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Ultrafast Heterodyne Optical Sampling applied to Picosecond Ultrasonics and Nanoscale Heat Transfer

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1. Introduction

Pump-probe techniques are widely used to measure events on time scales much shorter than the resolution of electronic detectors, and are applied in such diverse fields as ultrafast spectroscopy, photo-acoustics, TeraHertz imaging, etc. In ultrafast photoacoustics measurements for instance, a pump beam launches in the sample acoustic waves, which are detected by a second, temporally shifted probe beam. Typical detection methods rely on very small changes in the reflection coefficient of the sample surface, requiring an averaging of the signal to improve the signal to noise ratio. Traditional pump-probe methods use a mechanical delay line to shift the two pulses in the time domain, where each measurement point corresponds to a single mechanical position of the delay line. Although very efficient for small measurement ranges, extending this method in the hundreds of picoseconds or nanosecond lead to a very long acquisition time, and unpractical length for the delay line.

We present a new, compact detection system, using a compact dual-oscillator ultrafast laser system, specifically designed for pump-probe measurements over time scales as long 20 ns, with a sub-picosecond resolution. This system does not use any mechanical delay line, and allows for extremely fast acquisition time.

2. Experimental setup

The system uses a new, compact diode-pumped ultrafast laser, incorporating two asynchronous femtosecond oscillators, frequency locked to each other, with a small difference (1 kHz) between the repetition frequencies of both lasers.

Each pulse is therefore temporally shifted from the previous one without any mechanical adjustment. Since the repetition rate of the laser is very high (50MHz), a complete measurement can be done in a very short time (1ms). Since there is no need for delay line control, pump beam modulation, lock-in amplifier detection, the detection system is extremely simple, and only requires a high speed detector and an averaging system.

The experimental setup is shown in figure 1 :

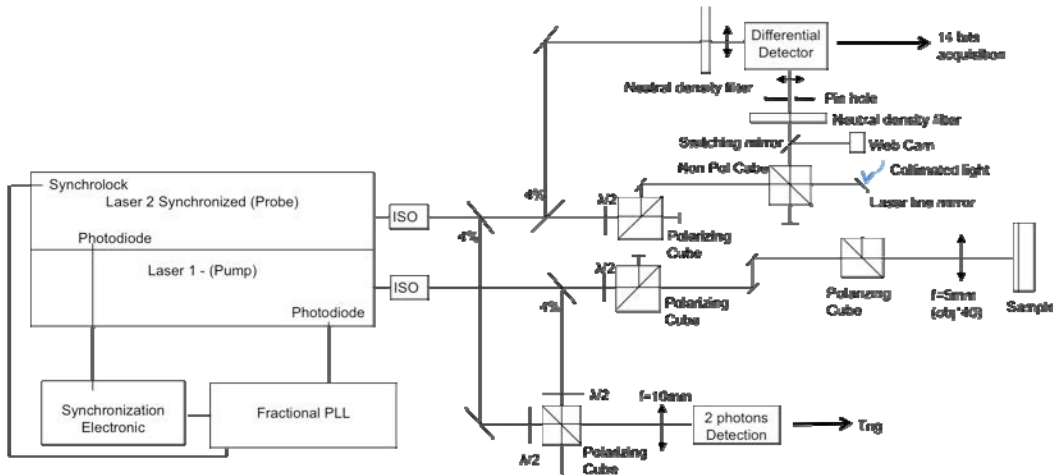


Figure 1 : Pump-probe experimental setup

Parts of the isolated pump and probe beams are combined in a polarizing prism to generate a low frequency ($\sim 1\text{kHz}$) trigger signal with the help of a two photon detection setup. The other part of the pump signal reaches the sample after intensity and polarization optimization. The probe signal reaches the sample with a slightly different angle (a few degrees) after passing through two polarizing cubes. The pump and probe beam wavelength are slightly shifted in order to eliminate the residual pump signal on the differential detector. This probe signal also loses half of its power in a non polarizing prism that is used for forward and backward beam dissociation. The probe beam reaches the non polarizing prism after being reflected on the sample, and another half of the power reaches the differential detector. A pinhole is used just before the detector to remove the slightly shifted residual parasitic pump beam. Another small part of the forward probe beam had also been selected just at the laser end to reach the second photodiode of the differential detector. This significantly improves the SNR by removing most of the probe laser fluctuation noise. The electrical signal coming from the differential detector is then acquired with a PXI NI-5122 14-bit/100MHz acquisition card. A collimated lamp and a basic web cam are used to observe the position of the laser spots on the sample.

By using this experimental setup, averaging 10000 times the signal takes only 10 s to reach a signal to noise ratio better than 10^{-7} . This high speed acquisition allows to acquire signal curves at various pump and probe beam positions. It is thus possible to acquire an image of the sample signal response by positioning the sample on a motorized XY translation stage. It is also possible to observe the photothermal and photoacoustic signal propagation by translating the pump signal using an afocal lens system with one of the lens fixed on a motorized XY translation stage.

3. Picosecond ultrasonics

The first result shown in figure 2 is the photothermal response of a 250 nm Tungsten film deposited on an Al substrate. The signal exhibits a typical fast behavior coming from the electronic response of the material followed by a slow thermal decay. Superimposed with thermal background appear up to 8 acoustic echoes showing the longitudinal acoustic wave reflected back and forth in the Tungsten film. The time of flight between successive echoes

allows the determination of mechanical properties of the film while the thermal decay can be used to identify the thermal properties of nanomaterials.

