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DELIVERABLE REPORT

WP11 Real-time observation and control in microscopy and spectroscopy of nano-objects

D11.3

Development of FWM setup with extended wave-vector region and with 20 fs time duration

Due date
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DELIVERABLE DESCRIPTION

Development of FWM setup with extended wave-vector region and with 20 fs time duration. Discussion of technical developments.

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NATURE

☐ R - Report

☒ P - Prototype

☐ DEC - Websites, Patent filing, Press & media actions, Videos, etc

☐ O - Other

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INTRODUCTION

During the second year of the NEP project, the development and extension of a Four-Wave-Mixing (FWM) technique, called Transient Grating (TG), has been continued and finalized. The deliverable has been postponed due to a huge problem with the main laser that was stopped for a very long period. The requirement to short pulse length didn't allow the use of a different laser currently available in our lab.

The main scope of this deliverable is the development of a FWM setup (in detail, TG), based on short pulse duration and covering an extended wave vector region, allowing to probe various dynamical ranges, not reachable by already existing schemes. The new setup was installed at SPRINT lab of the Istituto Officina dei Materiali (IOM) of Consiglio Nazionale dellle Ricerche (CNR) in Trieste.

The TG approach is mainly devoted to the excitation and detection of optical and acoustic phonons in materials. In the first case, a very short pulse duration is required, in order to follow the typical high frequencies of optical phonons (THz range). In the second case, the possibility of varying the induced wave-vector in the sample allows to measure the dispersion of acoustic phonons, and so the speed of sound extraction. The explored wave-vector region in standard laser-based TG setup is limited by the use of special optics, which cannot allow for large angles between pump pulses. The same optics fix a finite number of selected wave-vectors, not variable in a continuous way. Moreover, the use of very long pulses (hundreds of fs) in TG devoted to acoustic phonons, doesn't allow to measure contemporary the optical ones. Here we propose a new setup for measuring both optical and acoustic phonons, with a continuous variation of the induced wave-vector, which is also extended above any previous setup.

The system has been tested on various samples, and the measured frequencies have been compared with the literature. In detail, we report the analysis on optical phonons on α-quartz and optical and acoustic phonons on a glass slab, covering a wave-vector region up to 10 um^{-1}.

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Experimental progress

Transient grating setup

The source is a Ti:Sa-based amplified femtosecond laser (LEGEND, Coherent). The system produces <35 fs pulses at 800 nm with 1 kHz repetition rate, 7W output power, corresponding to 7 mJ/pulse.

This section describes the optical setup we built to perform TG measurements. A sketch of the optical system is shown in Figure 1.

The beam from the laser source is splitted in two parts. The reflected beam is used as probe, the transmitted one as pump. The last is rotated in polarization (we work with s-polarized light) using a half-wave plate, and then it is splitted again in two pulses with equal energy. The two pump pulses have been focalized on the sample at a fixed angle. The dynamics induced in the sample for acoustic oscillations depends on this angle (see Results and Discussion). The main improvement of the proposed setup is the possibility of varying this angle in a continuous way, covering a larger range with respect to any other existing setup. This can be obtained by moving the last two mirrors using two rails that are collinear to the beam path. This allows to translate the mirrors maintaining the beam in the centre of the mirrors. Also the rotation of the mirror in on axis with the optical post, and so with the beam path. In other words, any mirror position on the rail corresponds to a different angle between the pump pulses. The correct spatial overlap can be easily obtained by inserting a pinhole at the sample position. Considering the total symmetry of the two rails with the sample axis, and the use of a graduate scale on the two rails, by moving the mirror at the same position on the relative rail, also the spatial overlap is preserved (a fine tune of the delay between pump pulses is also possible by means of the pump delay line).

The probe, after passing through a delay line, is sent on a BBO generating the second harmonic (400 nm) and focalized on the sample after an interferential filtering to remove the fundamental wavelength at 800 nm. The angle between pumps and probe has to satisfy the so-called phase matching condition in order to be diffracted by the induced grating. The correct angle for any pumps configuration is guaranteed by a similar rail-plus-rotatable-mirror system, used for the pumps.

