Investigation of the effect of ultrashort pulse spectral phase modulation on the two-photon absorption processes

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Abstract

In this work we present the findings obtained on the investigation of the effect of ultrashort pulse spectral phase modulation on the two-photon absorption (2PA) process of well-known organic compounds. Using a pulse shaping technique (deformable mirror), we modulated femtosecond ultrashort pulses using two different spectral phase configurations, periodic (sine-cosine) and quadratic (linear chirp) functions, and analyzed the effect of these spectral phase modulations on the 2PA process of the compounds. We observed that in the case of the periodic phase, differences in the 2PA process modulation were observed for each compound, while for the quadratic phase similar modulations were observed.

Samples

The investigation of the effect of ultrashort pulse spectral phase modulation on the two-photon absorption (2PA) process was realized using the organic compounds MEH-PPV, coumarin, fluorescein, BePTCD and AzoPTCD (perylene tetracarboxylic derivatives), Fig. 1. The absorption spectra in the UV-Vis region, obtained with a Cary 17 spectrophotometer, are presented in Fig.2. All organic compounds present two-photon excited fluorescence (2PEF) when excited above the linear absorption wavelength, in the nonresonant region, with a strong laser pulse.

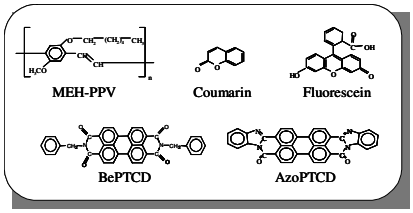

Figure 1: Molecular structures of the investigated compounds.

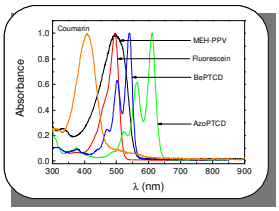

Figure 2: Absorbance spectra of the investigated compounds.

Experimental setup

The experimental scheme used in this work is showed in Fig. 3. The ultrashort laser pulse are produced by a Kerr-lens modelocked Ti:sapphire oscillator from (≈ 5 nJ energy pulse and 80 MHz repetition rate). For this work the laser cavity was adjusted to provide pulses with 40 nm spectral bandwidths (≈25 fs temporal duration) and center at three different wavelengths, 760, 800 and 840 nm. A micromachined deformable mirror (MMDM) [1] was used to shape the phase of the pulse spectral components. The MMDM is a 600 nm gold-coated silicon nitride membrane (8 mm x 30 mm) suspended over an array of 19 actuator electrodes on a printed circuit board. The MMDM is placed at the Fourier plane of a zero-dispersion stretcher consisting of a 600 grove/mm ruled grating and a 25 cm focal-length mirror. The deformation of the MMDM is controlled via computer using a program implemented.

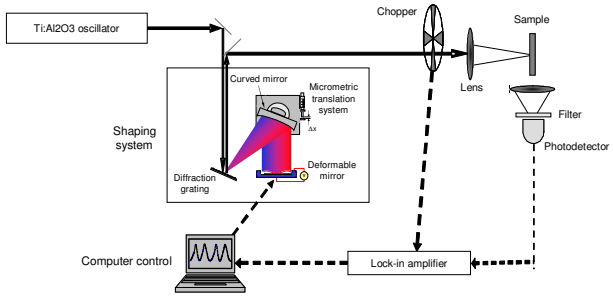

Figure 3: Experimental setup for ultrashort laser pulse shaping by spectral phase modulation and monitoring of the 2PA process.

The phase-modulated pulses are focused into the sample using a convergent lens of 10 cm focal distance. The excitation beam is modulated by means of a mechanical chopper, allowing the nonlinear processes signals to be monitored using a lock-in amplifier. The 2PEF intensity was collected perpendicularly to the cuvette using an optical fiber and an infrared filter to cut the laser beam scattering, and monitored using a photodetector. In the case of the second harmonic generation (SHG), the effect of the pulse spectral phase modulation on the process generated in a KDP crystal (0.7mm-thick) was monitored in the far field using also a photodetector and infrared filter to cut the transmitted laser beam intensity.

