Two-photon absorption optimization in ZnSe crystals

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Abstract

In the present work we have used an adaptive feedback control apparatus, which basically uses femtosecond pulses generated by a Kerr-lens modelocked Ti:Sapphire oscillator, whose spectral phase is shaped by a micro-machined deformable mirror, in order to optimize the two-photon absorption (2PA) process in ZnSe crystals.

Sample

The zinc selenide (ZnSe) crystals are semiconductors materials that present significant absorptive nonlinearity as well as a refractive one. Owing to its nonlinear optical properties, they have been widely studied as potential components of various optical devices. Among the areas of interest are optical switching and optical limiting. The absorption spectrum in the UV-Vis region, obtained with a Cary 17 spectrophotometer, is presented in Fig.1. The ZnSe crystal presents wide transparency range in the Vis-IR region and strong two-photon absorption (2PA) process, with subsequent fluorescent relaxation, when excited above linear absorption wavelength (nonresonant region) with strong laser pulse.

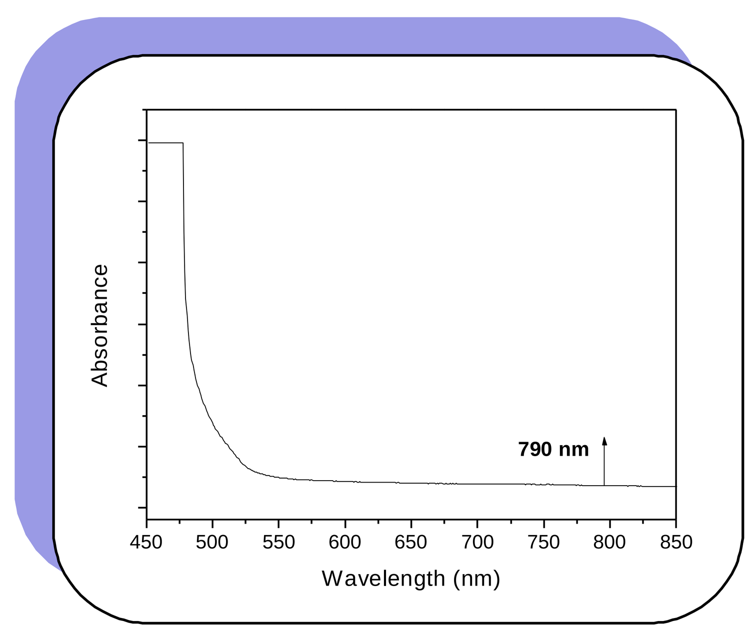

Figure 1: Absorbance spectrum of the zinc selenite crystal.

Experimental setup

In this experiment we have used laser pulses with around 60 nm of bandwidth and 15 fs of pulse duration, centered at 790 nm. The pulses were delivered by a commercial Ti:sapphire Kerr-lens modelocked (KLM) laser oscillator from K&M company, operating with a repetition rate of around 80 MHz. The typical average power used was 400 mW (~5 nJ per pulse).

In this work we have used a micromachined deformable mirror (MMDM) from OKO technologies to pulse shaping in the phase domain. The mirror in 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 maximum deflection is 4 μm with response of 1 ms. Potential applied to the actuator creates an electrostatic attraction between the membrane and the electrode, deforming the mirror surface. The total surface deflection of the mirror is a linear combination of the influence functions for all actuators. Deviation of the mirror surface causes the light to travel a different path, changing the phase of the spectral component in the area of the deformation. 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, Fig.2. In order to control the deformation of the MMDM we have used a GA program implemented in LabVIEW. Such program is very powerful in our case of multiple variable problems.