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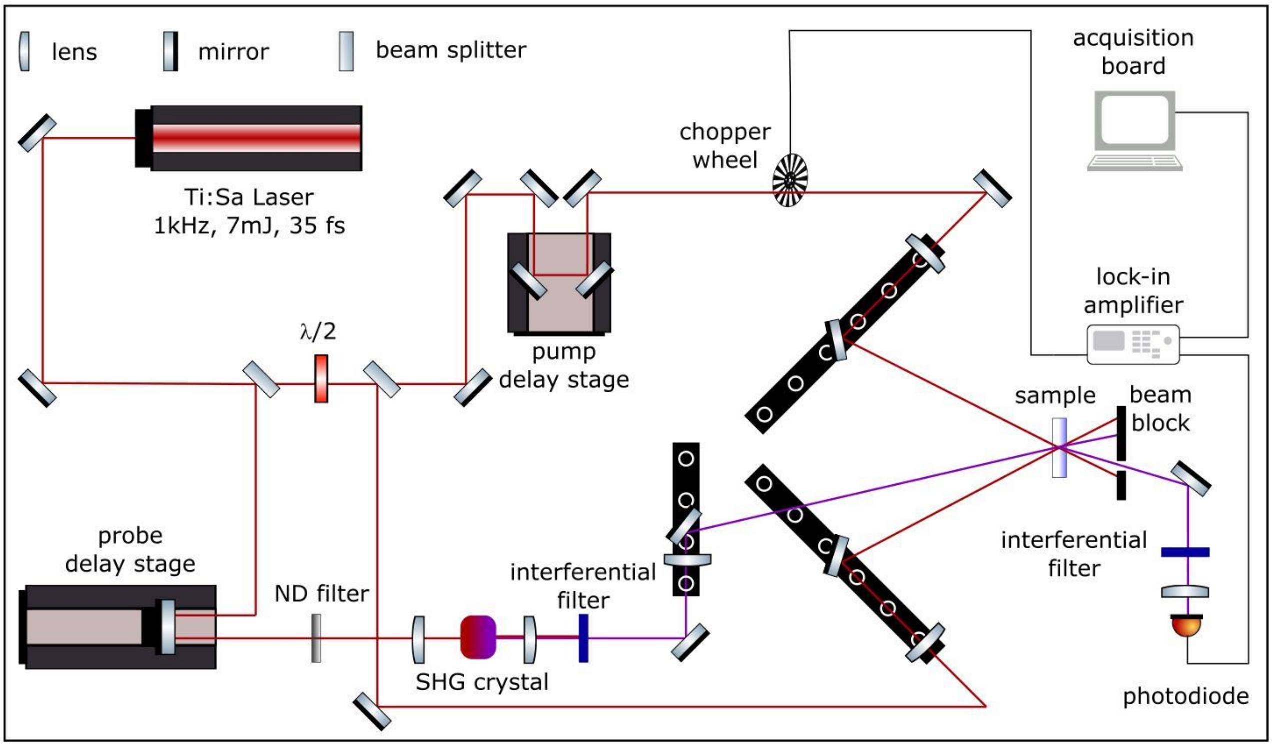

Figure 1: Transient grating setup. The laser source is the LEGEND, which generates < 35 fs pulses at 800 nm. The first beam splitter generates the pump (reflected) and probe (transmitted) beams. The pump is then splitted again to produce two pump pulses with the same energy per pulse. The two pulses are overlapped in space and time (by the use of a delay line) on the sample, producing the transient grating. The probe beam is duplicated in frequency, filtered and focalized on the sample. The scattered beam is then focalized on a photodiode and amplified by a lock-in.

