Direct Generation of 7 fs Whitelight Pulses from Bulk Sapphire

Emanuel Wittmann, Maximilian Bradler, and Eberhard Riedle
LS für BioMolekulare Optik, Oettingenstr. 67, 80538 München, Germany
e.wittmann@physik.lmu.de

Abstract: Generation of sub-10 fs continuum pulses without external compression is demonstrated. We investigate the propagation of the newly generated wavelengths and find that a short crystal in combination with an achromatic telescope leads to nearly chirp free continua.
OCIS codes: (320.6629) Supercontinuum generation; (190.4720) Optical nonlinearities of condensed matter

1. New insights into bulk filamentation

Supercontinuum generation (SCG) in bulk material is a generally applicable method to broaden the spectrum of femtosecond laser pulses at various wavelengths. The Fourier limit for a possible compression of, e.g., a 800 nm pumped continuum from sapphire, amounts to about 4 fs. Yet, no results have been published which show that bulk continua have intrinsically such short pulse durations. This is in striking contrast to the situation in continua generated in gas-filled hollow core fibers or in gas filamentation. There compression to below 4 fs has been shown.

In precise investigations of the continuum generation and propagation we now find that the inability to compress the continuum stems from the highly wavelength dependent effective generation locus and propagation. This knowledge gives us the chance for ideal control of the process and therefore the ability to generate sub-10 fs pulses without the use of any external compression scheme. This validates that the new frequencies generated during filamentation develop highly coherently.

2. Propagation properties of new frequencies and generation of sub-10-fs pulses during SCG

Two processes should be differentiated when the over-all appearance of continuum generation in a bulk material is considered. First, the spatial area or depth into the material where the new colors are developing has to be considered. Second, the propagation in the remaining material before exiting into free space has to be understood. That these issues are far from trivial is proven by the fact that a full collimation of a bulk continuum has not been reported and consequently the full temporal compression has not yet been achieved.

In preliminary experiments we imaged the continuum from a 3-mm sapphire with a singlet lens and found that the blue part of the spectrum focuses earlier than the red part. In a semi-quantitative interpretation this could be attributed to the chromatic error. To circumvent this issue, we used a *Schiefspiegler* [1] that images all spectral components without chromatic error. We still found the blue spectral components focused earlier. An explanation would be that during filamentation all colors are generated at once, but short wavelengths fall behind the filament channel because of their lower group velocity. Without guiding by the filament, they start to diverge. The wavelengths close to the pump follow the channel longer and diverge later, leading to the observed color dependent propagation.

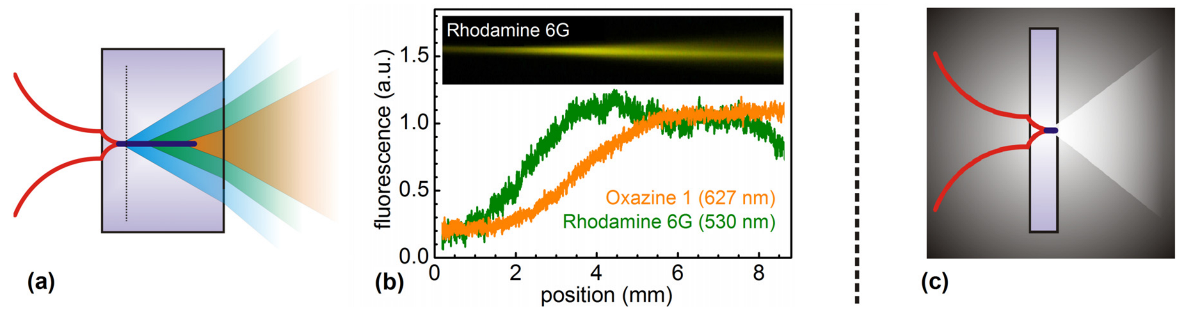

Fig. 1. a) Scheme of bulk continuum generation and wavelength dependent beam propagation. b) Fluorescence of Rhodamine 6G (green) and Oxazine 1 (orange) to monitor the beam propagation during filamentation in ethanol. c) All colors diverge equally in a short crystal.

To verify this concept, a continuum is generated in ethanol and monitored from the side with a high resolution camera. To visualize the propagation of selected wavelength ranges we use a solvent (instead of sapphire) and Rhodamine 6G (absorbing around 530 nm) or Oxazine 1 (627 nm) were added separately. These laser dyes partly absorb the newly generated light and fluoresce so that the beam propagation can be monitored from the side. Fig. 1 (b) shows the corresponding fluorescence signal and the side view from filamentation in ethanol with Rhodamine 6G (top). A strong signal is only found when the light has already broadened from the 8 µm filament and the dye absorption is not saturated as inside the channel. We find that different colors start to diverge at different positions in the solvent. Our proposed model is confirmed and we can explain the difficulty of properly imaging a continuum.