Deformation details

Well-known phase configuration: $\Theta(\Omega) = \alpha \cos(\beta\Omega+\delta)$, $\alpha = 2\mu m$, $\beta = 3\pi/\Delta\omega_{pulse} = 3{,}1\ 10^{-14}\ rad^{-1}$

Deformation Scan: 4π $(\delta: 0 \rightarrow 4\pi)$

Two-photon absorption

$$S_2(\Delta\omega) \propto \left| \int_{-\infty}^{+\infty} g(\omega) A(\omega/2-\Omega) A(\omega/2+\Omega) \exp\{i[\Phi(\omega/2-\Omega)+\Phi(\omega/2+\Omega)]\} d\Omega \cdot d\omega \right|^2$$

Results

By scanning a periodic modulation on the deformable mirror, we imposed symmetric and asymmetric phase distributions on the pulse spectral components. Through this method, we were able to analyze the 2PA modulation in organic compounds via the intrapulse quantum interference effect [2,3]. Measurements of the SHG process modulation in a thin KDP crystal was also carried out, and the modulation observed was used as a comparison signal for the measurements in the organic compounds. The results obtained in the study of the 2PA dependence on the periodic phase modulation of femtosecond pulses centered at 800 nm are presented in the Fig. 4. The data were normalized in such a way that the maximum intensity observed was equals to unity. The 2PA modulation efficiency of each compound was also investigated using ultrashort pulses centered at different wavelengths, 760 and 840 nm. The results obtained are gathered in the Figure 5.

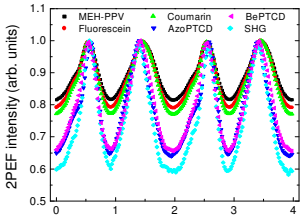

Figure 4: 2PA modulation of the investigated compounds obtained scanning periodic spectral phase modulation on the deformable mirror.

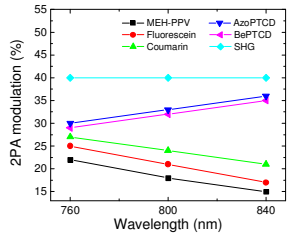

Figure 5: 2PA modulation of each investigated compound using ultrashort pulse in three different spectral conditions (●"40nm).

Using a pulse stretcher, we investigated the behavior of the 2PA and SHG as a function of the amount of linear chirp (quadratic phase configuration) introduced to the ultrashort pulse (Fig. 6). The purpose of these measurements was to confirm that the differences observed in the modulation of the 2PA process of the investigated compounds are actually related to the distinct effect that the symmetric spectral phase configuration makes on the intrapulse quantum interference process in each compound, and it is not just an effect of the intrinsic pulse intensity modulation due to the spectral phase modulation.

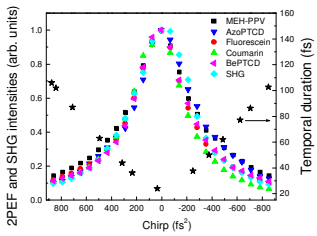

Figure 6: 2PEF and SHG intensities in function of the linear chirp introduced in the ultrashort pulse.

Conclusion

We observed distinct amplitudes of modulation on the 2PA process for each investigated compound, which we believe are related to the detuning between the pulse spectrum and the material absorption band. The effect produced for the detuning of the laser pulse spectral band on the 2PA process control of these organic compounds confirm ours interpretations.

The behavior of the 2PA process of all compounds in function of the introduction of linear chirp on the ultrashort pulse confirmed that the differences observed in the 2PA process modulation of the investigated compounds have their origin on the distinct effect that the symmetric spectral phase configuration makes on the intrapulse quantum interference process in each compound.

Acknowledgement

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