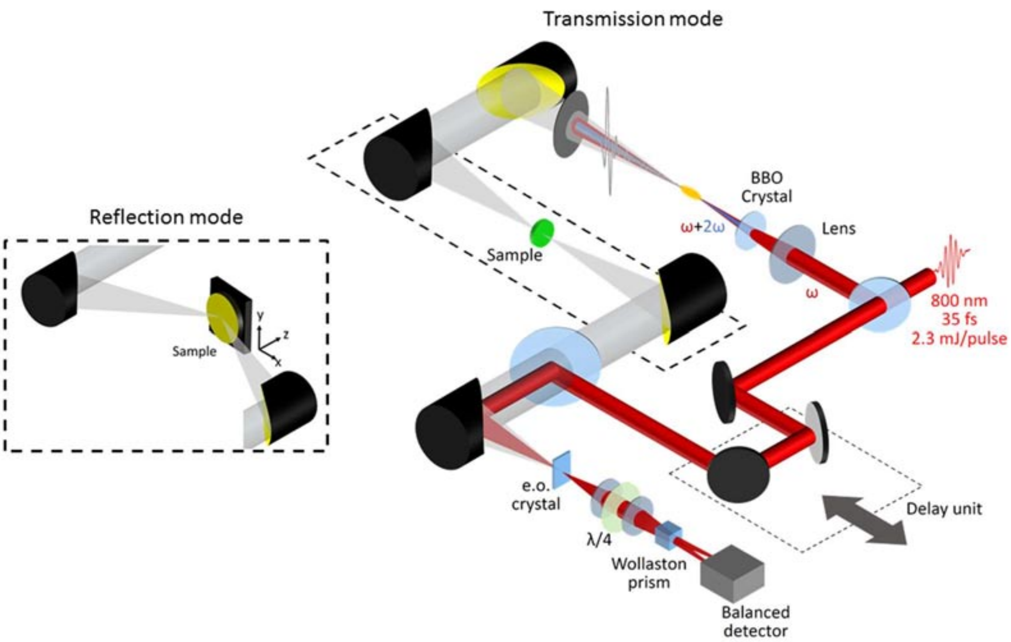

Figure 7. Terahertz time-domain spectroscopic (THz - TDS) setup.

In particular, an amplified kHz Ti : Sa laser system is used, delivering 35 fs pulses at 800 nm central wavelength and maximum energy of 2.3 mJ/pulse. Intense THz fields (> 200 kV/cm) are produced through two-color filamentation in air and their detection is achieved following either a time-resolved electro-optic (EO) sampling method for frequencies up to 7 THz (depending on the crystal), or Air Biased Coherent Detection (ABCD) technique for frequencies up to 15 THz. The amplitude and the phase of the THz pulse are obtained in the frequency domain, providing both the absorption coefficient α and the refractive index n of the studied material with high signal-to-noise ratio. Both transmission and reflection THz-TDS systems work under the same principles. However, a reflection THz-TDS setup is more appropriate for samples with high absorption or dispersion while at the same time tomographic images in THz frequencies can be obtained providing an in-depth analysis of the studied sample. The depth resolution is limited by the wavelength (~ 150 μm), while the spatial resolution by the size of the beam at the THz focus (~ 800 μm).

As an example, the optical properties in the THz frequency range of two different types of Silicon were extracted using the above transmission setup. In detail a low resistivity silicon (LR-Si) and a low resistivity black silicon (Black LR-Si) were studied (*Figure* 8). Black Si is produced by femtosecond laser ablation and is known for some interesting optical properties. The most prominent one, is that it presents high absorption up to 2500 nm compared to simple LR-Si that barely reaches 1000 nm. Figure 8 shows the absorption coefficient and the refractive index of the two samples in the THz regime up to 15 THz. Although up to 5 THz they present the same behaviour, after that frequency black Si presents higher absorption and slightly higher refractive index.

