Remote sensing of nuclear materials using ultrafast laser filamentation in air

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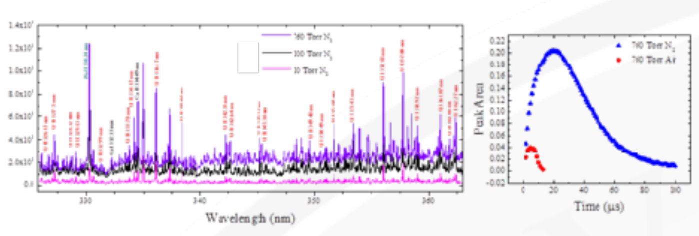
The use of high-power ultrafast laser pulses enables remote interrogation of solids, liquids, or gases on the scale of O(1 km) by employing the phenomenon of filamentation. Since such laser pulses produce high intensities, they give rise to significant third-order nonlinear optical effects in all media, including air. This phenomenon enables the delivery of a laser pulse to a small spot over considerable distances. Characteristic atomic, ionic, isotopic, or molecular signatures may be induced by such filaments and detected via optical spectroscopic techniques such as laser-induced breakdown spectroscopy (LIBS). We discuss our recent work in which we optimized the interaction between ultrafast laser filaments and solid targets to maximize the LIBS signal. We further discuss the experimental measurements of the scaling of characteristic atomic and ionic signal intensity with laser energy. Understanding this scaling is important to predict the efficacy with which excitation can be produced over long distances, when more complex effects such as multiple filamentation arise. Finally, we discuss our progress in understanding the practical aspects and constraints of collection of emission produced by filaments in high-background, long-standoff scenarios.

Laser-based sensing offers unique capabilities for fast, remote detection of any element, molecule, or even isotope

Standard ns-LIBS limited in standoff delivery of excitation source by the large aperture of the optical system



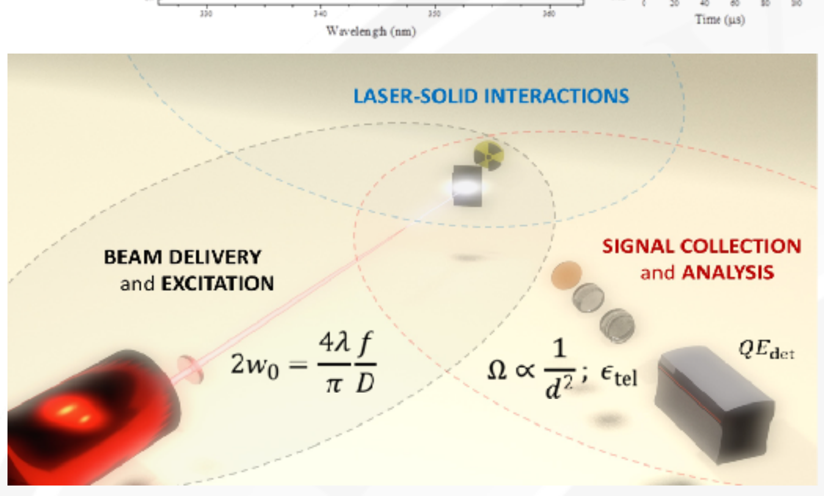
Laser-produced optical signals carry spatial, spectral, and temporal signatures

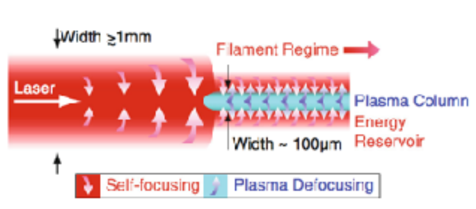
plasma defocusing

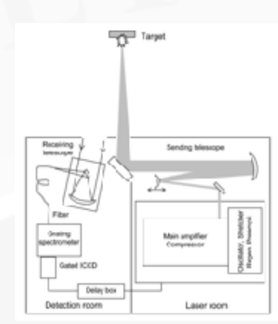
$$n_2 I = \frac{\omega_p^2}{2\omega^2} + \frac{1.22\lambda_0^2}{8\pi n_0 \omega_0^2}$$

Kerr self-focusing

diffraction



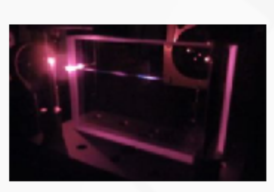




Ph. Rohwetter *et al.*, *Spectrochim. Acta B*, Vol. 60 (2005)

