Non-instantaneous polarization decay in dielectric media

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Abstract: We demonstrate experimental evidence for non-instantaneous polarization decay in dielectrics. The few-femtosecond relaxation times agree favorable with solutions of the time-dependent Schrödinger equation and relate to resonances of the quantum mechanical dipole.

Nonresonant nonlinear optical effects in dielectric media are typically instantaneous, with the nonlinear susceptibilities $\chi^{(2)}$ and $\chi^{(3)}$ having no explicit dependence on time. The invariability of the susceptibilities has important technical implications. The generation of few-cycle pulses via Kerr-lens mode-locking critically relies on this assumption, and pulse characterization methods like FROG or autocorrelation would produce incorrect results if the employed nonlinearity had a temporal dependence. The instantaneous character of the nonlinear response is expected to break down whenever one of the optical frequencies involved is in the spectral range of a resonance of the nonlinear medium. Here we provide experimental evidence that conditions exist where the induced polarization in a dielectric medium does not directly follow a power of the electric field but exhibits a non-instantaneous decay, with decay times that can last up to a few femtoseconds. More specifically, this lasting effect appears when the respective harmonic comes into resonance with the band gap of the dielectric. This effect is seen, both experimentally as well as in solutions of the time-dependent Schrödinger equation. This finding may have important consequences for pulse characterization in the ultraviolet and for Kerr-lens mode-locking in narrow-band dielectrics. The demonstrated relaxation time constants are among the fastest processes ever observed with near-infrared ultrashort laser pulses.

In our experiments, we used a Venteon Ti:sapphire oscillator delivering 6.5 fs pulses, which 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 widely similar to an interferometric autocorrelator. Group-delay dispersion of the beam splitter substrates is pre-compensated by two chirped mirrors. The pulse pair is then either focused onto a TiO_2 thin film (600 nm thickness, [1]) or onto a silica substrate for surface third-harmonic generation (THG). Subsequently, the generated third harmonic is 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. Our set-up duplicates the interferometric FROG technique, yet with the noted difference of employing third-harmonic generation rather than second-harmonic generation [2,3].

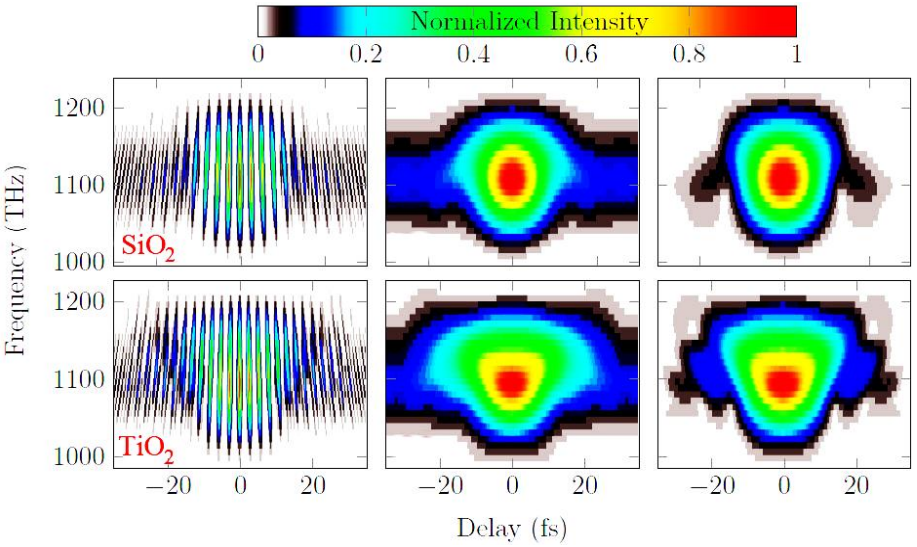

Fig. 1. Interferometric FROG measurements of a 6.5 fs Ti:sapphire pulse using surface THG in silica (top row) and in a 600 nm thick titania film (bottom row). Original measurement data is shown in the left column, the extracted cycle averaged dc FROG trace $S_0(t,\omega)$ in the middle column, and the amplitude of the fundamental oscillatory component $S_1(t,\omega)$ in the right hand column.

Example measurements for SiO_2 and TiO_2 are shown in Fig. 1. Using Fourier filtering techniques, we isolated the slowly varying dc kernel $S_0(t,\omega)$ of these traces as well as the modulation envelopes at the fundamental carrier frequency and its second harmonic, $S_1(t,\omega)$ and $S_2(t,\omega)$, respectively. In order to employ the full information contained in the traces, we wrote a specialized retrieval software to reconstruct the electric field structure $E(t)$ underlying the measured FROG traces. Specifically, this code allows for flexible weighting of the

different harmonic contributions $S_0(t,\omega)$, $S_1(t,\omega)$, and $S_2(t,\omega)$. Given that the second-harmonic trace $S_2(t,\omega)$ is already strongly corrupted by noise, it has proven most successful to put the strongest weight on retrieval of $S_0(t,\omega)$ and to ignore $S_2(t,\omega)$ altogether in the reconstruction process. Figure 2 shows the results of this procedure. Despite of identical input pulses and an identical beam splitting scheme, the retrieved pulse shape $E(t)$ is significantly longer when TiO_2 is used as the nonlinear medium. For deeper analysis we deconvolved the retrieved TiO_2 traces with a single-sided exponential kernel $k(t)=\exp(-t/\tau)$, $t \geq 0$, forcing the identity $E_{TiO2}(t)=\int E_{SiO2}(t')\,k(t-t')\,dt'$ in an iterative procedure. The result of this analysis are shown in Fig. 2, indicating a polarization decay time τ=6.5 fs in TiO_2.

