Sensitive interferometric frequency-resolved optical gating with an EMCCD based spectrometer

S. K. Das, M. Bock, G. Steinmeyer, R. Grunwald, and T. Elsaesser, Max Born Institute, Berlin (Dezember 2013)

Application Note

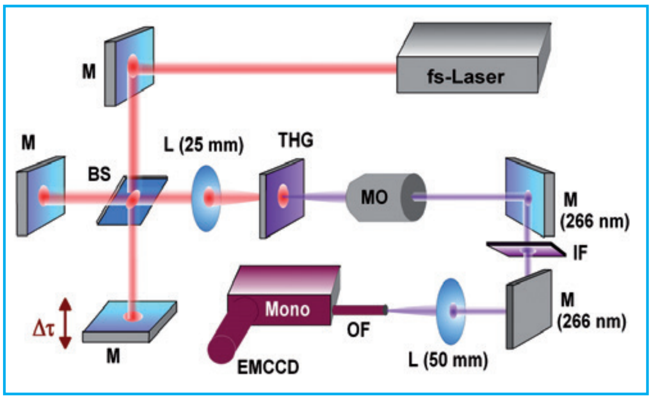
Introduction

The combination of high dynamic range detectors working at quantum efficiencies close to single photon capability with high-resolution spectroscopic devices opens new prospects for nonlinear optics. In particular, the sensitive characterization of the time-dependent response of materials induced by ultrashort laser pulses can be studied. Appropriate systems for the detection of such interactions are enabling tools for an improved understanding of basic excitation mechanisms as well as for the diagnostics of pulsed optical signals at extremely low intensities.

To characterize a temporal asymmetry in pulse analysis, third order autocorrelation functions have to be measured, e.g. by third harmonic generation (THG) [1]. To reduce phase matching problems, layers of sub-wavelength thickness are advantageous. In turn, because of the low excited volume and cubic intensity dependence of the THG efficiency, measurements with unamplified pulses are difficult. We report on recent experiments at Max Born Institute for Nonlinear Optics and Short-Pulse Spectroscopy (MBI) in Berlin with interferometric frequency-resolved optical gating (iFROG), which represents a particular kind of spectrally resolved nonlinear autocorrelation [2]. To this end, surface-enhanced THG of femtosecond laser pulses in sputtered nanocrystalline TiO_2 nanofilms was studied. Third-order autocorrelation and interferometric frequency-resolved optical gating (iFROG) traces were observed with nanojoule pulses emitted from a femtosecond laser oscillator [3]. The results help to tailor nanomaterials with large $\chi^{(3)}$ for next-generation optoelectronics.

Experimental setup and measuring strategy

The characterization of nonlinear material properties was performed with a combination of a Michelson interferometer, a UV-spectrograph and a UV-sensitized EMCCD detector. A fs-laser beam was tightly focused onto the surface of the samples.



Experimental setup for third order interferometric FROG measurements (M = mirrors, BS = beam splitter, EMCCD = single photon detector, mono = grating monochromator, OF = optical fiber, L = lenses or parabolic mirrors, MO = microscope objective, THG = nanomaterial for third harmonic generation).

The investigations were done in two steps. In a first set of experiments, the spectral dependent THG efficiency of different nanolayer structures was measured in comparison to standard surfaces, and the most efficient layers were selected. In a second set of experiments, the capability of these high efficiency nanolayers was evaluated to serve as low dispersion frequency converters in an iFROG configuration.

The FROG concept adds the spectral dimension to the temporal axis of an autocorrelation. A high (attosecond range) temporal resolution can be obtained by implementing the time delay by shifting one arm of an interferometer (collinear iFROG). This approach is particularly suited for diagnosing few-cycle pulses. The phase retrieval from the spectrally resolved interference plot requires the application of iterative mathematical procedures and enables additional consistency checks. The work reported in [3] was restricted on Fourier filtering the dc contribution and was processed with a commercial software.

Experimental details

Laser:

- Ti:sapphire laser oscillator (Femtosource), pulse duration 15 fs, center wavelength 790 nm, pulse energy 4 nJ, repetition rate 75.3 MHz

Spectrograph:

- Shamrock SR-303i-A (Andor Technology)
- Blaze gratings (1200 l/mm, 600 l/mm, 150 l/mm)

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Detector:
- EMCCD DU970N-UVB (Andor Technology)
- UV conversion layer
- cooling -75 °C
- 1600 x 200 Pixels, 16 x 16 µm²
- quantum efficiency >0.3 (UV)
- 1,515 spectra per s
- read out noise 2.8 e⁻ (<1 e⁻ in electron multiplying mode)
- dark current 0.00007 e⁻/pixel/s

