Continuum generation in bulk materials from the deep UV to the infrared with pump pulse durations over the entire femtosecond regime

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Abstract. We demonstrate continuum generation with pulses from the long femtosecond up to the picosecond regime. To understand the mechanism involved in this process we study the influence of the spectral width and the chirp of the pump pulses. We further investigate the threshold for stable continuum generation with smooth, plateau-like spectra from the deep ultraviolet to the infrared. We visualize the processes of the generation and study filament parameter such as length, diameter and observe multiple filamentation. This visualization allows us to determine and understand the temporal jitter of newly generated frequencies.

1 Influence of pump pulse duration on continuum generation in bulk

When ultrashort intense laser pulses are focused in bulk materials, one of the most fascinating phenomena in ultrafast nonlinear optics can occur: supercontinuum generation. The octave wide spectral broadening gives access to wavelength regions not supported by available laser systems. These continua render smooth, flat and gap free spectra with a high amount of coherence, compressibility, and high stability on all time scales [1]. This favors them as a most valuable source for broadband radiation in laser technology and spectroscopy [2]. Continuum generation works well with a pump pulse duration below 150 fs, even with extremely short few cycle pulses. However, it fails with longer pulses in common crystals like sapphire owing to avalanche ionization. Nevertheless continuum generation with longer pulse duration would be of great interest for many applications. It would be particularly advantageous for strong pump lasers of optical parametric chirped pulse amplifiers which have pulse durations in the picosecond regime.

In this contribution we show first proof of ps continuum generation in laser crystals retaining the advantageous properties of fs continua like flat and smooth plateau-like spectra over the entire visible range (see figure 1 (a)). To understand the role of the pulse duration on continuum generation, experiments on the influence of the spectral width and chirp were performed. For this purpose we generate picosecond pulses from a Ti:Sapphire system (Clark MXR, CPA 2001, 775 nm, 150 fs): One time by narrowing the spectrum and one time by up-chirping the pulse with the entire spectrum by transmission through glass. With sufficient pump energy new spectral components can occur for these pulses during propagation in crystals. The stability at selected wavelengths serves as criterion for continuum generation. Figure 1 shows the pump energy for stable continuum generation in sapphire, YAG and KGW, when chirping (b) and spectral narrowing (c) the pump pulse.

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Further studies indicate that especially for longer pulses the spectral width is a limiting factor, because long pulses compress towards the Fourier limits during the propagation in the first few mm of the crystal due to self-steepening. If the spectral width is low, self-phase modulation has to first broaden the pulse to reduce the Fourier limit and than self-steepening can shorten the pulses. This configuration needs more energy and show a stronger broadening, as confirmed by the experiment. In upcoming experiments we will analyze the chirp of the newly generated frequencies and the temporal behavior of the pump region during filamentation.

2 Continuum generation with pump wavelengths from the UV to the IR

Nonlinear processes like optical parametric amplification, second harmonic generation, and sum or difference frequency generation allow the generation of energetic pulses from the deep UV to the infrared [3] largely independent from the original laser wavelength. However, these pulses often have a limited spectral bandwidth originating from the necessary phase-matching of the nonlinear processes. The generation of broadband spectra around these new frequencies is a challenging task and often requires high energies. Continuum generation in bulk materials on the other hand only needs µJ energies and therefore spectral broadening is possible. The choice of the crystals additionally allows to reduce the necessary energy or to increase the spectral width. To show that continuum generation is not only limited to small regions around the pump wavelength, we show continua generated with frequency doubled Ti:sapphire and Yb:KYW lasers, the original wavelength, and infrared pulses from an optical parametric amplifier. Figure 2 shows selected continua covering the ultraviolet, visible and infrared. The pump wavelength is blocked with filters, so that only the newly generated frequencies are shown.

Fig. 1. Continuum generation in selected crystals with picosecond pulses (a), chirped (b) and spectrally narrowed (c) femtosecond pulses.

Fig. 2. Continuum generation with pump wavelengths from the UV to the IR (400, 800 and 1600 nm).

All these continua show a smooth, gap free and wide spectrum with a proper beam profile and a high stability. This favors them for the use in femtosecond pump probe spectroscopy. The spectral overlapping allows combining the results from different continua. The short wavelength continuum
cut-off remains constant if the pump wavelength is increasing. This allows octave-wide gap free flat continua when pumping with an infrared source. Combining the principles of optical parametric amplification (OPA) with the fact that the energy threshold is low, these infrared sources can easily be generated by nearly any laser systems. For OPCPA systems passive carrier-envelope phase (CEP) stabilization can be obtained in this conversion.

3 Visual observation of continuum generation in YAG

During filamentation and continuum generation many nonlinear processes take place like self-focusing, self-steepening, self-phase modulation, or multi-photon excitation. In Yttrium aluminum garnet (YAG) not only the continuum can be observed as beam profile along the propagation axis, but also the filament channel is clearly visible from the side. This is presumably due to the recombination of the electrons from the conduction to the valence band. By imaging the visible filament on a camera we can investigate the position and length of the filament. The filament length of up to 2 mm is independent of the incidental numerical aperture and can therewith be significantly larger than the Rayleigh length. This is due to the balance between self-focusing and plasma defocusing which overcomes the natural divergence. Also filamentation starts closer to the entrance surface of the crystal, if the input energy is increased. This is due to the stronger self-focusing, what leads to a shorter self-focal distance.

From the side view of the filamentation also the temporal jitter of the newly generated frequencies can be analyzed and optimized to 0.5 fs/nJ. The jitter comes from the fluctuation of the filament position which originates from the energy fluctuations of the pump. This is for example important for passively CEP stabilized pulses from an OPA [4]. If the energy for continuum generation is increased and exceeds twice the threshold for continuum generation we observe a second filament arising. These filaments line up on the propagation axis and show spatial and spectral interference. Figure 3 shows this spatial (a) and spectral (b) interference, and the side view of two filaments in a YAG crystal (c).

![Fig. 3. Spatial (a) and spectral (b) interference and side view (c) of two filaments in YAG](image)

The field of continuum generation in bulk materials is still under investigation and offers many rewarding opportunities for advanced laser applications and broadband spectroscopy. Continuum generation with multi-ps pump pulses is within reach, as confirmed by our first successful experiments. The ease of handling and the very compact setup qualify continuum generation as excellent alternative broadband source for many applications.

References