Laser filamentation-based nonlinear optics for near-single-cycle spectroscopy

J. H. Odhner[1,2], D. A. Romanov[2,3], E. T. McCole Dlugosz[1,2], and R. J. Levis[1,2]

[1] Department of Chemistry, Temple University, Philadelphia, Pennsylvania 19122

[2] Center for Advanced Photonics Research, Temple University, Philadelphia, Pennsylvania 19122

[3] Department of Physics, Temple University, Philadelphia, Pennsylvania 19122

Corresponding author: rjlevis@temple.edu

Abstract

We present and discuss theory, measurements, and applications of laser filament-based optical spectroscopy. Direct measurements of filamentation in air remain challenging, motivating several methods for *in situ* investigation of filamentation dynamics. Impulsive stimulated Raman scattering measurements can be used to predict the pulse dynamics. Vibrational and time-frequency-mapped rotational Raman spectroscopy can additionally be used as tools for applications in IED detection, radioactive materials detection, and atmospheric sensing. Finally, gas phase filamentation provides a bright, low dispersion seed source for advanced laser technologies.

Filamentation Dynamics

Laser filamentation in the gas phase has been generally understood for nearly twenty years under the paradigm of the dynamic spatial replenishment model [1], but experimental investigations of filamentation dynamics, particularly in air, remains challenging due to the high intensities, extremely short pulses, and broad spectral bandwidths formed in the filament channel during propagation. Sophisticated theoretical studies have predicted many interesting propagation effects in multi-color filaments and shaped beam filamentation. Providing new methods of rigorously studying the dynamics of filamentation *in situ* is critical in order to fully understand how implementations of proposed schemes compare to the predictions.

In Situ Filament Pulse Characterization

The characterization of the dynamics of a femtosecond laser pulse undergoing filamentation has been accomplished by implementing a transient grating cross-correlation frequency-resolved optical gating (TG-XFROG) scheme. This scheme allows for non-invasive investigation of the pulse dynamics, and demonstrates the validity of pulse reshaping models directly.

Filamentation White-Light Spectroscopy

A femtosecond laser pulse will impulsively excite vibrational and/or rotational wavepackets in a molecular medium. Rotational coherences are characterized by periodic revivals of the wavepacket that are often spaced tens of picoseconds apart for large molecules. The broad white-light continuum generated during filamentation makes it possible to map the rotational dynamics into frequency space by chirping the pulse, enabling single-shot measurement of the dynamics over a 100-ps temporal window. This technique has potential applications in detection and control, allowing rapid detection and identification of the rotational structure of molecules in the beam path.

A challenge in the theoretical description of chirped-filament rotational spectroscopy is properly handling a pulse whose Fourier bandwidth limit is a few femtoseconds, but whose chirped temporal duration is 100 ps. We have developed a theoretical description of this situation that avoids the breakdown of the slowly varying envelope approximation, making the correct treatment of the phenomena possible.

Filament-seeded Visible NOPA

Finally, gas phase filamentation offers a source of bright, low dispersion continuum radiation that can be amplified by optical parametric amplification for applications in high field physics or multi-dimensional spectroscopy. Sub-4-fs visible pulses with energies in the several hundred micro-Joule range have been achieved using non-collinear optical parametric amplification with a newly developed method for handling pulse front tilt for short pump pulses (Figure 1).

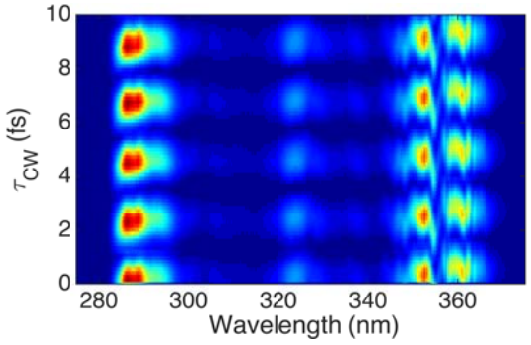

Fig. 1. 2DSI trace of an amplified filament pulse with a FWHM duration of 3.5-fs.

References

[1] M. Mlejnek et al., *Opt. Lett.* **23**, 382-384 (1998).