A. Valenzuela *et al.*, *App. Phys. B*, Vol. 116 (2014)

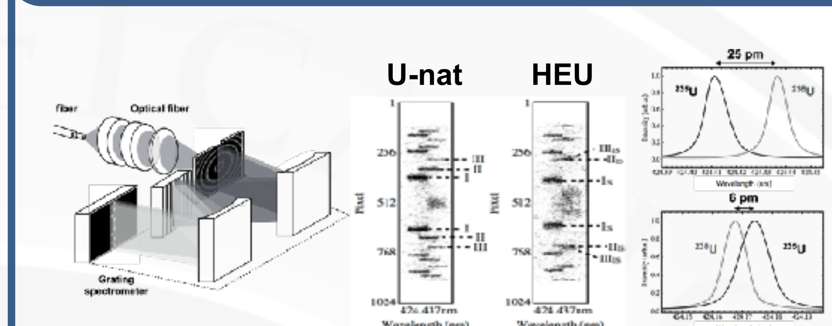
fs-filamentation allows for the extended propagation of laser pulses to distances $\mathcal{O}(10^2 - 10^3\ \text{m})$



Introduces questions regarding interaction of filament with target and standoff collection and detection

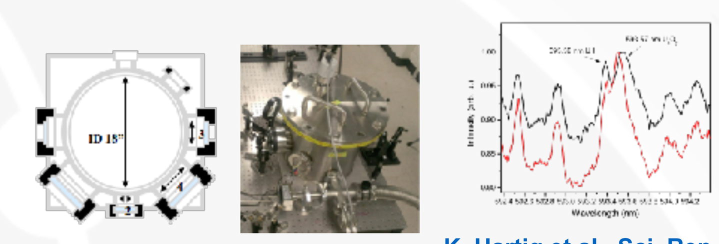
Optical emission spectroscopy (OES) and laser-induced luminescence (LIL) allow for the interrogation of various targets

Optical emission spectroscopy



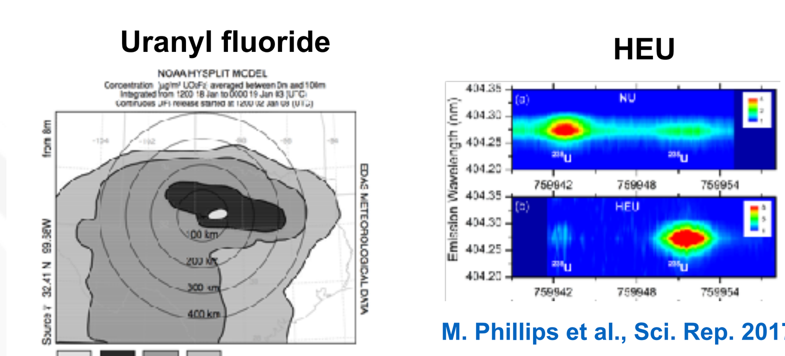
P. Ko, J. Scott, and I. Jovanovic, Opt. Commun.: 357, 95-99 (2015).

P. K. Morgan, J. R. Scott, and I. Jovanovic, SCAB: 116, 58-62 (2016).



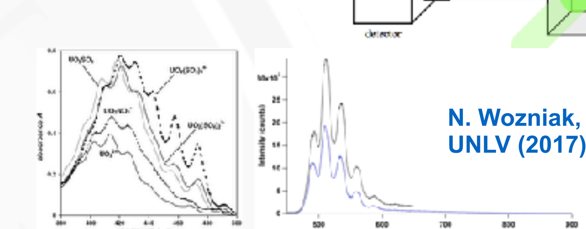
K. Hartig et al., Sci. Rep. 2017

Laser-induced luminescence



M. Phillips et al., Sci. Rep. 2017

R. S. Kemp, Sci. Glob. Sec., Vol. 16 (2008)



N. Wozniak, *Dissertation*, UNLV (2017)

Filamentation allows for the standoff delivery of a laser excitation source

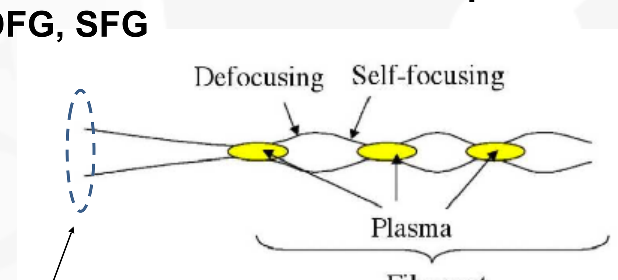
$$P = \varepsilon_0\left(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \cdots\right)$$