The scattered beam is spectrally filtered and focalized in a 2151 Newport femtowatt photoreceiver. This photoreceiver is an extremely sensitive detector to be used in synchronous detection with a lock-in amplifier. The lock-in amplifier used is a SR850 by Stanford Research Systems. The signal is acquired directly using a homemade LabVIEW software.

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Results and discussion

We performed measurements on a SiO_2-based glass slide from Carlo Erba Reagent. The measurements on optical phonons have been performed also on a α-quartz sample, a material that is well-known for a strong signal from optical phonons, and can be considered as a standard for optical phonons frequencies.

The measured signal on the SiO_2 slab on the entire time scale is reported in figure 2. The signal is characterized by a very fast signal at time zero (the so-called "coherent peak"), shown by the very intense peak at time zero, which is a sort of cross correlation of the laser pulses, and describes the Gaussian profile of the pulses (not reported in the zoom). After the first hundreds of femtoseconds the optical phonons signal rises up, showing an oscillating behavior, with a frequency of 2.6 THz (see green zoom). Around 100 ps after time zero also the acoustic phonons signal appears. The time duration of the signal is limited by the delay stage length (around 30 cm). The achievable time resolution is around 7 fs.

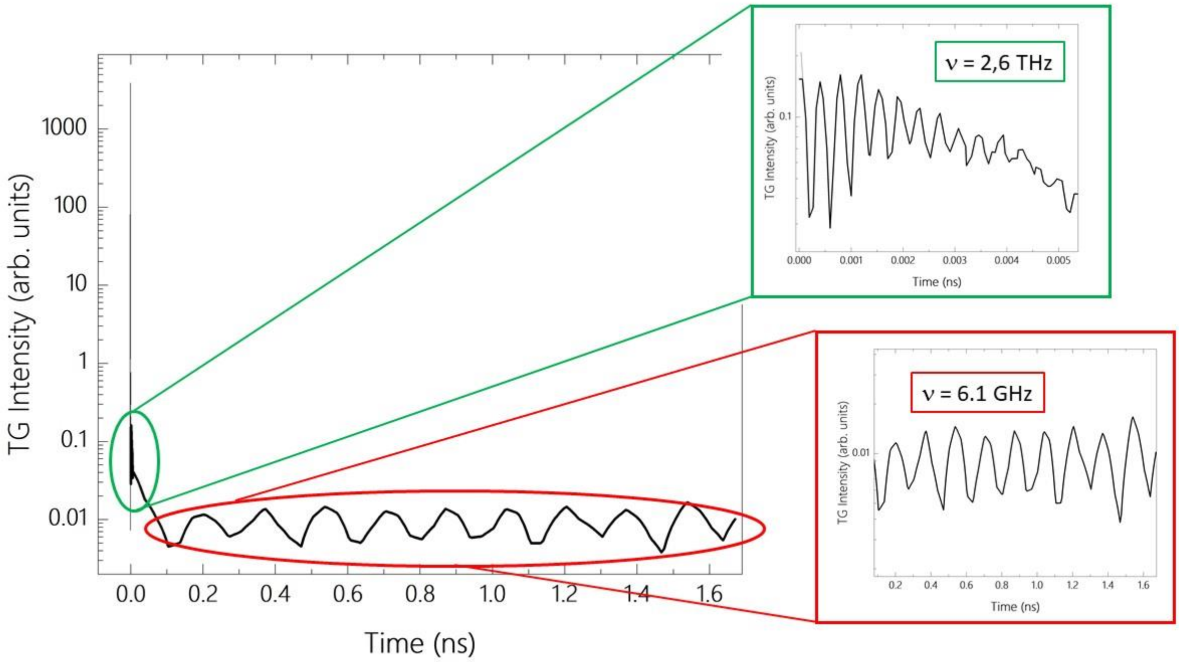

Figure 2: TG signal on SiO_2-based glass slab on the entire accessible time window. The two graphs in green and red boxes show a zoom at short and long times, where optical and acoustic dynamics appear, respectively.