To avoid the wavelength selective propagation, the continuum has to be generated at the very end of the crystal. The crystal should be terminated at the dashed line in Fig. 1 (a) so that no spectral and local separation occurs and all colors start to diverge simultaneously as shown in Fig. 1 (c). We find a short crystal length on the order of 1 mm suitable. This should lead to a chirp free continuum as the newly generated colors pass through no extra material, and all colors having the same spatial properties.

With a 1 mm sapphire plate, we optimize the continuum generation onto the output face. A careful alignment still renders a continuum with little fluctuations. The pump source is a small fraction of the output of a Ti:sapphire amplifier (CPA 2001; Clark MXR) with a pulse duration of 170 fs and a central wavelength of 778 nm, focused with a f = 50 mm plano-convex lens. We obtain a continuum spanning down to 430 nm. For an anastigmatic and achromatic collimation or imaging of the generated continuum we use a reflective *Schiefspiegler* telescope [1], consisting of a suitable combination of a convex and a concave mirror. This allows us to measure the pulse duration of the newly generated frequencies with a SHG intensity autocorrelator without the loss of any frequencies due to aberrations. The main peak of the autocorrelation signal corresponds to a 7 fs continuum pulse.

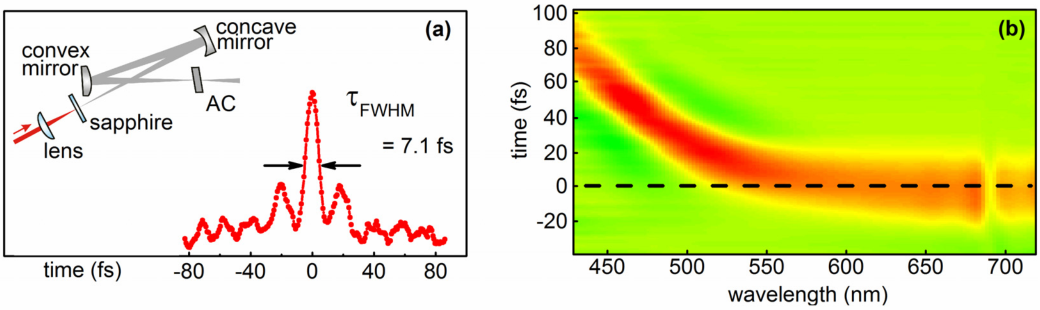

Fig. 2. a) *Schiefspiegler* geometry for imaging the newly generated colors of a 1-mm sapphire continuum and autocorrelation trace demonstrating a 7 fs pulse. b) Transient absorption measurement in 200 µm GG400 to determine the chirp of the continuum generated in a 1-mm sapphire plate.

For a better insight into the spectro-temporal distribution of the continuum and to show that the autocorrelation signal does not correspond to a coherence spike [2] due to the complexity of the continuum pulse, the chirp of the sapphire continuum is determined with our transient spectrometer [3]. Fig. 2 (b) shows the transient signal of the continuum generated in the 1-mm sapphire plate. With a 200 nJ, 25 fs pump pulse at 470 nm and the sapphire continuum as probe we measured the cross correlation via two-photon absorption in a 200 µm GG400 (Schott AG) substrate. Two-photon absorption of pump and probe only occurs for temporal overlap and allows determining the group delay for every wavelength. Since there are only reflective optics in the probe beam path, the signal represents the intrinsic spectral chirp of the continuum after propagation through just a short length of air. Fig. 2 (b) shows that all spectral components from 500 to 700 nm coincide in time. The chirp for the short wavelength range mainly originates from the propagation in air. These chirped components of the pulse appear as broad and structured wings in the autocorrelation trace.

3. Conclusion & Outlook

It is possible to obtain sub-10 fs white light pulses directly from bulk filamentation. In the literature only a few examples for white light compression can be found, but until now the appropriate setup always is accompanied by huge complexity. By studying the propagation properties of the continuum we found a straightforward way to simplify the effort for generation of short continuum pulses. An elaborate apparatus can be replaced by a lens, a 1-mm sapphire plate and the adequate adjustment for the incident pulse energy.

With the imaging by a *Schiefspiegler* also any chromatic aberrations as well as astigmatism can be avoided. Such pulses are highly interesting for broadband amplification, ultrafast 2D spectroscopy, or spectroscopic experiments.

[1] A. Kutter, *Der Schiefspiegler: Ein Spiegelteleskop für hohe Bilddefinition* (Weichhardt, 1953)

[2] Rick Trebino, *Frequency-Resolved Optical Gating: The measurement of Ultrashort Laser Pulses*, (Kluwer Academic Publishers, Boston / Dordrecht / London, 2000), Chap. 4

[3] U. Megerle, I. Pugliesi, C. Schriever, C.F. Sailer, E. Riedle, "Sub-50 fs broadband absorption spectroscopy with tunable excitation: putting the analysis of ultrafast molecular dynamics on solid ground", Appl.Phys.B **96**, 215-231 (2009)