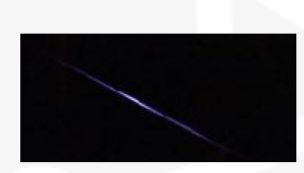
linear optics

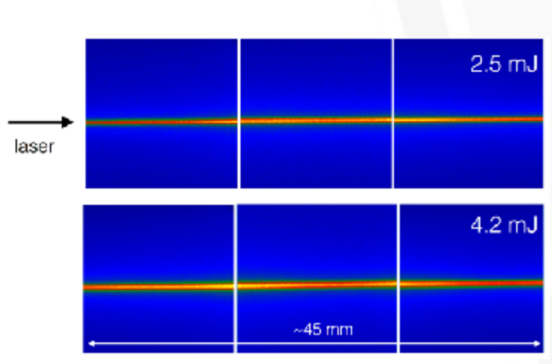
SHG
DFG, SFG

Kerr effect
self-phase modulation (SPM)

Third-order susceptibility gives rise to intensity-dependent refractive index change $\Delta n = n_2 I$



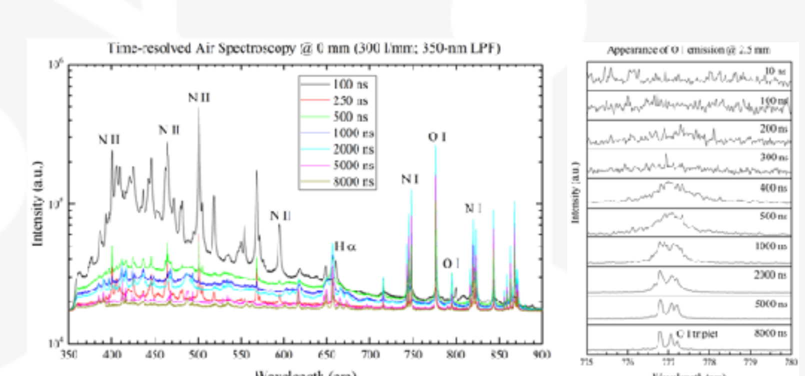




Refractive index change forms a *Kerr lens* which focuses the laser pulse

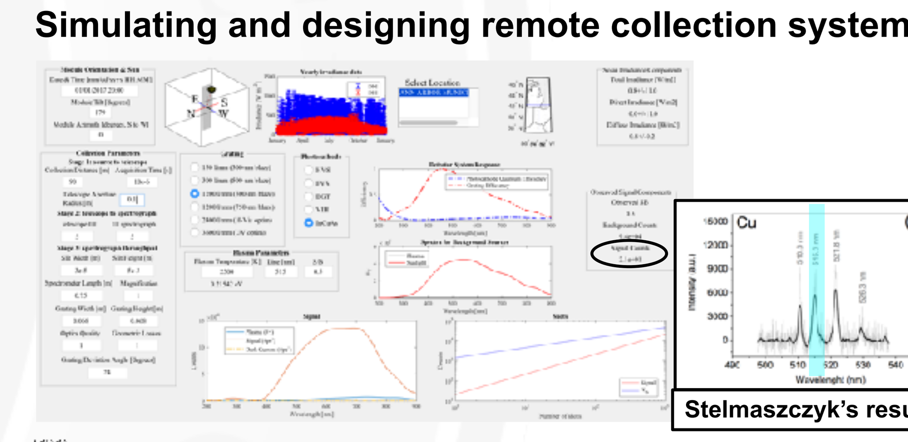
High laser pulse intensity ionizes air and forms a plasma column – *filament*

We study the spatially-, spectrally-, and time-dependent fingerprints in optical signals generated by laser excitation



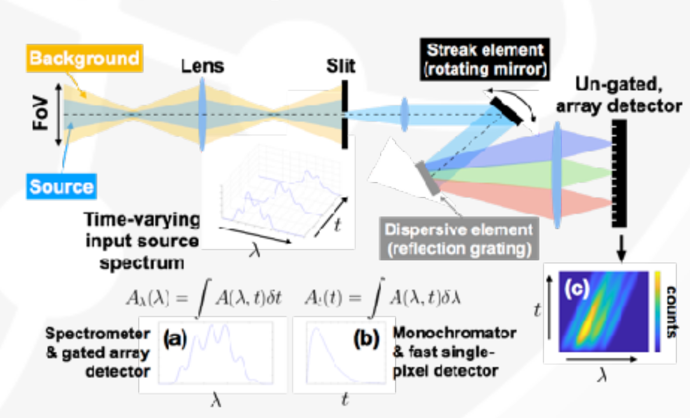
Breakdown spectra carry unique temporal signatures