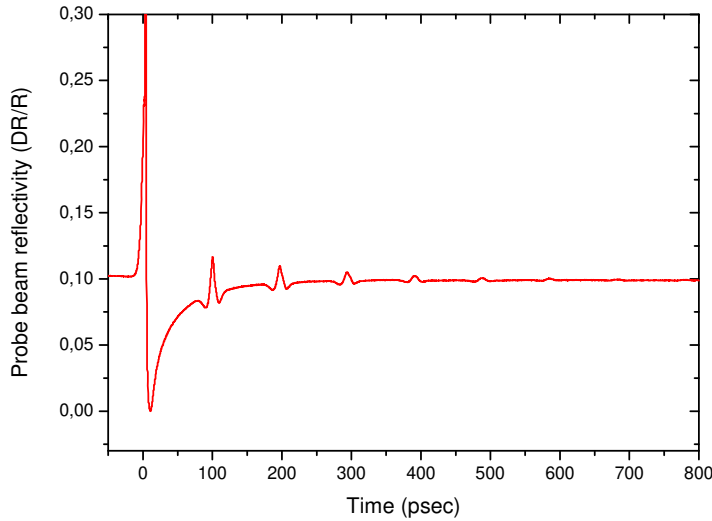


Figure 2: Transient response of a 250nm Tungsten film on an Al substrate

Once a point measurement is done, one can scan the whole surface of interest of the material and reconstruct the film of the material response. Thus figure 1 and 2 correspond to a single pixel response of the photothermal and photoacoustic movie. Frame rate is about 1 Tera images per second during 20ns. Acquisition time of the movie is a few tens of minutes. Figure 4 shows a 20*20 resolution frame of the photothermal and photoacoustic movie. One can easily see the thermal contribution to the signal giving rise, through thermoelastic coupling, to surface waves in the film. The movie shows the propagation of these surface waves on the sample surface.

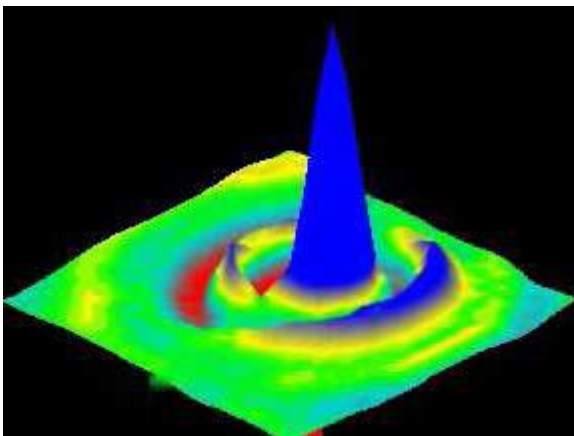


Figure 3 : Surfaces waves on a 250nm Tungsten film on an Al substrate observed with the experimental setup detailed above

3. Nanothermal applications

As the reflectance of the sample is modified by the surface thermal field one can measure as a function of time the thermal relaxation of the sample. Through the thermal decay we can identify the different thermal characteristic of complex nanometric layers. In picosecond transient thermoreflectance experiment, thermal conductivities and interface thermal resistances are carried out by comparing the experimental cooling curve to the theoretical model and optimizing the free parameters. For this purpose the active sample is generally covered by a thin metal film e.g Al, which acts as a transducer. This layer allows the conversion of optical power of pump beam into heat and the measurement of surface temperature by the probe beam through thermoreflectance [12]. The thermal model is based on the thermal quadripole method solving the heat transfer equation in a composite medium. An identification procedure allows to minimize the difference between experimental data and calculation. The thermal properties of nanometric layers are very different from bulk values. We determine the thermal conductivity of 70nm Si layer equal to 70W/m/K in excellent agreement with literature [6].

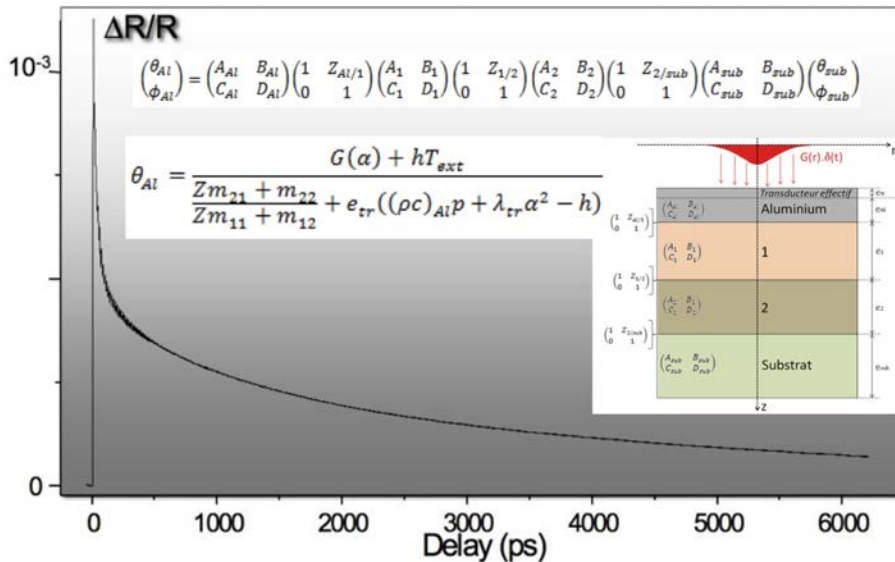


Figure 4: Thermal model of a multilayered structure.

5. References

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