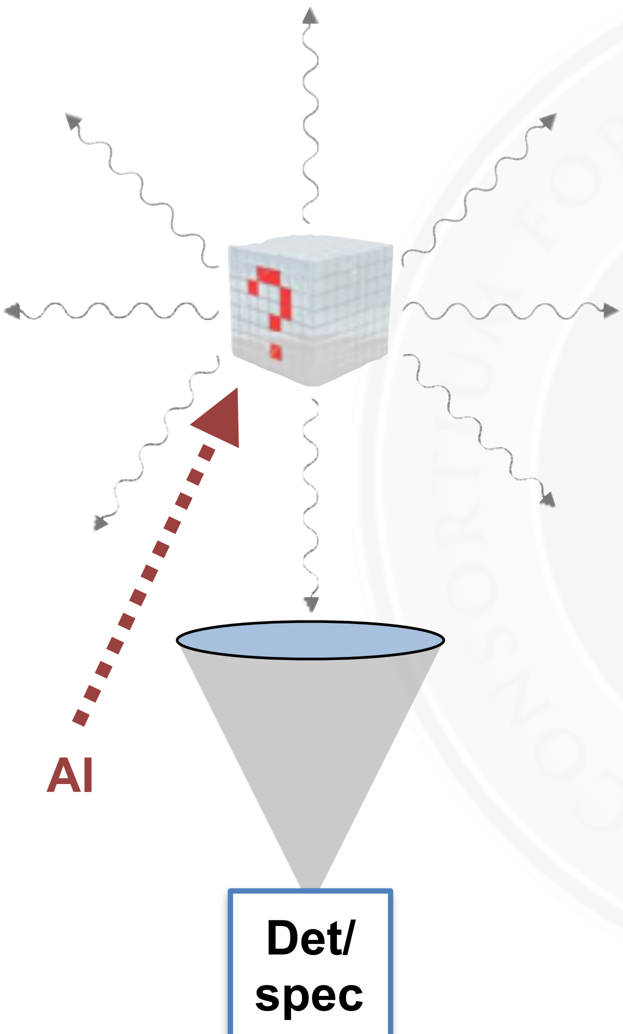
Development of advanced optical techniques for verification measurements

Igor Jovanovic
University of Michigan

Outline

1. Impetus and technical approach
2. Isotopic measurements from atomic and molecular spectra
3. Remote elemental and isotopic measurements by filamentation
4. Detection of chemical compounds as a signature of the enrichment activity
5. Team

Why use optical techniques?

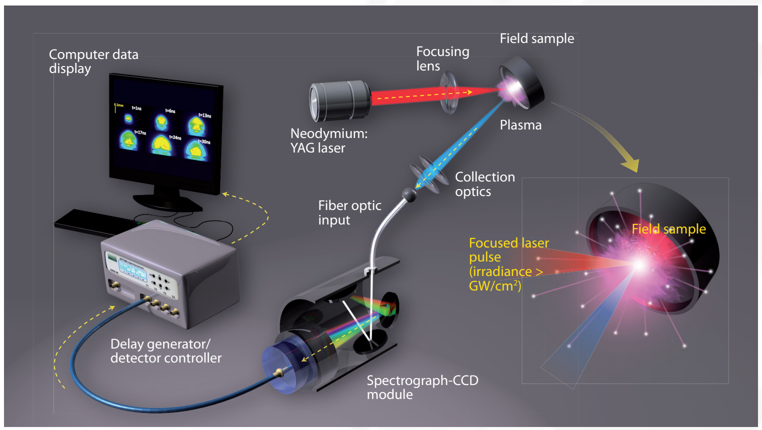


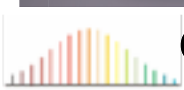
Criterion	Optical	n/γ
# of information quanta	✓	✗
Large collection aperture	✓	✗
High-resolution imaging	✓	✗
High energy resolution	✓	✗
High time resolution	✓	✓
Photopeak efficiency	✓	✗
Detector intrinsic efficiency	✓	✗
AI probe range	✓	○
Signature range	✓	○
Penetrating shielding	✗	✓
Non-radioactive material	✓	○
Detection of compounds	✓	✗
Isotope sensitivity	○	✓
Speed of detection	✓	✗

The focus of our work is detection of proliferation activities via optical techniques