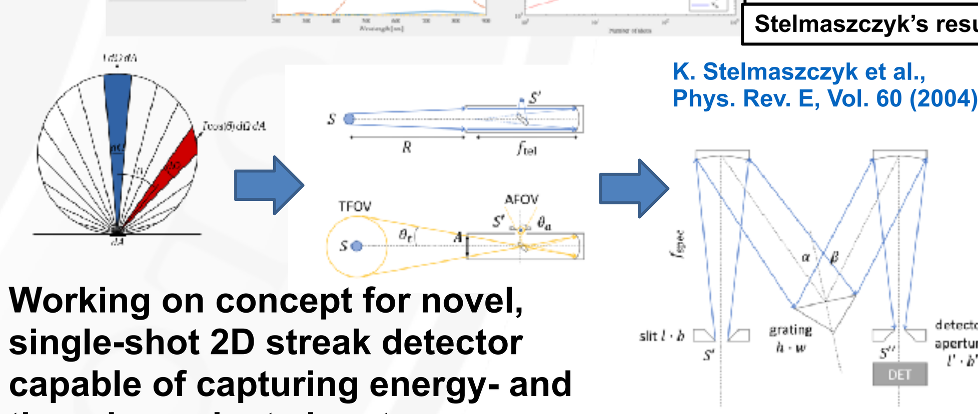
Simulating and designing remote collection systems



Stelmaszczyk's result

K. Stelmaszczyk et al., Phys. Rev. E, Vol. 60 (2004)





Working on concept for novel, single-shot 2D streak detector capable of capturing energy- and time-dependent signatures

We study energy loss mechanisms in the filament in order to optimize propagation distance

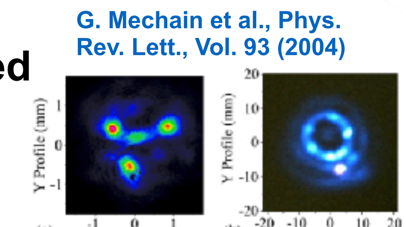
Filamentation is a threshold phenomenon limited by self-focusing then ionization intensity

$$P_{cr} = \frac{0.148\lambda^2}{n n_2}$$

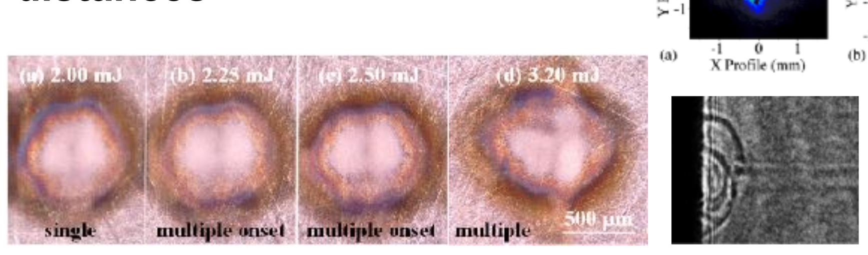
Wavelength λ [nm]	Photon Energy [eV]	Critical Power P_{cr} [GW]
400	3.1	0.45
800 (nominal)	1.6	3.1
2050	0.6	10

High laser energies are required for propagation to longer distances

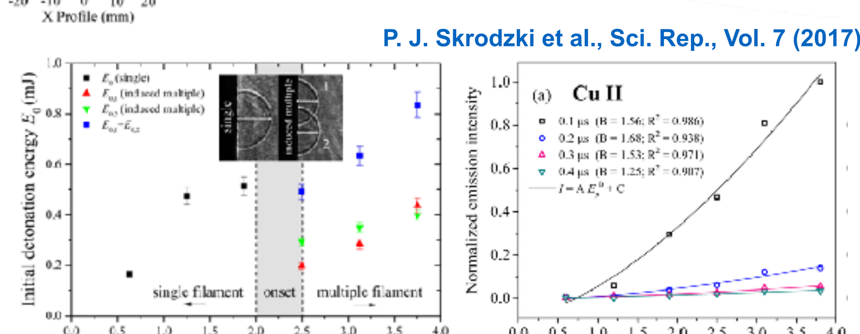
G. Mechain et al., Phys. Rev. Lett., Vol. 93 (2004)