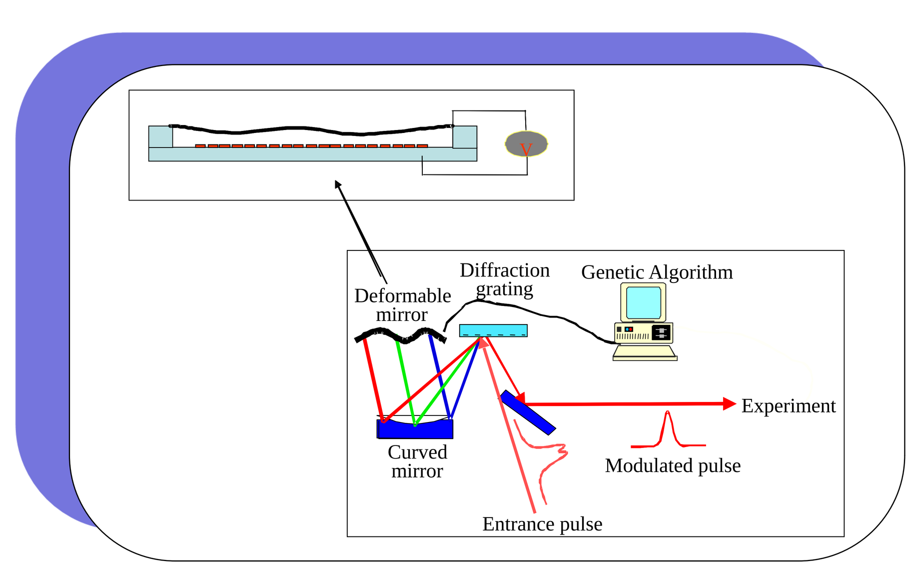

Figure 2: Experimental setup for pulse shaping.

The two-photon absorption optimization is obtained using an evolutionary strategy which begins with a set of random pulse shapes whose associated two-photon absorption effect is measured. Those pulses that produce the most intense two-photon absorption effect are retained, duplicated, perturbed and reproduced, as the GA requires, Fig.3. This process is repeated until a desired number of interactions (generations). Basically, the GA is a search and optimization methodology inspired in the biological evolution, consequently, the biology concepts such as chromosome (father and children), reproduction, crossover, mutation, etc are applied in the computational GA code.

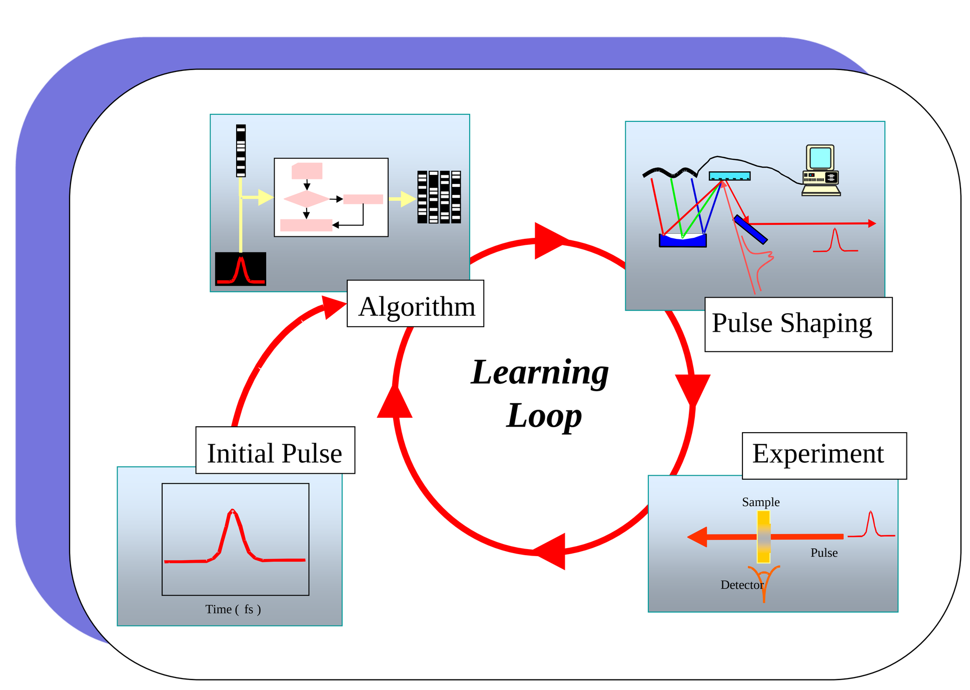

Figure 3: Experimental setup for pulse shaping optimization using GA..