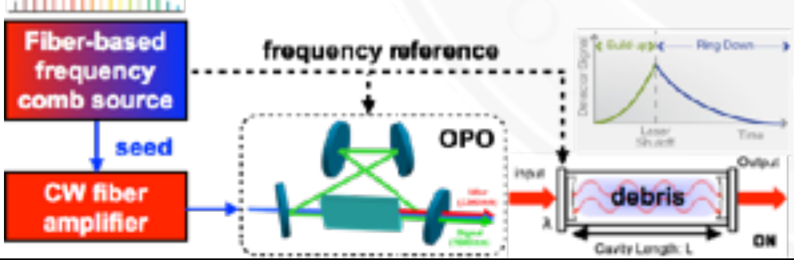
Near-Field Detection of Proliferation

Laser ablation - optical emission spectroscopy



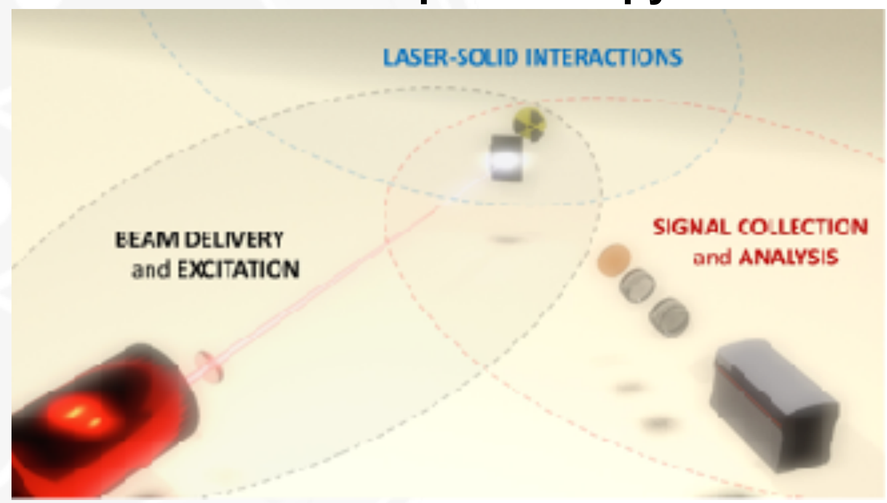


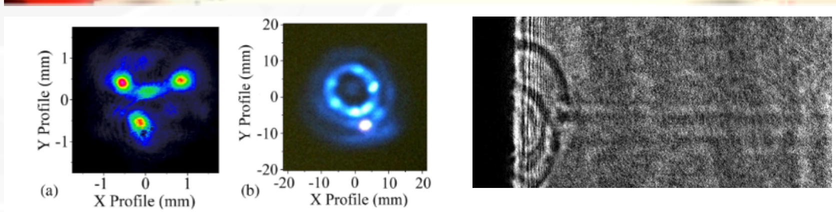
Cavity ringdown spectroscopy



