Enhanced Nonlinear Spectral Broadening in Multi-Pass Cells Using Molecular Gases

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A nonlinear spectral broadening technique based on the phenomenon of self-phase modulation (SPM) using a waveguide-like approach, namely the multi-pass cells (MPC), has been shown to have excellent performance in numerous experiments [1]. One of the main benefits of such a geometry is simple and flexible dispersion management within the cell. Bulk or noble gases are typically used as nonlinear media for spectral broadening. Recently, molecular gases have been proposed as an alternative to noble gases in hollow-core fibers for enhanced and efficient spectral broadening [3,4]. In this work, we experimentally investigate the use of molecular gases in multipass cells and perform its direct comparison with the nonlinear spectral broadening in the noble gas. Such a realization promises to serve as a power-scalable, extremely broadband, simple and tunable source of ultrashort pulses. Such a source can facilitate many applications in ultrafast spectroscopy and microscopy [3, 4].

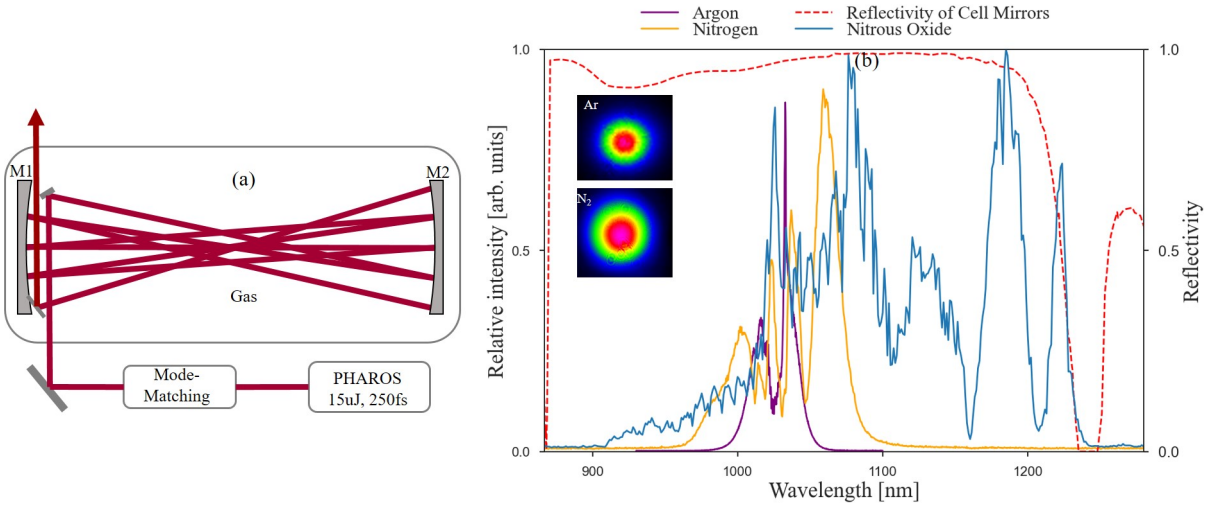

Fig. 1 (a) Schematic of a concave-concave multi-pass cell (MPC). (b) The output spectrum of Ar, N_2 at pulse energy of 15µJ, a repetition rate of 100 kHz and for N_2O at pulse energy of 15µJ and 10kHz repetition rate **inset**: output beam profile for Ar and N_2 .

We designed a MPC setup consisting of two concave mirrors with a 100 mm radius of curvature (ROC) introducing overall negative dispersion of -11.4 fs2 per roundtrip. The cell mirrors were separated by a distance of 130 mm along the optical axis. The eigenmode inside the MPC has a 1.3 mm and 0.20 mm $1/e^2$ radius on the mirrors M1 and M2, respectively. A commercial laser (Light conversion PHAROS) with 15 µJ pulse energy and 250 fs pulse duration was used as a driver for the experiment. The input beam was mode-matched to the eigenmode of the MPC using a single curved mirror with 1000 mm radius of curvature. Input pulses experienced 34 passes through the MPC. The MPC was placed in a gas-filled housing filled with 15.5 bars of Argon (Ar), Nitrogen (N_2), and Nitrous Oxide (N_2O). The overall transmission for Argon and Nitrogen was over 80 %, with clean output beam profile. However, the transmission dropped to less than 50% for N_2O.The output spectra were measured with a high resolution spectrometer (waveScan APE) (Fig 1(b)), resulting in a spectral span of 128 nm and 204 nm for Argon and Nitrogen, respectively. The broadening in N_2 and N_2O shows a strong red shift, Raman contribution, and is limited by the mirrors' bandwidth.

Conclusively, we demonstrated that it is possible to efficiently perform extreme spectral broadening in molecular gases by exploiting their effective rotational and vibrational nonlinearity. Nonlinear absorption and gas turbulences seem to be the main problems in operating such systems at high repetition rates and average power. Systematic investigation of these instabilities is a work in progress.

References

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