Pulse non-linear post-compression with tunable wavelength by balancing SPM and SRS

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Abstract: In non-linear pulse spectral broadening via SPM, the original central wavelength is preserved; with SRS, it is red-shifted by an amount related to the broadening. We show that, by balancing SPM and SRS, red-shift and broadening can be tuned independently.

For the last 30 years, NIR, femtosecond laser pulses with increasing energy (from sub-mJ to the 100-mJ level) have been routinely post-compressed by non-linear spectral broadening via self-phase modulation (SPM) in noble gases and subsequent management of the spectral phase. This includes the compensation of the positive chirp of the broadened pulses by adding a controlled, discrete amount of negative group delay dispersion (GDD) with chirped mirrors, and eventually a continuously tunable amount of positive GDD with wedges.

The spectral broadening $\Delta\omega \propto \int dl/A \cdot P \cdot k_2 . p/\lambda_0^2$, where dl is the length of the interaction, A the section area, P is the pulse peak power, λ_0 is the central wavelength, and k_2 is the ratio between the nonlinear index coefficient and the gas pressure p [1]. The spectral broadening $\Delta\omega$ cannot be extended by arbitrarily increasing the pulse intensity nor the gas pressure, in order to avoid ionization and to operate below the critical power of self-focusing. The solution has been to increase the length of laser-gas non-linear interaction at controlled levels of laser intensity and pressure: this is achieved in hollow-core fibers (HCF) [2], where a relatively large (tens to hundreds of μm) focal spot size of the laser is preserved by the guiding effect of the capillary over long distances (from several cm to few meters), and for the last few years, also in multi-pass cells (MPC) [3], where instead the laser beam is mildly focused several (tens of) times between two curved mirrors, typically in a Herriot configuration. With either type of setup, the spectral broadening obtained via SPM in noble gases is symmetric around the central laser frequency, which is therefore preserved and constant regardless of the amount of spectral broadening.

In 2020, few groups [4] demonstrated that, with hollow-core fibers filled with molecular instead of noble gases, the spectral broadening is strongly asymmetric towards longer wavelength, thus resulting in a net red-shift of the broadened spectrum: similarly to the case of SPM, the pulses can be also compressed to similar durations, with comparable efficiencies and potentially with the very same experimental setup. As an example of a possible application, in [5], the pulses from an Yb:CaF_2 regenerative amplifier (1030 nm, 250 fs, up to 14 mJ) were compressed to ~20 fs either via SPM or stimulated Raman scattering (SRS), and used for high harmonic generation (HHG). The cut-off photon energy, which scales with λ^2 driver, could be extended from 220 eV for driving pulses broadened by SPM to 290 eV for pulses broadened and shifted by SRS.

The amount of spectral broadening can be easily tuned for both SPM and SRS, within certain limits, by tuning the gas pressure. The central wavelength though is fixed in the case of SPM, and dependent on the gas pressure in the case of SRS, but through the amount of spectral broadening: i.e. it is not possible to tune independently the central wavelength and the spectral broadening with SRS only. It is however quite straightforward that, in a controlled mixture of noble (for SPM) and molecular (for SRS) gas, the total broadening can be controlled by the total pressure, while the central wavelength can be tuned by the ratio between the gases.

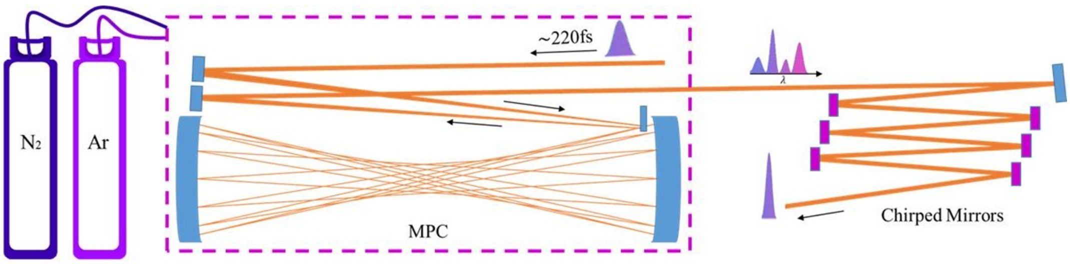

Fig. 1. Schematic of the setup. Left: The beam is coupled into a MPC for spectral broadening in a chamber filled with gas. Right: set of chirped mirrors for post compression.