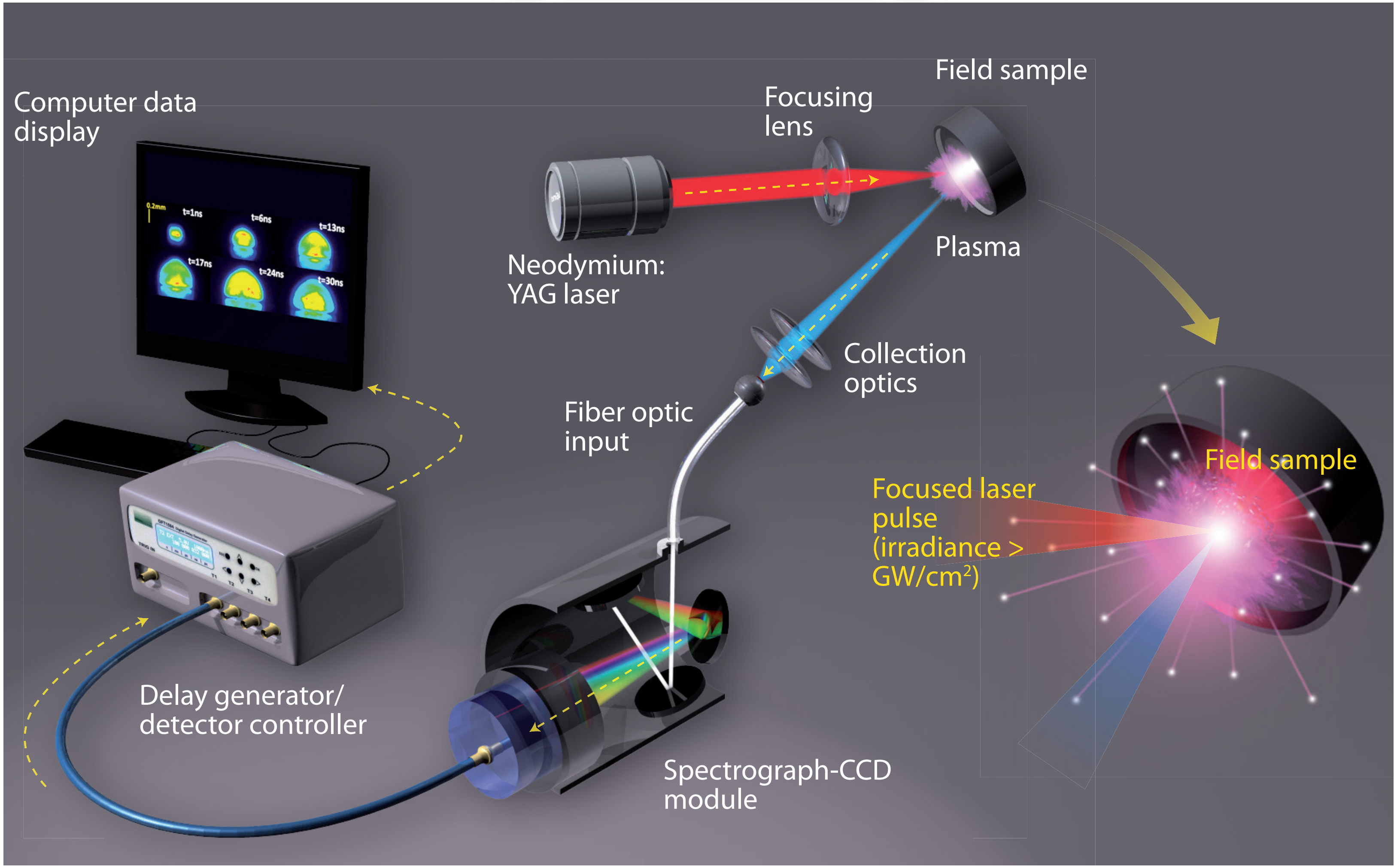
Remote Detection of Proliferation

Femtosecond filamentation laser ablation - optical emission spectroscopy

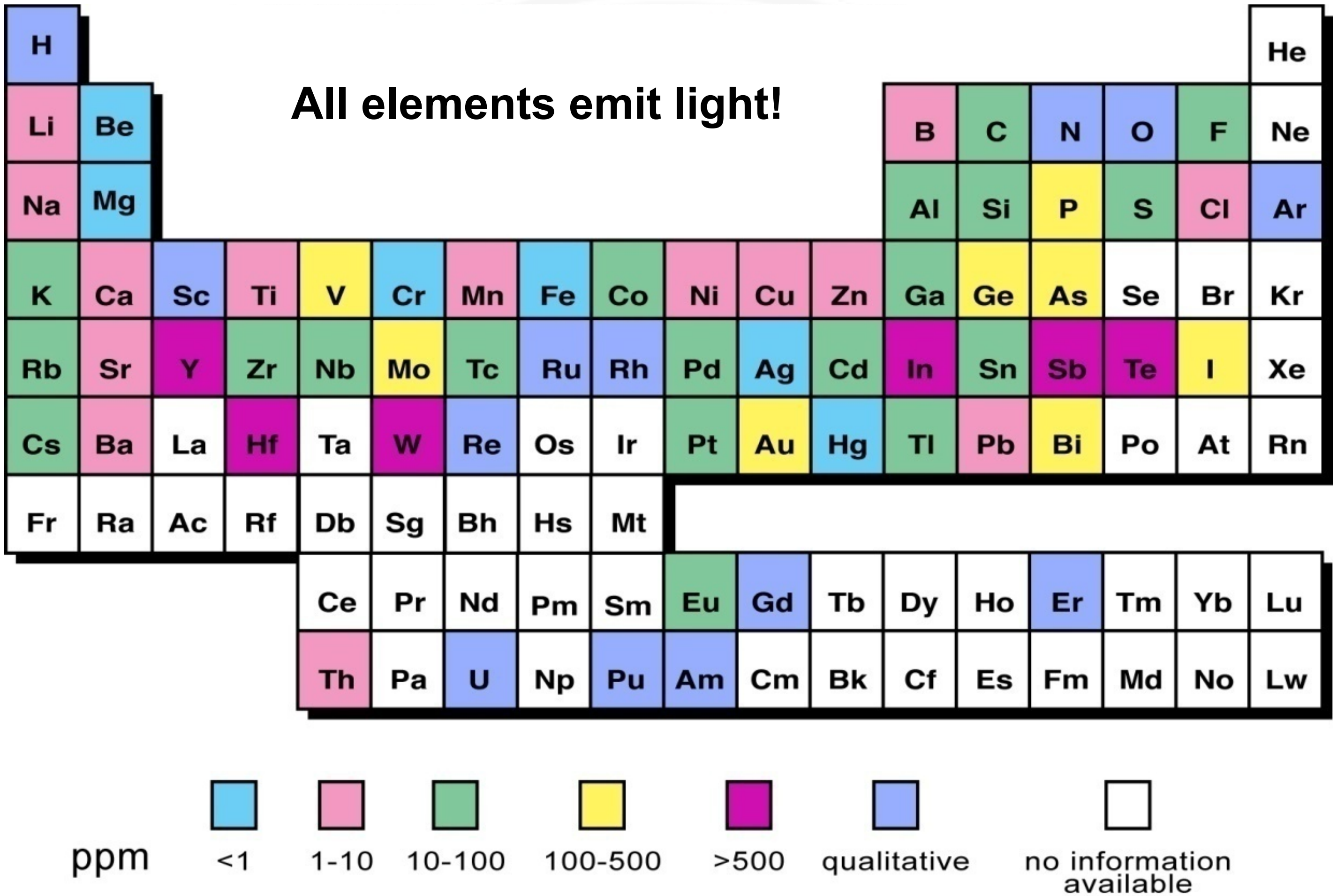




Laser-induced breakdown spectroscopy is a representative example of an optical technique