The recorded signal covers around 4 order of magnitude in time, allowing to measure three different dynamics (coherent peak, optical and acoustic phonons). This can be achieved avoiding the use of diffractive elements (usually used in laser-based TG setup) and with the use of very short pulses (< 35 fs). The suggested technical solutions allow to combine the two requirements without losing an easy-alignment condition. Another important feature is the possibility to cover 5 orders of magnitude of intensity, fundamental aspect to measure dynamics with very different efficiencies in a single measurement.

In order to compare our results with a well-known sample, we measures the optical phonons on α-quartz sample (see figure 3). The signal shows an optical phonons frequency of 3.85 THz (128 cm^{-1}), in perfect agreement with the value reported in literature for the first optical branch on this material (Opt. Comm, **44**, 411,1983). The very good signal-to-noise ratio indicates how the TG technique should be suitable also for the optical phonons dynamics. In particular, the optical phonons are usually measured with time-resolved reflectivity, which is a well-established approach. But it requires a reflective sample, making this technique not so useful with transparent samples. The reported measures demonstrate how it can be easily achieved with TG approach. The solution suggested on this report indicates how both the dynamics (optical and acoustic) can be measured in the same setup.

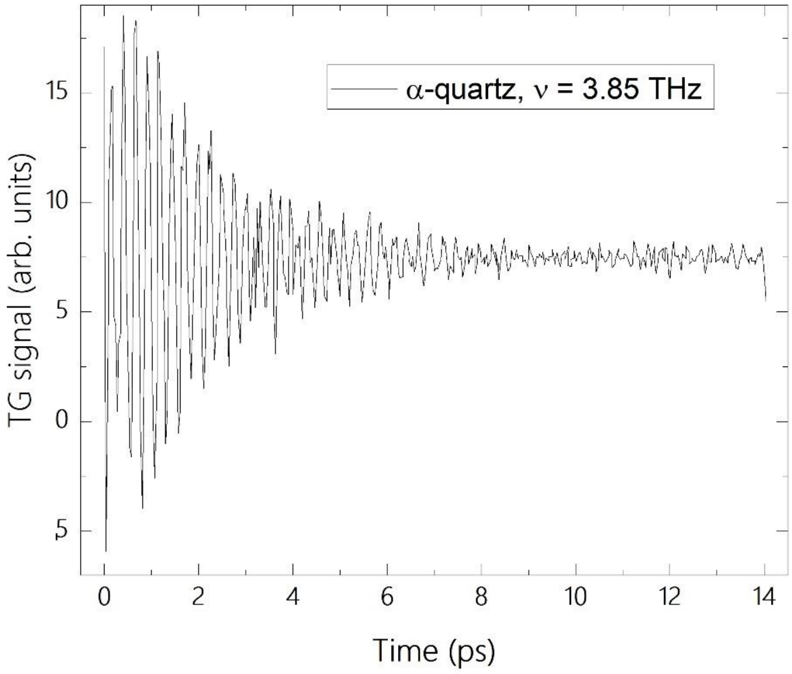

Figure 3: Optical phonons on α-quartz. The measured frequency ν= 3.85 THz (128 cm^{-1}) corresponds to the first optical branch.

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The possibility to change in a continuous way the angle between pump pulses, allows to measure the acoustic dispersion in materials. The angle between pumps and their wavelength fix the line spacing of the induced grating Λ, and so the induced wave-vector q, following the relation $q=2\pi/\Lambda = 4\pi\sin(\theta)/\lambda.(\theta)/\lambda$, where θ is the half angle between pump pulses, and λ is the pump wavelength. By varying the value of q, it is possible to induce acoustical phonons with different frequencies ω. By plotting the frequency as a function of q, it is possible to extract the sound speed, by $\omega=cq$. So, in order to measure the dispersion of acoustic waves, a continuous variation of the angle between pump pulses is required.