In our experiment, we used a 59 cm MPC (fig. 1.) wherein the laser beam gets reflected by two r=-300 mm mirrors with 20 bounces. Meantime, by placing Argon and mixing with Nitrogen, it was observed that the symmetric spectrum acquired from SPM has become intermediate with SRS. Fig. 2.a shows experimentally spectrum obtained from spectral broadening of 1 mJ, 220 fs pulses for SPM (700 mbar Ar), SRS (260 mbar N_2) and gas mixture of 200 and 260 mbar N_2 added to 700 mbar Ar. We saw that by changing the mixture ratio we could be able to change the relative intensity of blue and red part of the broadened pulses, therefore changing the central wavelength of the compressed pulse.

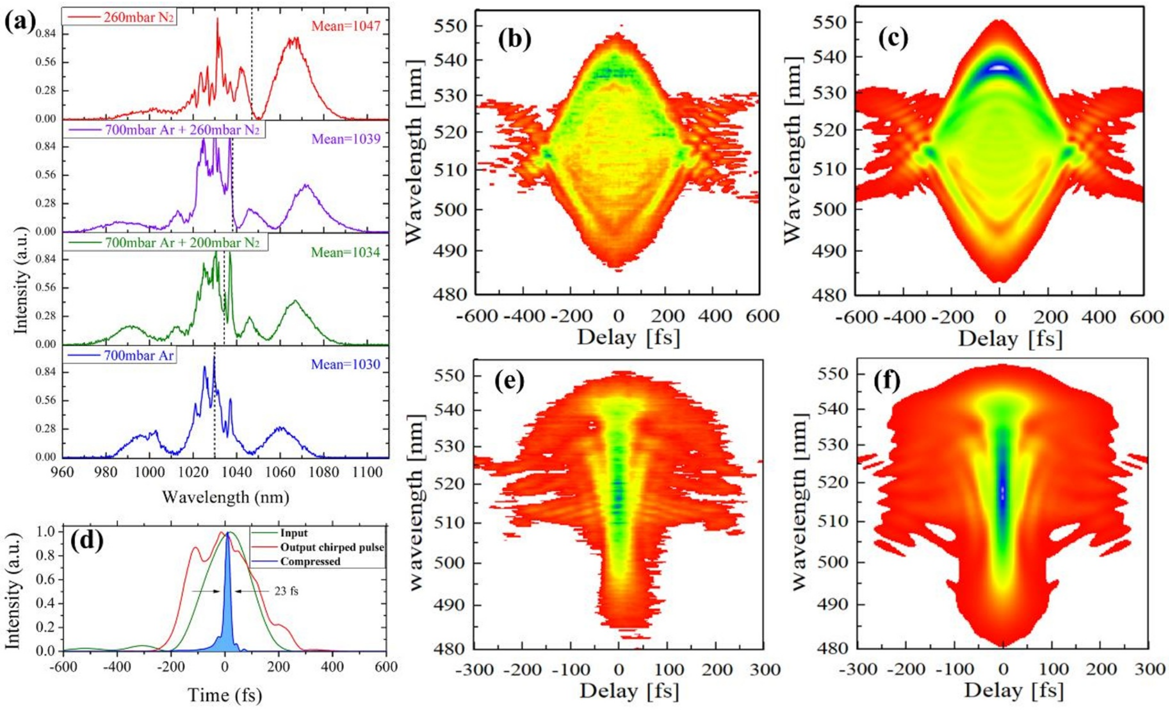

Fig. 2. (a) experimentally obtained spectral broadening of 1 mJ, 220 fs pulses for SRS only (red), gas mixture with 260 mbar (violet) and 200 mbar (green) N_2 added to 700 mbar Ar, and SPM only (blue), (b) SHG-FROG traces of the pulse at the output of the MPC with the gas mixture of 700 mbar Ar and 200 mbar N_2, (c) retrieved from the experimental FROG traces after MPC, (d) temporal envelopes of the input (green), output of the MPC (red), and compressed (blue) pulses retrieved from the experimental FROG, measured (e), and retrieved (f) SHG-FROG of the compressed pulses after chirped mirrors.

Fig. 2.b & c display the SHG-FROG traces and retrieval for the broadened pulses with the gas mixture of 700 mbar Ar and 200 mbar N_2. In principle we could compress SRS only, SPM only, therefore we managed to do it also with the gas mixture. In fig. 2.d the temporal properties of the input, output pulses has been plotted which indicates the compressed pulses down to 23 fs. As it is shown in measured FROG (fig. 2.e) and retrieval (fig. 2.f) from compressed pulses after set of chirped mirrors, the weighted mean value of the wavelength changed to 1041 nm for this specific gas mixture comparing to SPM and SRS, and in the future we plan to do it also in HCF.

References

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