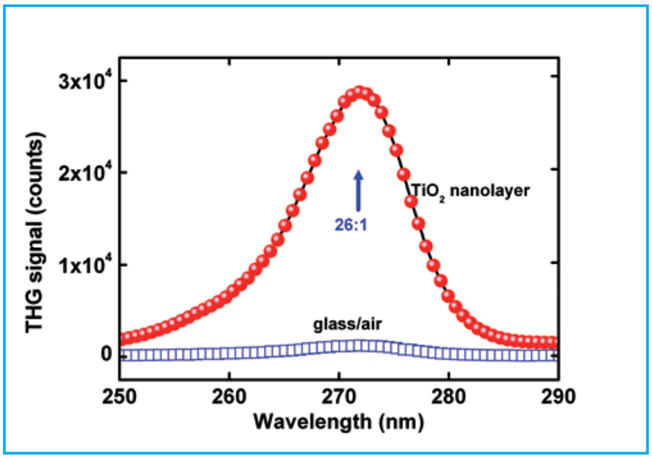
Phase retrieval software:
- FROG 3.0 (Femtosoft Technologies)

Experimental results

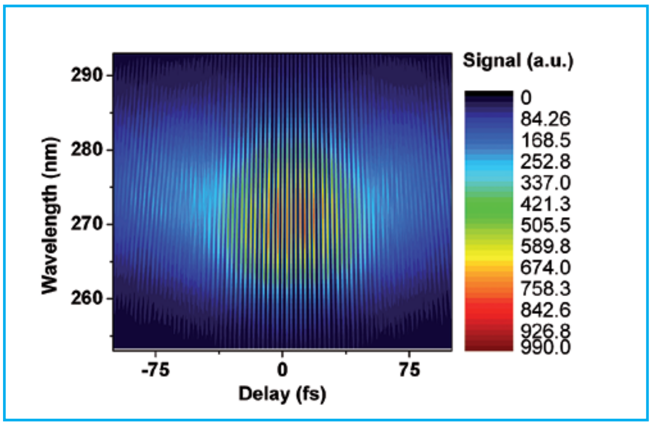
TiO_2 nanolayers with thicknesses between 100 and 400 nm and different morphology were first characterized by SEM, XRD and via detecting the THG. Maximum THG signals were found at 180 nm thickness. Third-harmonic generation of near-infrared ultrashort pulses in sputtered TiO_2 nanolayers was studied. Optimized samples of 180 nm thickness provided 26 times higher THG efficiency than the air-glass interface. The nonlinear susceptibility was estimated to be $\chi^{(3)} = 7.1 \times 10^{-11}$ esu (in SI units: 9.94×10^{-19} m^2V^{-2}) and thus significantly exceeds reference values for 1064 nm and 532 nm. The enhanced THG efficiency enabled, for the first time, to characterize broadband nanojoule ultrashort pulses with third-order interferometric FROG in a compact experimental arrangement.

The retrieved spectral phase corresponds to a pulse duration of 22 fs, indicating a parasitic positive quadratic chirp induced by optical components (beam splitter).

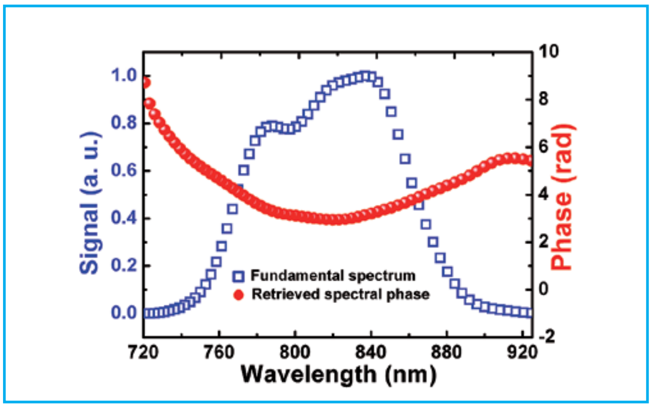
Recently, similar iFROG studies were performed with ZnO nanorods at even shorter pulse durations (6 fs) [4].



THG signal for the maximum efficiency TiO_2 nanolayer (thickness 180 nm), after [3].



Measured iFROG signal for the maximum efficiency nanolayer (thickness 180 nm), after [3].



Retrieved spectral phase and fundamental spectrum for the maximum efficiency TiO_2 layer, after [3].

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References

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Contact

Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy im Forschungsverbund Berlin e.V.
Dept. C2
Dr. Rüdiger Grunwald
Max-Born-Str. 2a
12489 Berlin
Germany

e-mail grunwald@mbi-berlin.de
phone +49 30 6392-1457
www.mbi-berlin.de