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First Measurement of the Non-instantaneous Response Time of a $\chi^{(3)}$ Nonlinear Optical Effect

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Abstract. The third harmonic of a few-cycle pulse, generated at different dielectric surfaces, is investigated using interferometric frequency-resolved optical gating. We present direct experimental evidence for a non-instantaneous nonlinear response in a TiO_2 thin film whereas surface third-harmonic generation in a SiO_2 sample does not show any indication for non-instanteneity. To the best of our knowledge, this constitutes the first report of a non-instantaneous nonlinear optical response of a dielectric optical material.

1 Introduction

Nonlinear optical $\chi^{(3)}$ effects such as third-harmonic generation, four-wave mixing, self-focusing, and self-phase modulation are the keystone of nonlinear optics and appear in any optical material. These effects are the basis of supercontinuum generation and, together with anomalous dispersion, give rise to the appearance of optical solitons. Self-phase modulation, in particular, is a key mechanism for the implementation of ultrafast optical absorbers. Using mechanisms like Kerr-lens mode-locking (KLM) or additive-pulse mode-locking (APM), the reactive quasi-instantaneous Kerr nonlinearity can be converted for the implementation of an ultrafast optical absorber. This mechanism is key for few-cycle pulse generation directly from the oscillator, and, in turn, the generation of attosecond pulses and high harmonics. Little is known, however, about the speed of these saturable absorbers. In literature, estimates in the range of 0.1 to 5 fs have been given for the response time of the Kerr effect in dielectric media [1,2]. It is generally accepted that the Kerr nonlinearity is significantly faster than the shortest oscillator pulses. Therefore, this nonlinearity is generally labeled "quasi-instantaneous". Deviations from instantaneity have been reported for nonlinearities in semiconductors or for resonantly driven $\chi^{(2)}$ systems [3]. In the following, we will demonstrate a significant temporal broadening in the third harmonic response of a $\chi^{(3)}$ nonlinear optical material. To the best of our knowledge, this report constitutes the first measurement of a finite lifetime of a $\chi^{(3)}$ effect in a dielectric material.

2 Experiment

We used a Venteon Ti:sapphire oscillator delivering 6.5 fs pulses, which is among the commercially available laser sources with shortest pulse durations. Pulses from this laser are launched into a dispersion balanced Michelson interferometer to generate a pulse pair with nearly equal energy at adjustable delay in a collinear geometry. This part of the set-up is therefore similar to interferometric autocorrelators, as frequently used for pulse characterization. Bulk dispersion of the beam splitter substrate and other group-delay dispersion contributions are compensated for by two chirped mirrors. The pulse pair is then either focused onto a TiO_2 thin film (600 nm thickness, [3]) or onto a silica substrate for surface third-harmonic generation. The generated third harmonic is then analyzed in a spectrometer, using an electron magnifying CCD camera (Andor Newton) in the Fourier plane. This camera enables a spectrally dispersed analysis of the interferometric autocorrelation signal at the single-photon level, which essentially duplicates the interferometric FROG technique, yet using third-harmonic generation rather than second-harmonic generation [4,5].

Example measurements for SiO_2 and TiO_2 are shown in Fig. 1. Using the $\chi^{(3)}$ nonlinearity in fused silica (SiO_2) the measured interferometric FROG traces [Fig. 1(a)] and autocorrelations [Fig. 2(b)] are perfectly symmetric. Using Fourier filtering techniques, the dc kernel of these traces are isolated. Unfortunately, one cannot use standard FROG software for retrieval of pulse shapes from these kernels, as they consist of a linear combination of standard SHG and THG FROG traces. We therefore used a simple retrieval software that is based on the Levenberg-Marquardt algorithm for local optimization. Seeding this retrieval procedure with an unchirped sub-10 fs Gaussian pulse, we can in fact retrieve a short slightly asymmetric pulse with 6 – 7 fs pulse duration for the input pulses from the Venteon Ti:sapphire laser. This duration is also independently confirmed with other $\chi^{(2)}$-based autocorrelators and SPIDER diagnostics. Exchanging the fused silica $\chi^{(3)}$ medium by the TiO_2 thin film medium, however, we observe interferometric FROG traces and autocorrelations that are significantly wider than those measured with SiO_2, see Figs. 1(b) and 2.