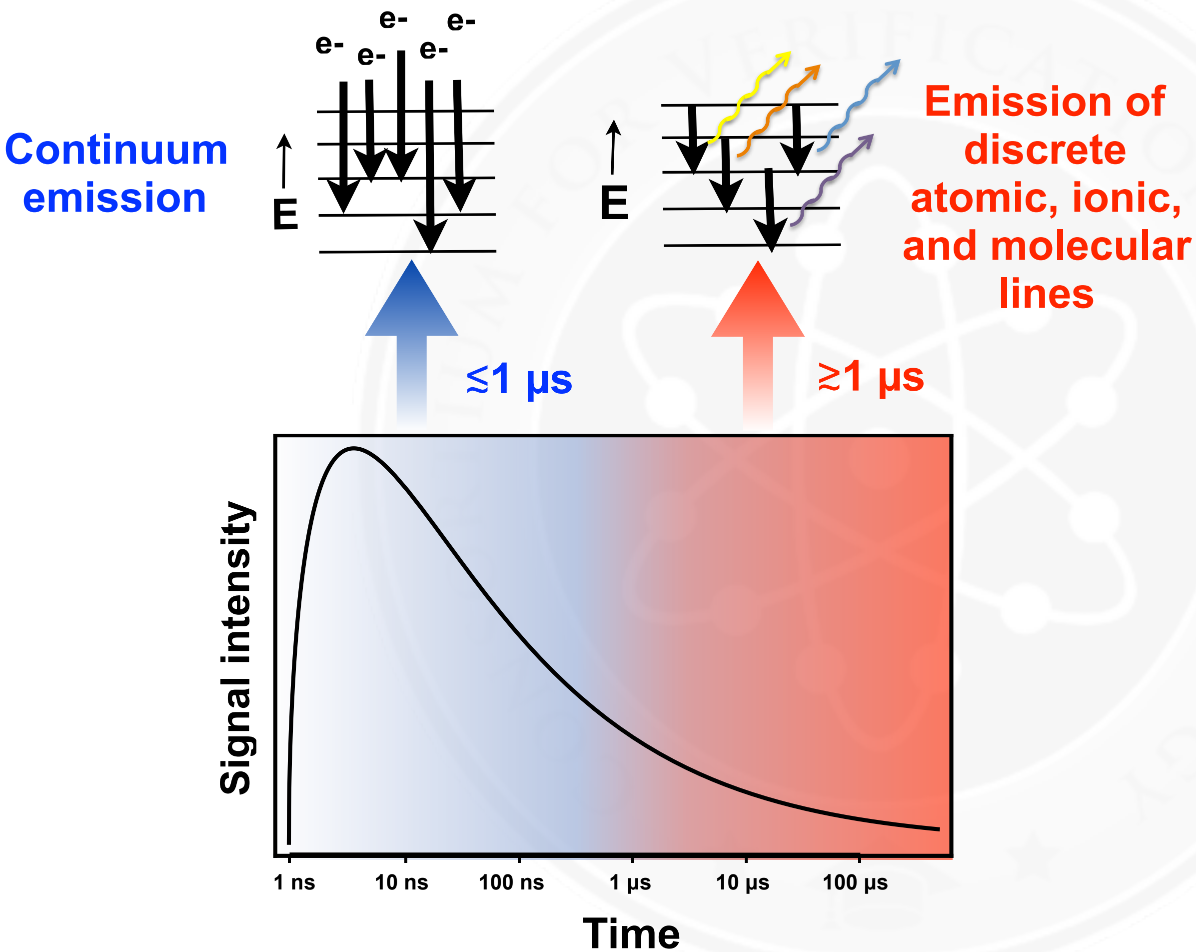


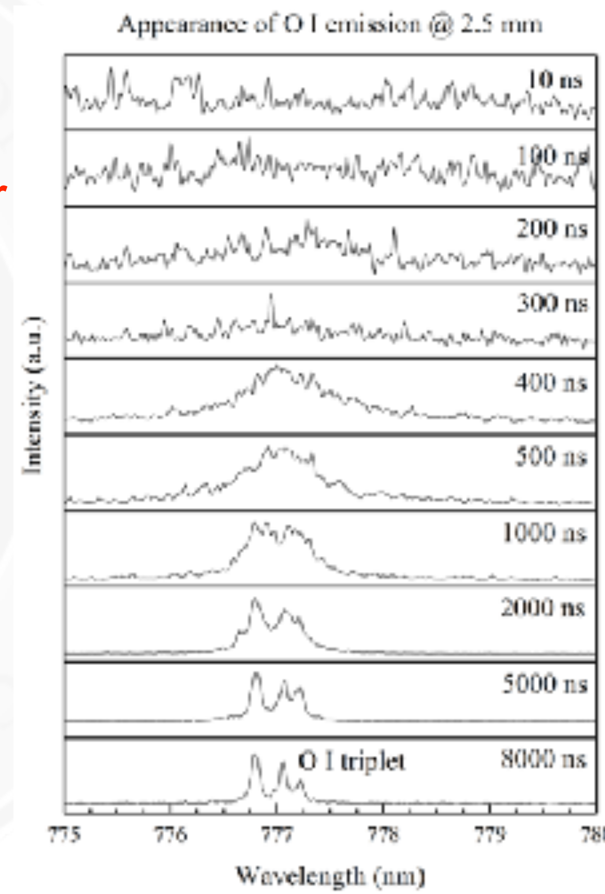
Elemental coverage of LIBS



J. E. Barefield II, INMM (2011)

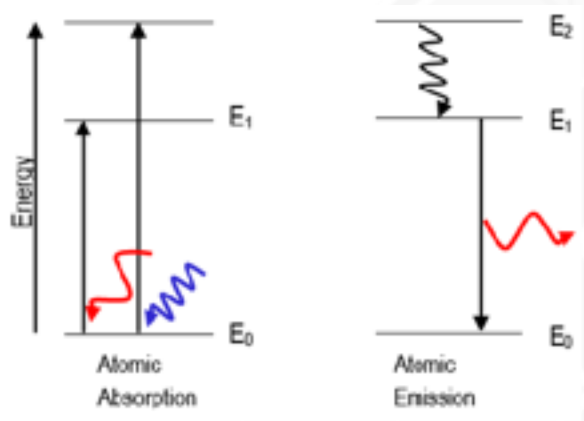
Spectral features of LIBS signals are time-dependent