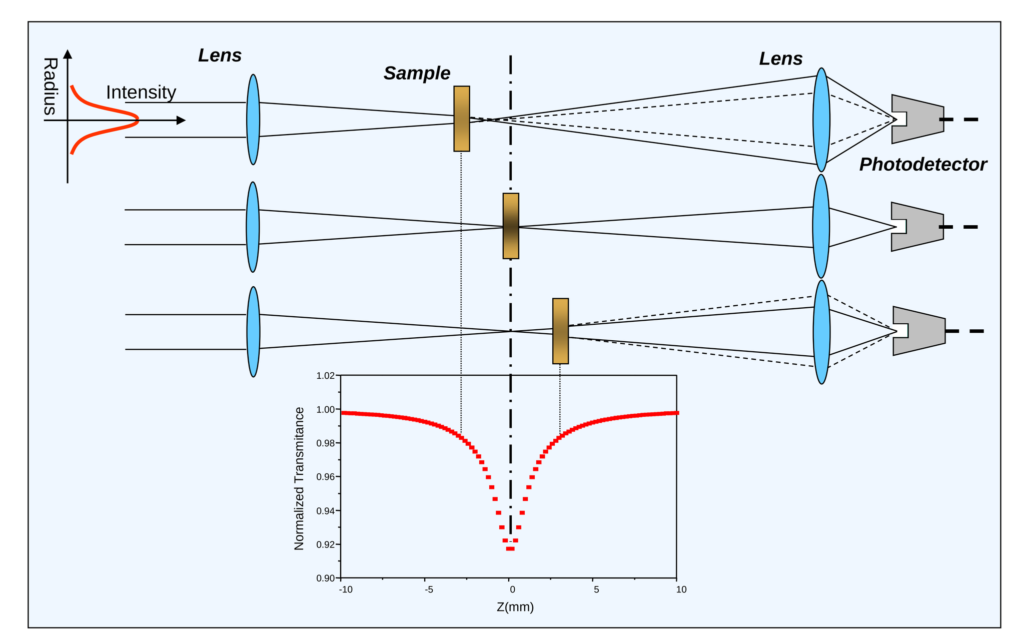

Figure 4: Experimental setup of the absorptive Z-scan technique utilized at the 2PA optimization process in ZnSe crystals.

The adaptive feedback control scheme proposed here, which basically consists of a absorptive Z-scan technique, is shown in Fig. 4. In this experiment, the transmittance of a sample is measured in the far field as the sample is moved along the propagation path (z) of a focused Gaussian beam. The magnitude of the nonlinear absorption can be easily observed from such a transmittance curve (Z-scan). Such Z-scan signature is symmetric with respect to the focus (z=0) where it have a minimum transmittance, in the case of 2PA processes. The sample nonlinear transmittance can be detected and used as the feedback signal in a closed-loop control using the GA.

The 2PA process intensity is detected by monitoring the beam intensity pattern in the far field, by measuring the nonlinear transmittance. This method is similar to the absorptive Z-scan technique, however, here the sample is not scanned but positioned at the focal point of the first lens, once it corresponds to the point where occurs a 2PA peak and hence, a minimum transmittance in the absorptive Z-can signature. This nonlinear transmittance signal is used as the feedback signal in our evolutionary strategy.

Results

Using our pulse shaping setup, 2PA optimization in ZnSe crystals (0.7mm thickness) was achieved through the evolutionary strategy after approximately 30 interactions. The transmitted light was collected by a PIN photodetector with lock-in amplifier. In order to confirm the 2PA optimization we have carried out absorptive Z-scan measurements before and after the optimization processes. We have carried out also FROG (Frequency-Resolved Optical Gating) measurements of the pulse before and after the optimization processes so as to obtain the pulse duration shortening and phase correction occurred. Fig.5 illustrates the evolution of the fitness parameter (2PA magnitude) during the GA optimization process and the absorptive Z-scan and FROG measurements before and after the optimization process in the ZnSe crystal. The duration of the optimization processes was twenty minutes.

