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Femtosecond interferometry: absolute measurements of optical susceptibilities

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ABSTRACT

An improved time resolved interferometric technique characterizes the optical nonlinear response of various liquids and solids. Absolute values of optical third order susceptibilities have been measured and molecular dynamics extracted from coherent signals. Subfemtosecond processes have been evidenced using this new analysis.

INTRODUCTION

A Mach-Zehnder interferometer is used to measure and time resolve the optical nonlinear response of various liquids and solids. The basic experimental setup is presented in fig 1. The sample is located in the probe arm and interacts with the stronger copropagating pump beam orthogonally polarised. A KLM Titanium-saphir laser (Coherent MIRA 900) emitting ultrashort pulses around 800 nm has been chosen as the source with respect to its unique stability in amplitude and pulse shape. We will report here on time resolved measurements in which the time delay τ between the pump and the probe pulses gives the basis of this sampling interferometry. We have implemented a fringe locking technique to stabilize the phase difference between the probe and the reference beam and thus increase the sensitivity of the detection.

As all the relevant parameters such as optical power, beam profiles and sizes, pulse shape, time delay and interferometer sensitivity are precisely measured, absolute values of the nonlinearity can be computed⁽¹⁾.

Moreover, in this particular experimental fully frequency degenerate configuration, we showed⁽²⁾ that, in our experimental setup, coherent effects between pump and probe pulses have to be fully analyzed to extract absolute values of the relevant susceptibilities. The complete recording of the signal as shown in fig 2 (left) displays coherent modulations, i.e "nonlinear fringes" oscillating at twice the optical period, whose shapes and amplitudes are dependant of the involved nonlinear process. Using this analysis, we present new results

where this nonlinear fringes are interpreted in terms of speed of response at the femtosecond time scale, much below the pulsewidth.

EXPERIMENTAL SETUP

Interferometric basis

The experimental setup is presented in the figure 1.

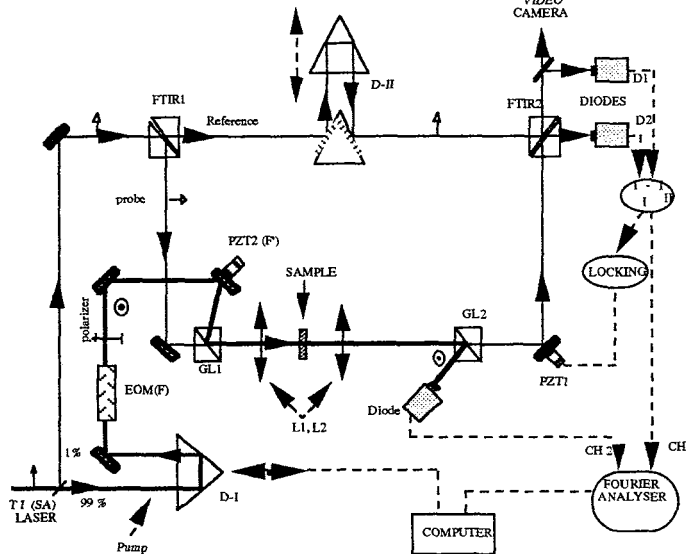


Fig 1: Experimental setup. Modified Mach-Zehnder three beams interferometer.

First, without the pump beam, the interference between reference and probe beam is adjusted via a delay DII and the fringe contrast optimized with the help of two adjustable Frustrated Total Internal Reflectors to take into account all the differential losses between the two arms intensities. While keeping a perfect overlap between the two beams, an afocal in the probe arm provides a convenient beam waist in the sample and controls the phase front to maintain a far field interference pattern free of any fringes. The sample is located at the beam waist (size $w_0 = 10 \mu\text{m}$) at the focus of a 4 cm focal lens. Since the two outputs display complementary interference states, the two photodiodes $D_{1,2}$ (DT-25, EGG) are connected in opposition in such a way that a zero difference photocurrent indicates an optical phase difference of $(2k+1)\frac{\pi}{2}$ within the coherence length of the laser. The corresponding error signal is used to lock the interferometer to this value via a high voltage amplifier and a piezoelectric transducer PZT1. At this locking point, any small phase variation induced by the pump beam in the sample will appear as a voltage proportional to the optical phase excursion. In order to improve the nonlinear optical phase measurement, we have stabilized the phase difference between the probe and the reference beam within 1 milliradian. The scaling factor was easily measured by recording the whole interference fringe as the path difference via D-II

is varied- servo unlocked. An electronic servo bandwidth of $f_c = 1,5$ kHz was sufficient to reduce mechanical instabilities and index fluctuations at this performance level.

The variation of the same difference signal as the pump beam was applied in the sample was used to measure the nonlinear phase shift.