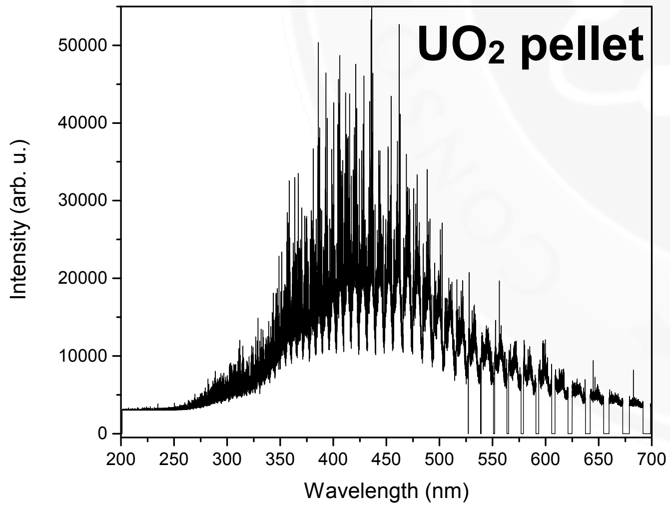




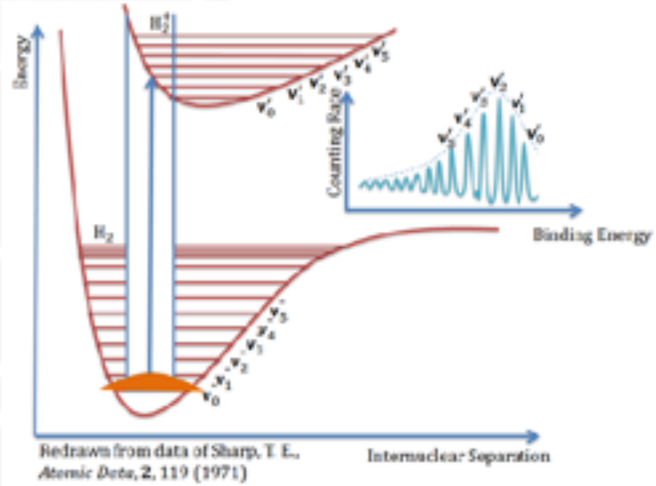
Atomic and molecular emission can be used for identification of elements or compounds

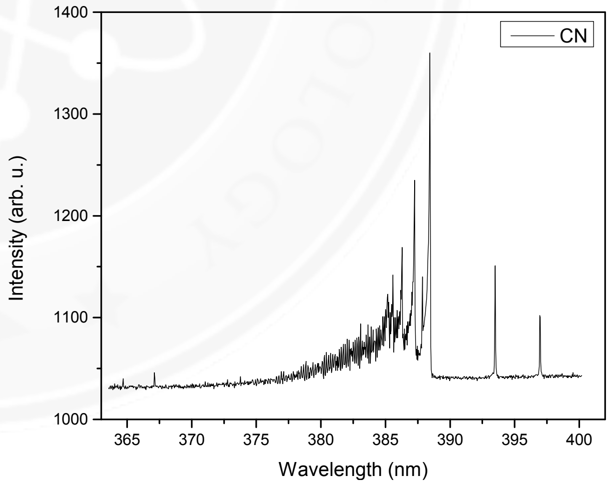
Atomic





Molecular





Origin of the optical isotope shift

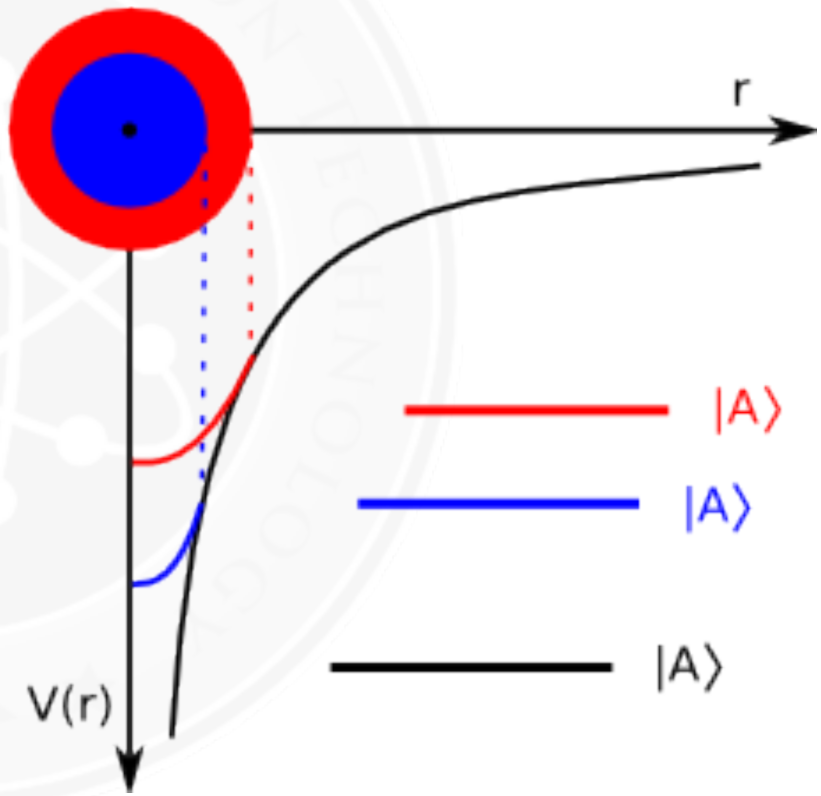
Mass effect

$$\left(-\frac{\hbar^2}{2}\nabla^2 + V\right)\psi = i\hbar\frac{\partial}{\partial t}\psi$$