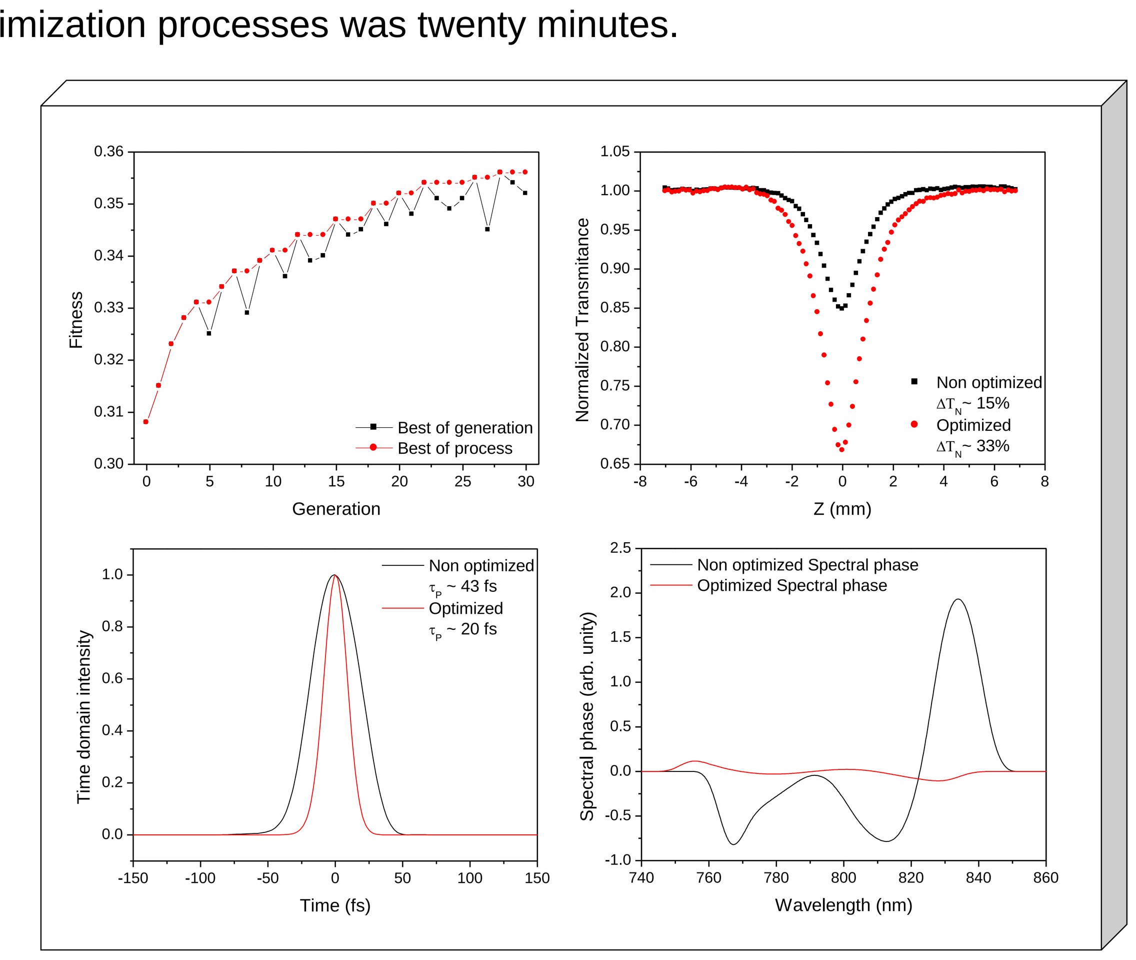

Figure 5: Evolution of the fitness parameter and absorptive Z-scan and FROG measurements before and after the 2PA optimization process.

An increase of approximately 100% was observed in the normalized transmittance change (two-photon absorption) for the ZnSe crystal in the optimized condition when compared to the non optimized one. The pulse characterization obtained with the FROG (Frequency-Resolved Optical Gating) method before and after the optimization reveals that a reasonable pulse shortening from 43 fs to 20 fs and a significant phase correction were obtained.

Conclusion

The results indicate that the observed enhancement in the nonlinear absorption must be due to the achievement of a Fourier transform limited pulse (laser bandwidth ~ 60 nm). In this way, further investigation must be carried out in order to understand if the phase correction accomplished by the evolutionary strategy is really coherent controlling the nonlinear process or just optimizing the pulse duration (pulse chirp).

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