Pumping scheme of the sample

Introduction of the powerful pump beam into the sample and its ejection was realized using two high rejection ratio Glan prisms. It reduces the coupling between pump beam and interferometric signal to a leak η of the powerful pump beam into the detection system. In order to separate the nonlinear signal from the noise, we use the power of a Fourier analysis. We modulate the pump beam field amplitude at a frequency f_{mod} and its intensity at $2f_{\text{mod}}$ with an electrooptic modulator (Conoptic 354) and a polarizer. The resulting modulation of the signal is recorded using a spectrum analyzer (Tektronix 2622). The evolution in amplitude and phase of the relevant Fourier component at $2f_{\text{mod}}$ versus the delay τ (delay D-I) between the pump and the probe beam enables us to analyse the dynamics of the optical nonlinear processes .

COHERENTS EFFECTS

As pump and probe beams given by $E_{\text{pump,probe}}(t) = \xi_1 \cos(\omega t - kz)$ are at the same frequency ω are exactly copropagating inside the sample, a strong coherent coupling arise when they overlap also in time. It can be shown that the nonlinear polarisation of an isotropic medium where the fourth-rank tensor response $R_{ijkl}^3(t)$ reduces to 3 independent terms : $R_{xyxy}^3(t)$, $R_{xyyx}^3(t)$ and $R_{xyxx}^3(t)$ and can be written as:

$$P_x = P_{x \text{ Kerr}} + P_{x \text{ coh}}$$

$$P_{x \text{ Kerr}} = E_{\text{probe}} \int \frac{R_{xyxy}^3(t-t')}{2} \xi_{\text{pump}}^2(t'+\tau) \{1 + \cos(2\omega(t'+\tau) + 2kz)\} dt'$$

$$P_{x \text{ coh}} = E_{\text{pump}} \int \frac{R_{xyyx}^3(t-t')}{2} \xi_{\text{probe}}(t') \xi_{\text{pump}}(t'+\tau) \cos(\omega \tau) dt'$$

As a result the induced nonlinear phase shift in the transmitted probe field gives an interferometric response integrated over the pulse duration.

In our experimental setup, for a pure electronic case-instantaneous response-all the third order susceptibilities are equal to $\frac{\sigma}{6}$ and the nonlinear phase shift will appear as a signal proportional to the following expression as the delay τ between pulses varies:

$$\frac{\sigma}{3} I_p(\tau)(2 + \cos(2\omega\tau))$$

where σ is third order optical susceptibility dependant.

This signal plotted versus the time delay τ between pump and probe will display nonlinear interferences of a contrast of 1/2 weighted by the pump intensity time dependence. Indeed, a recording of the nonlinear response for such a material -the standard SF59 Shott

glass- displays such a shape as indicated in the figure 2 (left). This confirms the validity of the instantaneous response of the studied glass.

Moreover, as we can record simultaneously in this interferometric technique the applied optical field -the linear fringes in the interferometer between pump and reference beams- as well as the nonlinear response -nonlinear fringes due to pump-probe interaction, any delay will appear as a lag between the two set of fringes. A zoom in the preceeding figure close to zero delay shows a perfect synchronism between the two sets of fringes, indicating the ultrashort response time (\ll fs) of this optical nonresonant Kerr process, much below the pulse width.

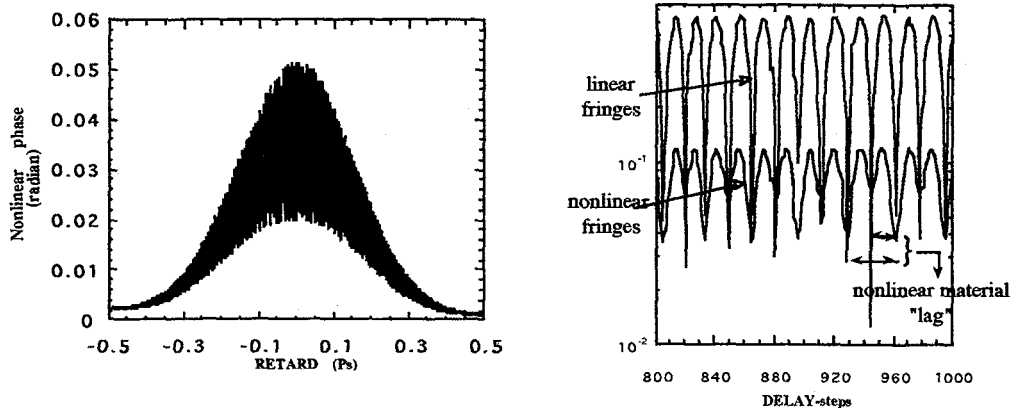


Fig 2: Left: Nonlinear response of a Glass(SF59) .Right: Zoom with linear and nonlinear fringes.

This sub laser pulsewidth technique has been applied to different nonlinear processes such as reorientation in CS_2 or even resonant processes like one or two photons absorption. In this case, the corresponding lag increase widely as the laser is tune into the corresponding resonance.

CONCLUSION

A powerfull interferometric tool has been developped to measure and time resolve nonlinear processes. A new analysis taking into account coherent effects has proven its ability to extract from the signal sub laser pulsewidth (i.e femtosecond) dynamics in disordered medias such as glasses or molecular liquids.

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