Dual-beam multipass cell compression for time-resolved femtosecond spectroscopy setups

Alan Omar[*]**, Boldizsar Kassai, Martin Hoffmann, Clara J. Saraceno**
Photonics and Ultrafast Laser Science, Ruhr-Universität Bochum, Universitätsstrasse 150, 44801 Bochum, Germany
*alan.omar@ruhr-uni-bochum.de

Abstract: We present a dual-beam single multi-pass cell compressor designed for applications in time-resolved pump-probe spectroscopy. A single MPC simultaneously compresses two beams with different pulse energies to the same pulse duration, allowing for independent modulation of the input beams. The two beams have different, high pulse energies (82 µJ and 296 µJ) and are compressed to the same pulse duration of 50 fs with high efficiency.

1. Introduction:

High-average power ultrafast Yb-based laser systems are increasingly being deployed in ultrafast science [1], thanks to the widespread availability of multi-pass cell MPC nonlinear compression setups, that enable a wide variety of pulse energies with high efficiency to be compressed to well below sub-30 fs. In particular, increasing the repetition rate of pump-probe time-resolved experiments is increasingly being sought after for shorter measurement times. In many of these setups, both pump and probe pulses require short pulse durations, and the two beams need to be modulated at different, high frequencies to provide best signal-to-noise ratio which is challenging to achieve with high peak power beams (after compression) – and is ideally done with the native moderate peak power of the laser for both beams. One approach is to build two multi-pass cell compressors for each beam, which is cumbersome and leads to path length differences that need to be compensated, increasing complexity. In this work, we present a compact and cost-efficient approach for nonlinear pulse compression of two high-pulse-energy beams in a single hybrid air/bulk MPC, designed for future ultrafast pump-probe experiments. We spectrally broadened via self-phase modulation and compressed simultaneously two beams with 82 µJ and 296 µJ pulse energy to 50 fs in one single MPC in ambient atmosphere with transmission efficiency of 88% and 92% respectively, at a repetition rate of 100 kHz. Such a setup significantly simplifies the implementation of MPCs in ultrafast spectroscopy setups at high repetition rates.

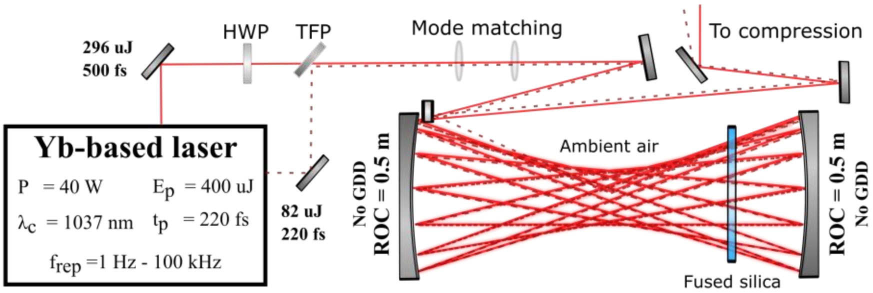

Fig. 1. Experimental setup. The seed laser is a commercial laser amplifier, and the Herriott cell mirrors are HRs with negligible GDD and separated by 94 cm, providing 38 passes through fused silica and ambient air. Both beams were combined using a TFP and then compressed after the MPC with two different dispersion setups at full power, and both were guided for characterization using a wedged sampler.

2. Experimental setup and results:

The seed laser is a commercially available Yb-based laser system (Carbide, Light Conversion) that provides an average power of 40 W and a repetition rate of 100 kHz, corresponding to a maximum pulse energy of 400 µJ. The laser output beam was divided internally into two beams, with two different pulse stretchers, enabling the pulse duration to be varied from 220 fs to a few picoseconds. The unchirped arm has a pulse energy of 82 µJ and a pulse duration of 220 fs, while the second arm has a pulse energy of 296 µJ, and the pulse duration is stretched to 500 fs to decrease its peak power. Both arms have two independent pulse pickers to vary the repetition rate from a single pulse to 100 kHz. Both beams were combined using a thin film polarizer (TFP), then passing the same mode-matching optics. Both beams enter the MPC in a closed-path configuration via a rectangular mirror. The MPC is carefully optimized using 3D pulse propagation simulations to balance the contribution of the nonlinear media (consisting of air and a fused silica plate) and maximize the MPC performance for both pulse energies simultaneously, achieving a spectral broadening of both beams supporting a Fourier transform limit (FTL) of sub-50 fs. The MPC consists of two 2-inch highly reflective (HR) mirrors with a radius of curvature (ROC) of 0.5 m each with close to zero group delay dispersion (GDD). This MPC is based on our previous approach of hybrid gas/bulk MPCs, which showed excellent spatio-temporal properties [3]. The mirrors were separated by 94 cm, providing 19 roundtrips corresponding to 38 passes through the nonlinear media. A 1-mm thick anti-reflection (AR) coated fused silica (FS) plate is placed 15 cm from one mirror in ambient air. Both media contribute significantly to the total nonlinearity. Both beams were collected using a 2-inch HR mirror and collimated separately. To separate them, the initial angle of the 82-uJ beam reflection on the TFP is slightly tuned off from the initial angle of the 296-µJ beam, leading to an elliptical spots pattern on the mirrors for the 82-uJ beam. The spectral broadening of the 82-µJ pulse after the MPC corresponds to an FTL of 48 fs, while the output spectrum of the 296-µJ pulse has an FTL of 41 fs. After collimation, both beams are compressed using two separate compressor arms. The 82-µJ arm is compressed with an overall GDD of -3000 fs^2, while the 296-µJ arm has an overall GDD of -3900 fs^2. More GDD was needed for this arm due to the pre-chirp of the 296-uJ pulses.