The figure 4 shows the dispersion relation for the glass slab, by varying the q vector in the whole available range. Any q vector can be chosen in this range, depending only by the mirror position on the rail (different positions correspond to different angles between pump pulses). Thanks to the very high signal-to-noise ratio, the sound velocity can be extracted with a relative error lower than 1%.

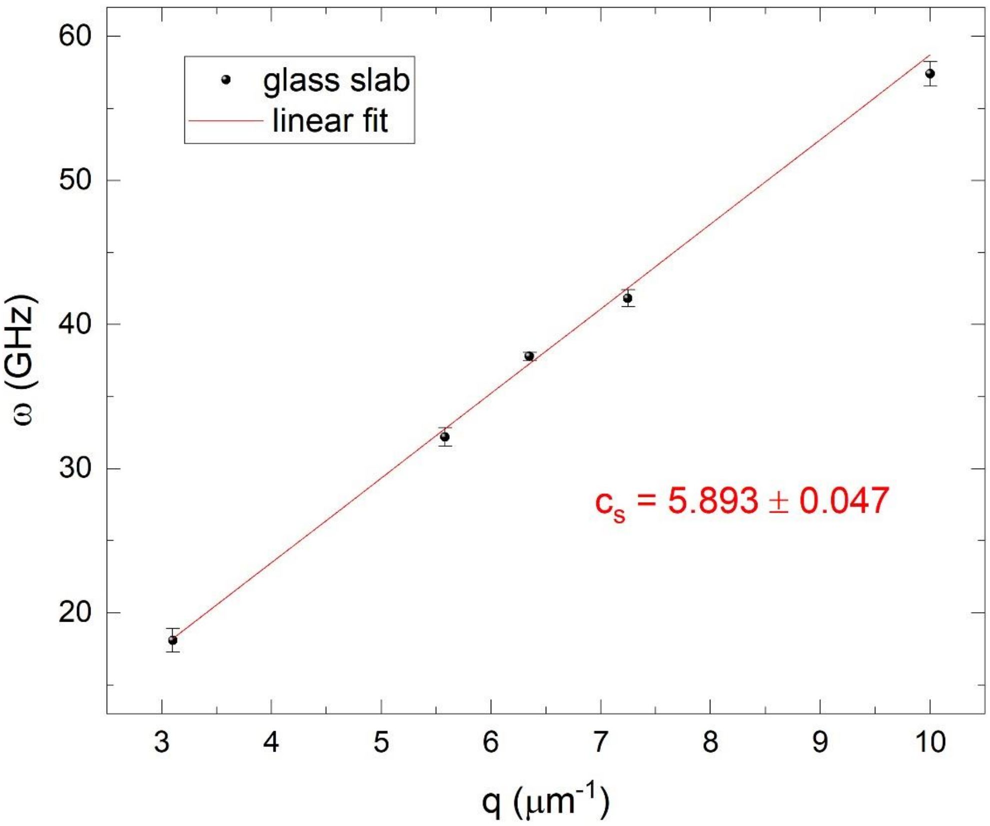

Figure 4: Dispersion curve of acoustical phonons, measured by choosing five different angles between the pump pulses. The extracted sound speed is in agreement with literature on SiO_2-based glasses.

The higher reported value of $q=10um^{-1}$ is almost a factor 2 higher than any other existing laser-based TG (the standard TG setups work in the range 0.5-3 um^{-1}). This range can be further extended working with the second or third laser harmonics. The setup requires only small adjustment, considering the almost totally use of reflective broadband optical elements. This will allow a complete superposition with other techniques like Brillouin light scattering, covering the currently unmeasurable q range between them.

In conclusion, we developed a new experimental setup based on FWM approach covering an extended wave-vector region, and suitable for measuring the optical and acoustical degrees of freedom in transparent media. The combination of short pulses and continuous variable angle between pump pulses allow to measure in the same setup both the degrees of freedom, with a very high signal-to-noise ratio. The setup can be considered an alternative method to time resolved reflectivity for transparent media.

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