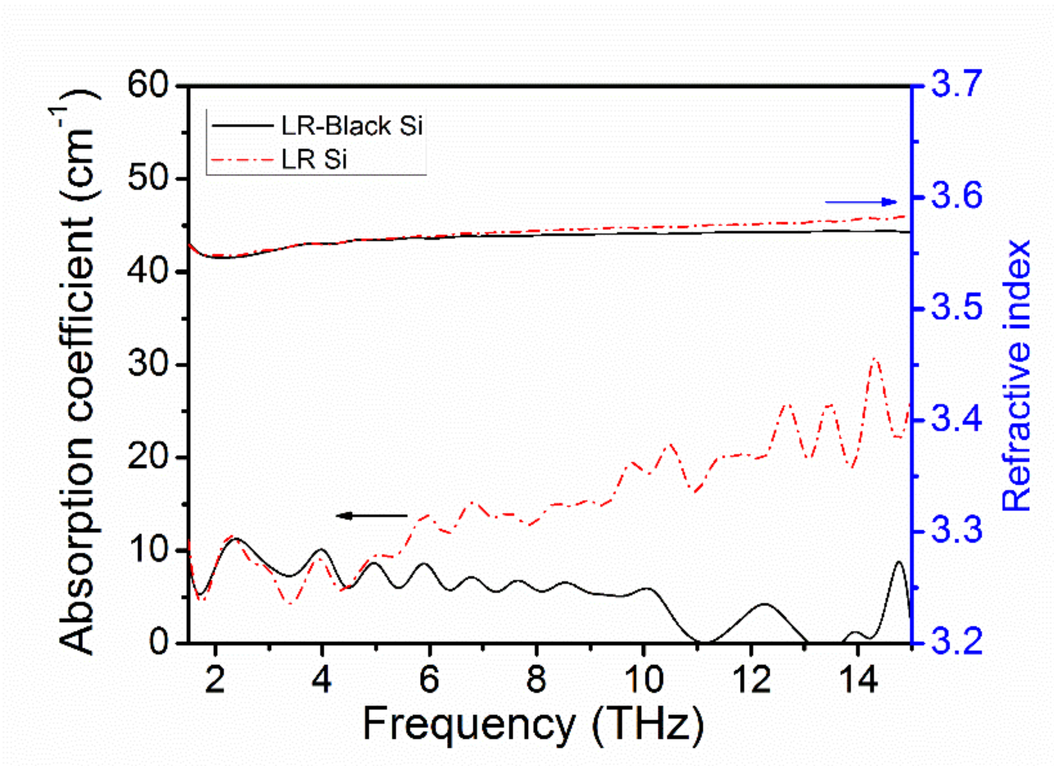

Figure 8. Optical properties of LR Black Si and LR Si in the frequency regime up to 15 THz.

Furthermore, time-resolved THz spectroscopy (TRTS) which is an optical pump - THz probe technique, can be used to study systems in which excitation initiates a dynamic change in far-infrared absorption properties on a sub-picosecond timescale. Such experiments can provide information about the lifetime and mobility of the photoexcited carriers and determine their recombination mechanism.

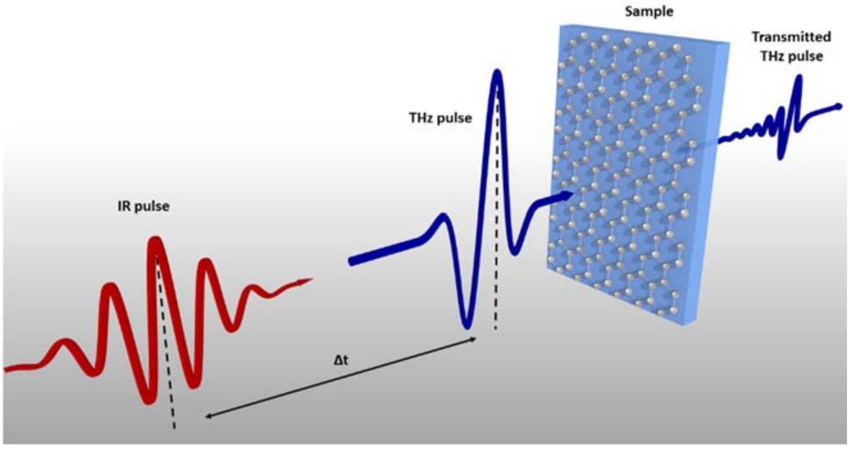

Figure 9. Time-resolved pump probe THz spectroscopy (TRTS) approach.

3. Conclusions and Perspectives

Pump-probe workstations for VIS near-VIS, near-IR and THz spectroscopy of nanosystems were realised, are currently fully operable and are readily available to NFFA-Europe users. The workstations can monitor the time-resolved differential transmissivity/reflectivity of nanosystems and can be employed in studies of the ultrafast optical, optoelectronic and structural-phononic response. The combination of the different wavelengths for pumping and probing allows one to follow in time electronic and lattice evolution, gaining thus insight in the material dynamics and allowing for better understanding and modeling of the nanosystem itself.