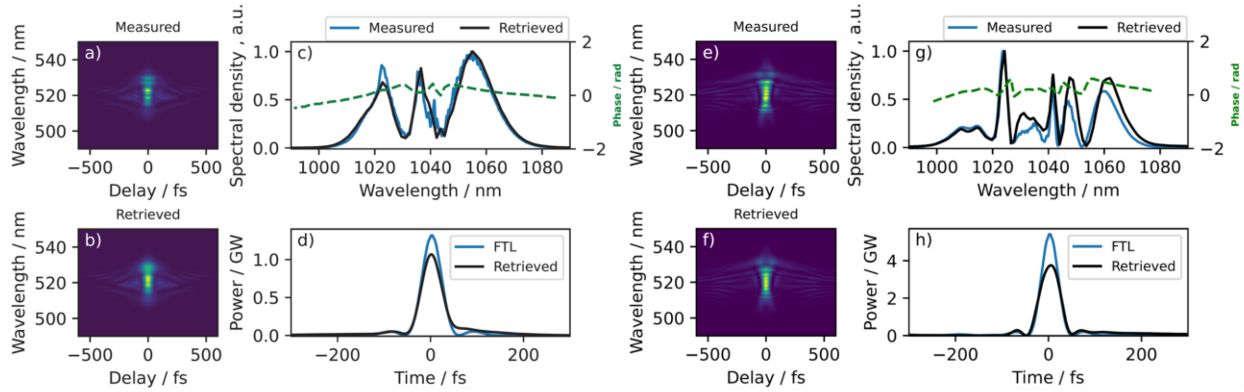

Fig. 2. Characterization of the compressed pulses of both beams after the compression setup. (a) to (d) are the results of 82-µJ arm pulses. a) Measured FROG trace. b) Retrieved FROG trace on a grid of 512 × 512 and an error of <0.45%. c) Measured and retrieved spectra and the phase. d) The retrieved temporal pulse with FTL of the measured spectrum. (e) and (f) are the results of 296-µJ arm pulses. e) Measured FROG trace. f) Retrieved FROG trace on a grid of 512 × 512 and an error of <0.7%. g) Measured and retrieved spectra and the phase. h) The retrieved temporal pulse with FTL of the measured spectrum.

We characterized the compressed pulses of both beams using a home-built second-harmonic frequency-resolved optical gating (SH-FROG). The results in Fig. 2(a) to 2(d) are the results of the 82-µJ arm pulses. The measured and reconstructed traces in Fig. 2(a) and 2(b) are in good agreement. Fig. 2(c) shows the measured spectrum after the MPC, the reconstructed spectrum, the spectral phase, and both spectra show excellent agreement. The compressed pulse intensity profile (Fig. 2(d)) is close to the FTL with a peak power of 1 GW and a pulse duration of 50 fs. The results in Fig. 2 (e) to (h) are the results of the 296-µJ arm pulses. The measured and reconstructed traces in Fig. 2(e) and 2(f) are in good agreement, Fig. 2(g) depicts the measured, the reconstructed spectrum and the retrieved spectral phase. Fig. 2(h) shows the compressed pulse intensity profile of 296-µJ arm with a peak power of 3.8 GW and a pulse duration of 51 fs; further compression of these pulses to the FTL and enhancing the peak power was limited by the GDD profile of the available dispersive mirrors at the time of the experiment. This work proposes a robust and simple approach for simultaneously compressing two different pulse energies in a single MPC in air atmosphere, adapted for experiments in pump-probe spectroscopy. We compressed the pulses of two beam arms of a commercially available, 40 W, Yb-based laser with pulse energies of 82 µJ and 296 µJ pulses to 50 fs and optical transmission efficiency of 88% and 92%, respectively. We believe such a simple design will facilitate the implementation of Yb-based lasers for ultrafast time-resolved spectroscopy.

3. References:

[1] P. M. Donaldson *et al*., “Breaking Barriers in Ultrafast Spectroscopy and Imaging Using 100 kHz Amplified Yb-Laser Systems,” *Acc. Chem. Res.*, vol. 56, no. 15, pp. 2062–2071, Aug. 2023.
[2] J. Schulte, T. Sartorius, J. Weitenberg, A. Vernaleken, and P. Russbueldt, “Nonlinear pulse compression in a multi-pass cell,” *Opt. Lett.*, vol. 41, no. 19, Art. no. 19, Oct. 2016.
[3] A. Omar, M. Hoffmann, G. Galle, F. Sylla, and C. J. Saraceno, “Hybrid air-bulk multi-pass cell compressor for high pulse energies with full spatio-temporal characterization,” *Opt. Express*, vol. 32, no. 8, pp. 13235–13248, Apr. 2024.