$$m \rightarrow \mu$$

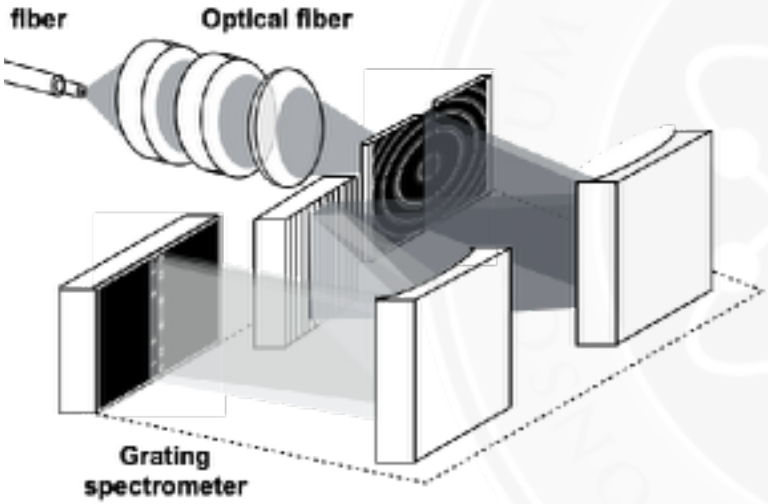
$$\mu = \frac{1}{1/m_1 + 1/m_2} = \frac{m_1 m_2}{m_1 + m_2}$$

Volume effect

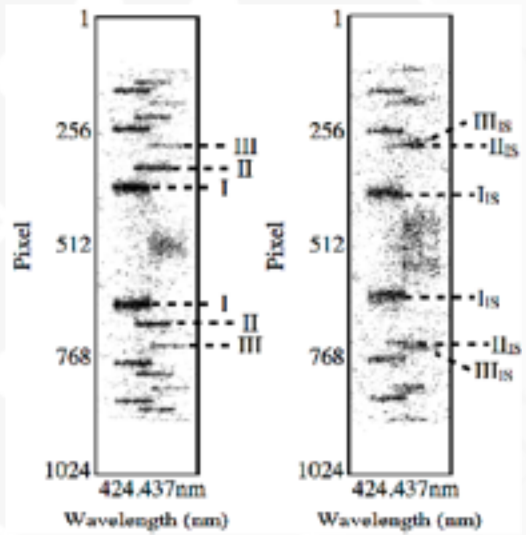


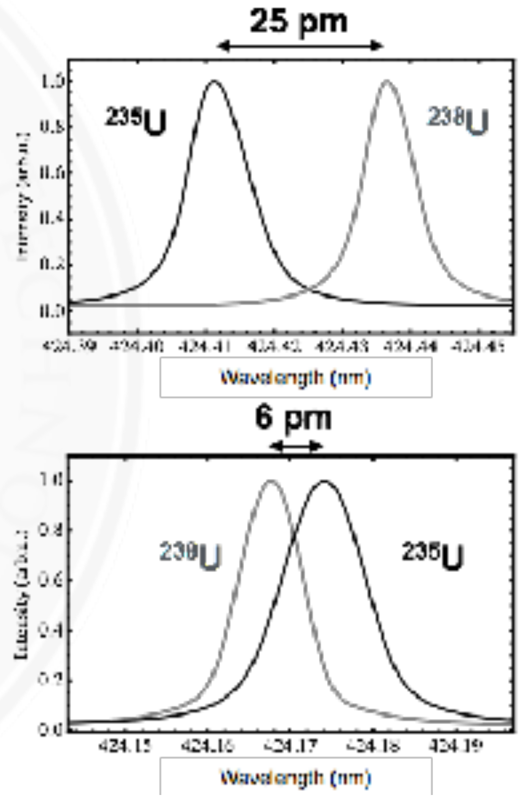
We developed a compact instrument and used it to measure $^{235}U/^{238}U$ isotope shift

We measured the $^{235}U/^{238}U$ atomic isotope shift down to 6 pm.