May improve propagation distance by varying laser wavelength



P. J. Skrodzki et al., Sci. Rep., Vol. 7 (2017)

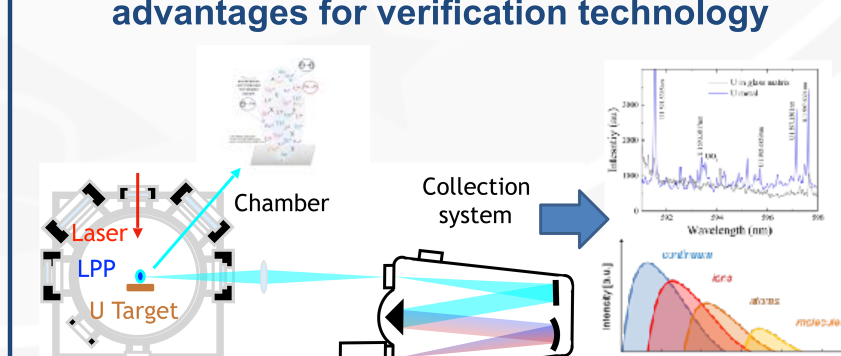


But stochastic multiple filamentation dominates at greater laser peak powers

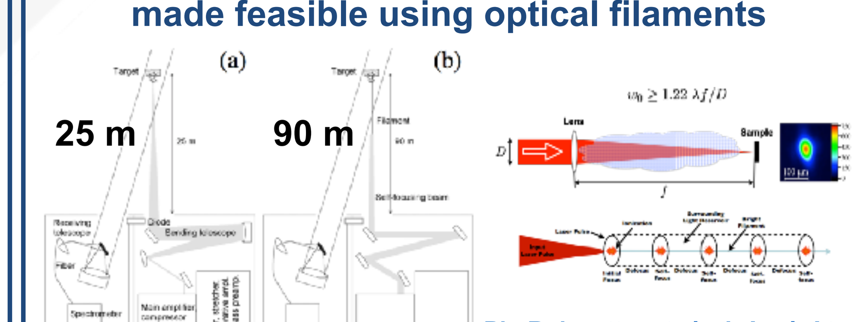
Detected spectroscopic signal continues to increase despite multiple filamentation!

Summary

Laser-based methods offer unique advantages for verification technology

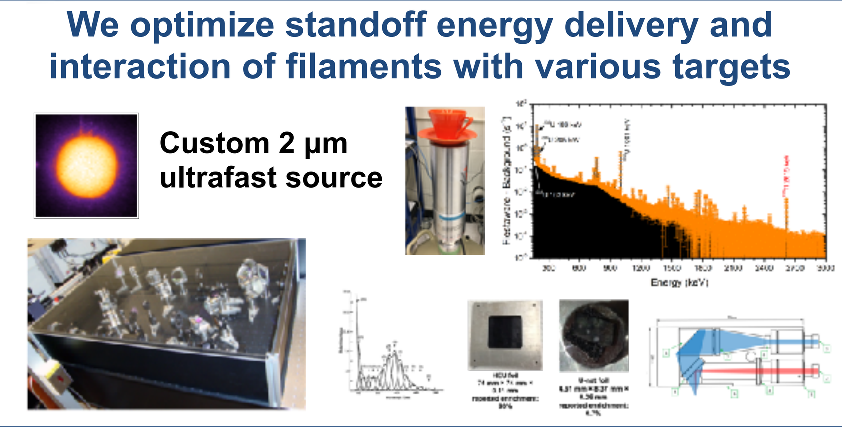


Remote delivery of a laser excitation source is made feasible using optical filaments

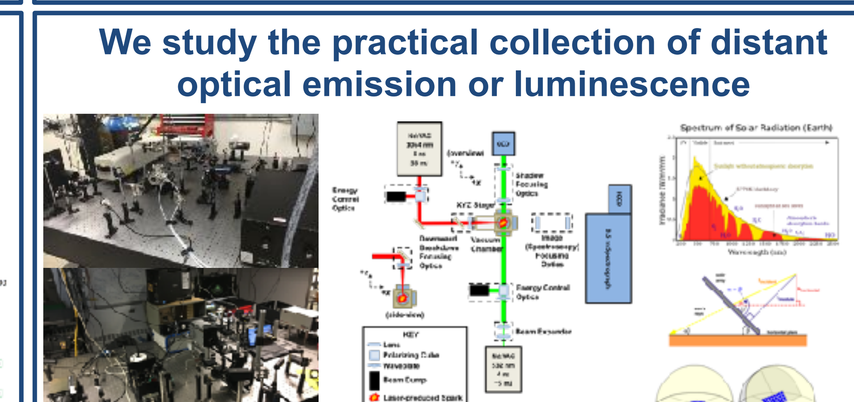


Ph. Rohwetter et al., J. Anal. At. Spectrom., Proc. SPIE 6158 (2004)

We optimize standoff energy delivery and interaction of filaments with various targets



We study the practical collection of distant optical emission or luminescence



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