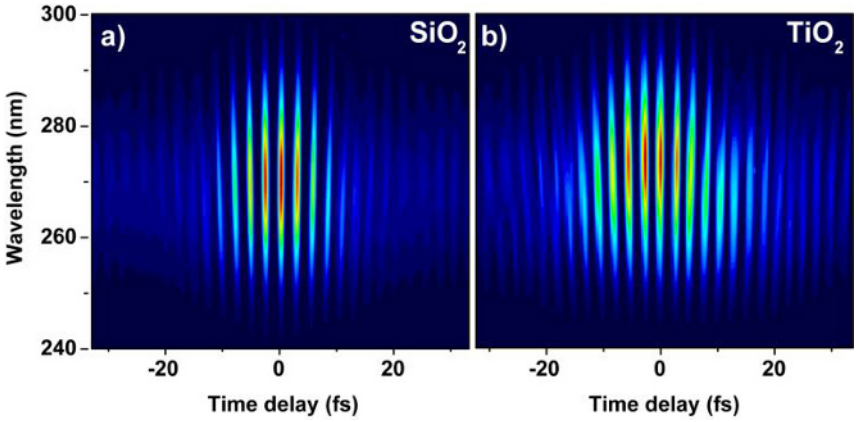

Fig. 1(a). Interferometric FROG measurements of a 6.5 fs Ti:sapphire pulse using surface THG in silica. The trace reveals a Fourier limited pulse. (b) The same measurement with a 600 nm TiO_2 thin film as the nonlinear medium. The trace is substantially broadened.

In many aspects, these measurements resemble the analysis of the relaxation dynamics of surface SHG on a plasmonic nanotip [6]. Comparing the SHG IFROG traces measured with the nanotip and with a BBO crystal, a relaxation time constant of about 20 fs of the plasmonic nonlinearity could be determined in a deconvolution analysis [6]. Using a similar deconvolution procedure for the data in Fig. 2, we estimate a relaxation time constant of about 5 – 6 fs in TiO_2. This is quite remarkable, as it surpasses the optical cycle at 800 nm by a factor of 2. To the best of our knowledge, these measurements constitute the first positive report of a non-instantaneous lifetime of a nonlinearly induced polarization in a dielectric material.

3 Discussion

The TiO_2 thin film used in the experiments already exhibits a measurable absorption for the third harmonic of the Ti:sapphire pulses. Therefore, THG may be connected to the build-up of free carriers inside the material, even though the observed few-femtosecond relaxation times do not appear to match respective recombination time constants. Alternatively, the finite life time may be explained in the harmonic oscillator framework of nonlinear optics in dielectric media. In the latter case, the relaxation time constant should stay constant, regardless whether the THG photon energy exceeds the band gap energy or not. In that case, one can also make conclusions on a lifetime of other $\chi^{(3)}$ effects, and one can derive scaling laws for the lifetime of the effect as a function of the bandgap of the material. Applying such simple scaling laws, we predict a lifetime of 0.8 fs in silica. This theoretical understanding corroborates that an increase of nonlinearity is always accompanied by an increase of the response time of the effective nonlinear absorber, clearly imposing a physical limit for the obtainable switching speed in passive mode-locking devices. Further experiments are underway to examine the wavelength scaling of the effect.

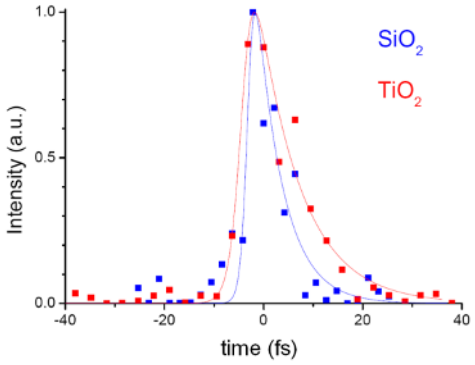

Fig. 2: Reconstructed temporal traces from the IFROG measurements in Fig.1. The broadening in the TiO_2 case is attributed to the finite dephasing time.

4 Outlook and Conclusion

In summary, we have been able to measure a finite lifetime of the nonlinearly induced polarization in dielectric $\chi^{(3)}$ media, enabling a quantification of the word "quasi-instantaneous" for the first time. While it has been considered that such effects only appear on the attosecond scale, we provided ample evidence its appearance on the femtosecond scale. Our measurements indicate a lifetime of the nonlinear polarization of 5 fs in TiO_2. Further experiments are underway to clarify wavelength and bandgap dependence of this effect.

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