U-nat HEU

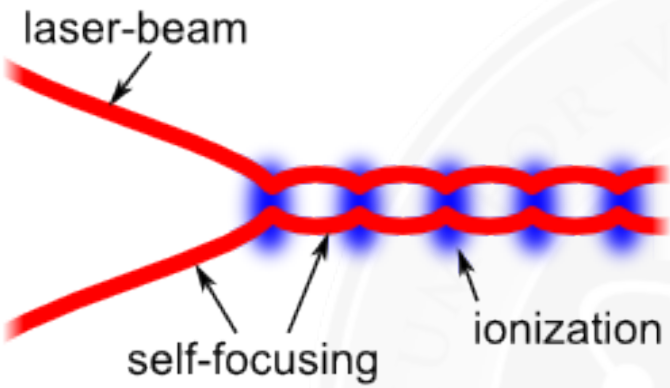


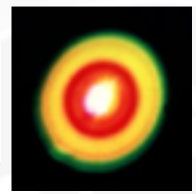


P. Ko, J. Scott, and I. Jovanovic, Opt. Commun. 357, 95-99 (2015).

P. K. Morgan, J. R. Scott, and I. Jovanovic, Spectrochimica Acta Part B: Atomic Spectroscopy 116, 58-62 (2016).

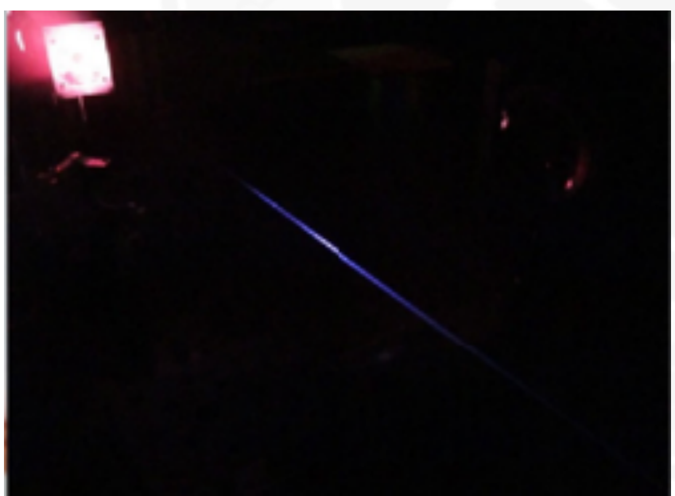
Remote excitation can be realized by use of laser filamentation





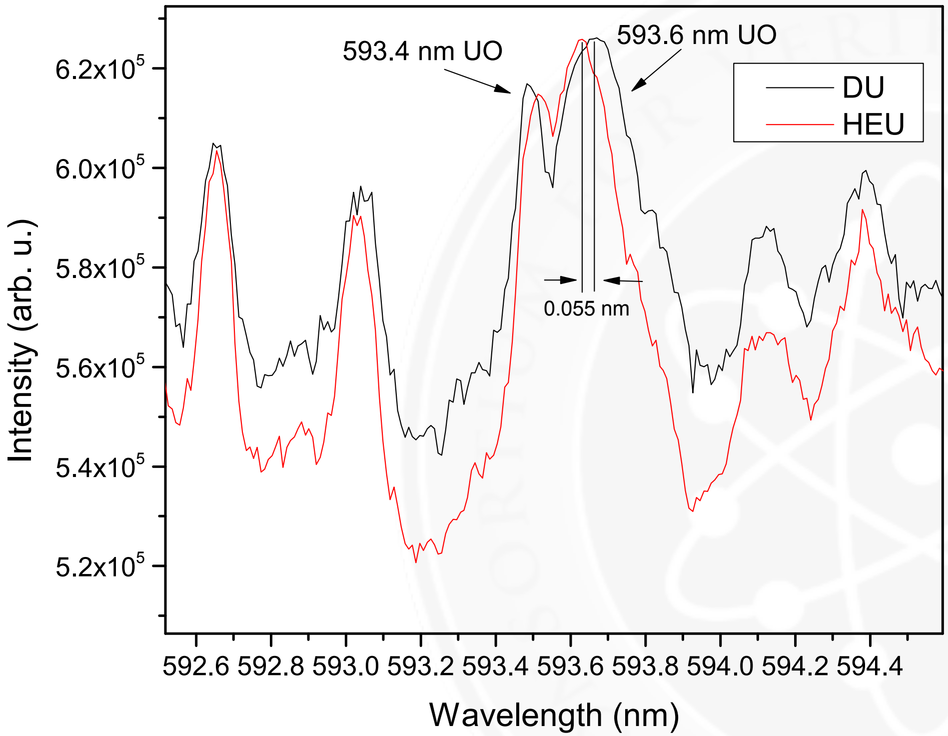
Filaments can be used to deliver laser excitation over multi-kilometer distances!