In addition to the experiments, we theoretically investigated this surprising outcome by numerically solving the TDSE. We used the numerical formalism originally proposed in [4] with periodic boundary conditions, employing the quasipotential for silica proposed in [5]. For titania, we adapted the quasipotential to correctly model band gap and effective mass of the material. The result of these calculations is the rapidly oscillating polarization $P(t)$, from which we extract the phase $\varphi_\omega(t)$ at the fundamental frequency ω by Fourier analysis. Additionally, we extract the amplitude envelope of the third harmonic contribution $P_{3\omega}(t)$. For the case of fused silica, this envelope follows exactly the third harmonic of the driver field, i.e., $E_3(t)$. In agreement with the experimental results, however, we see delayed relaxation $P_{3\omega}(t)$ when adjusting the band gap to match TiO_2. Again, we analyzed this marked difference by a deconvolution, which indicates a polarization decay time τ=6.2 fs for peak intensities of 10^{12} W/cm^2 in TiO_2. Appearance of a non-vanishing decay time at the third harmonic is linked to similar effects in $\varphi_\omega(t)$, i.e., the self-phase modulation also experiences a non-instantaneous polarization decay. However, both these effects only appear when the third harmonic of the driver frequency is in resonance with the quantum mechanical dipole, i.e., $3\hbar\omega \geq E_{gap}$.

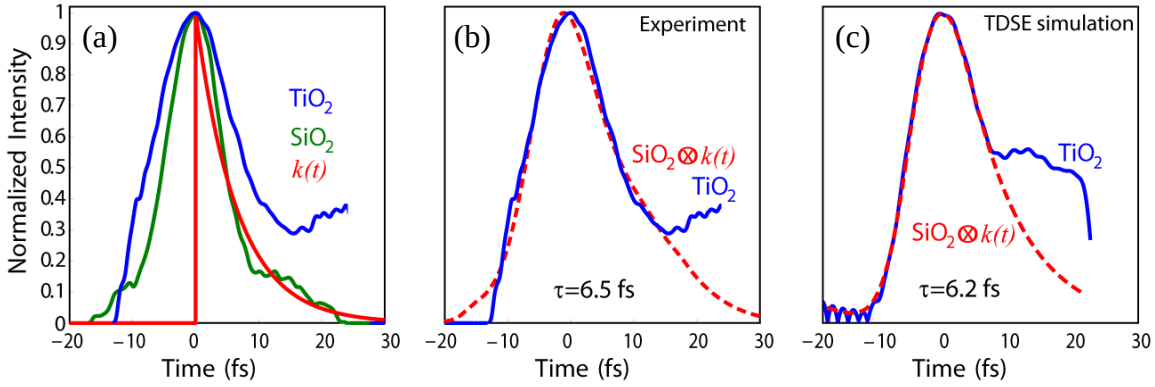

Fig. 2 (a). Retrieved intensity envelopes from the measured FROG traces. Blue: titania. Green: silica. Red: kernel function employed in deconvolution. **(b)** Titania envelope in comparison with convolved silica envelope. Time constant was chosen to match the response near zero delay. The lasting component appears to stem from free carriers. **(c)** Same as in (b), but for numerical results of the TDSE.

As a result of the non-instantaneous polarization decay, restrictions may apply to Kerr-lens mode-locking or other mode-locking mechanisms that are based on a Kerr-based self-amplitude modulator. While we are not aware of a report on operation of a KLM laser above $E_{gap}/3$, it seems impossible, for example, to exploit the KLM principle to a few-cycle dye laser in the visible. Moreover, $\chi^{(3)}$ based FROG or correlation techniques will inevitably record a longer pulse duration in the ultraviolet above $E_{gap}/3$. For nonlinear materials like silica, its practical use would therefore be restricted to wavelengths above 500 nm. Using LiF as the solid with the widest band gap, an absolute lower limit for any given $\chi^{(3)}$ FROG technique appears at 320 nm. We therefore believe that our findings may have important consequences for both, the generation and the characterization of few-cycle pulses. Moreover, the interferometric FROG method provides an interesting new access to the high-field physics inside solid-state materials.

References

[1] S. K. Das, C. Schwanke, A. Pfuch, W. Seeber, M. Bock, G. Steinmeyer, T. Elsaesser, and R. Grunwald, Opt. Express **19**, 16985 (2011).
[2] G. Stibenz and G. Steinmeyer, Opt. Express **13**, 2617 (2005).
[3] A. Anderson, K. S. Deryckx, X. G. Xu, G. Steinmeyer and M. B. Raschke, Nano Lett. **10**, 2519 (2010).
[4] H. G. Muller, Laser Phys. **9** 138-148 (1999).
[5] T. Paasch-Colberg et al., „Solid-state light-phase detector," Advance Online Publication, Nature Photon. (January 2014).