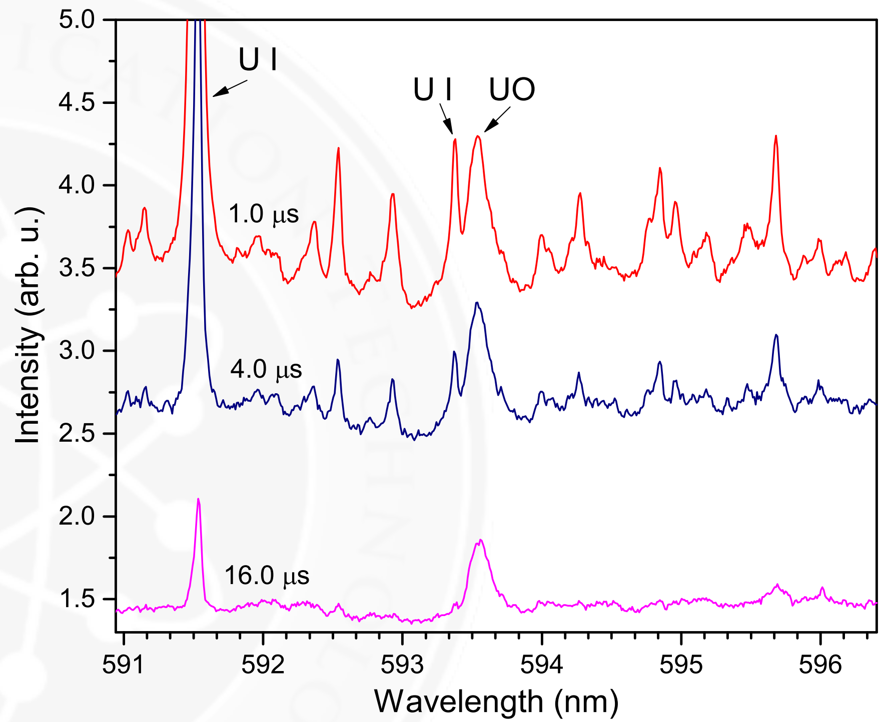
$$n = n_0 + n_2 I$$



Terramobile

Remote measurement of uranium enrichment via filament-produced uranium oxide

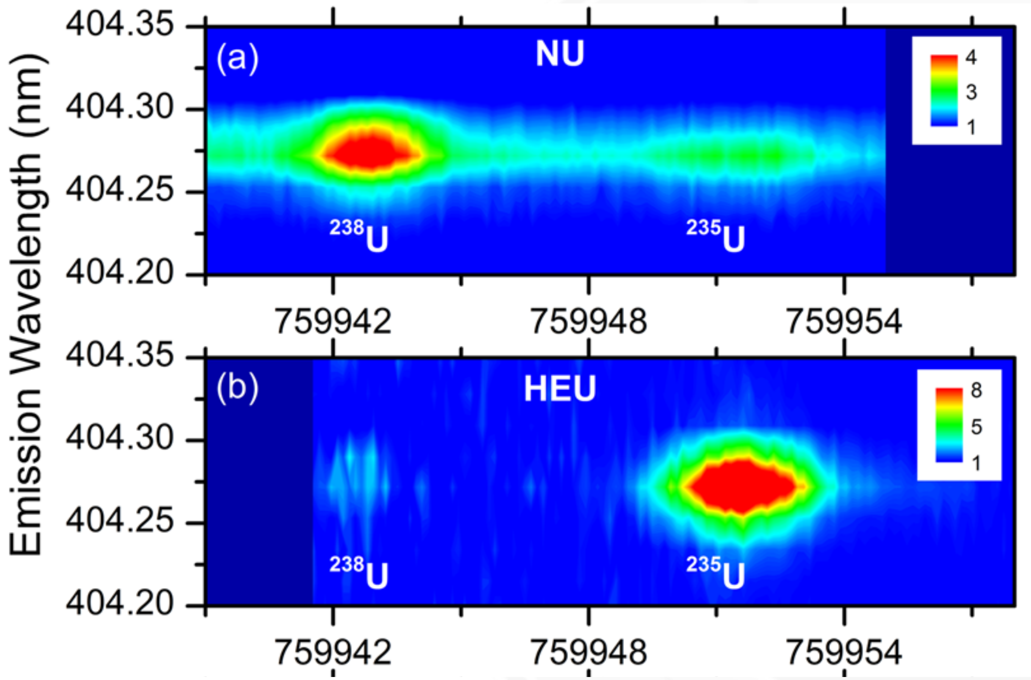


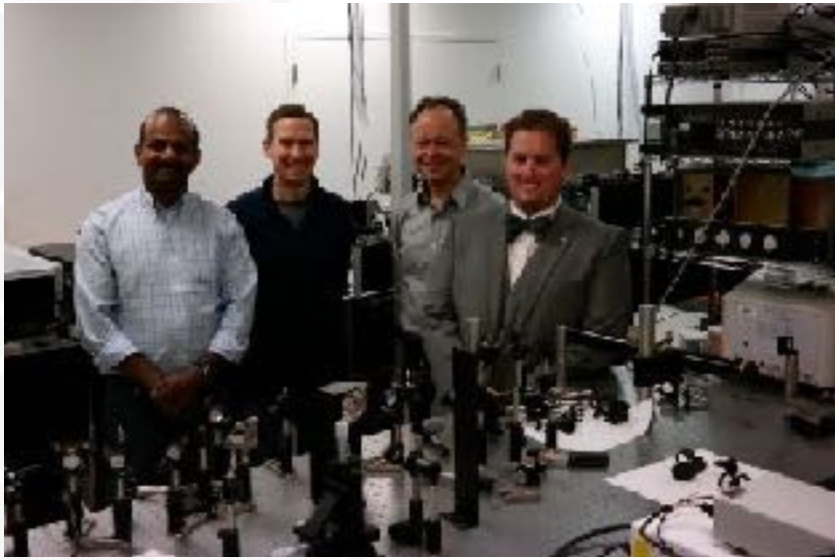


Molecular shift (Q-branch of the UO molecular transition, 593.6 nm) is ~2.5 times the atomic shift (U II, 424.43 nm).

K. C. Hartig, I. Ghebregziabher, and I. Jovanovic, Sci. Rep. 2017

The first measurement of uranium isotopic composition via 2D fluorescence spectroscopy



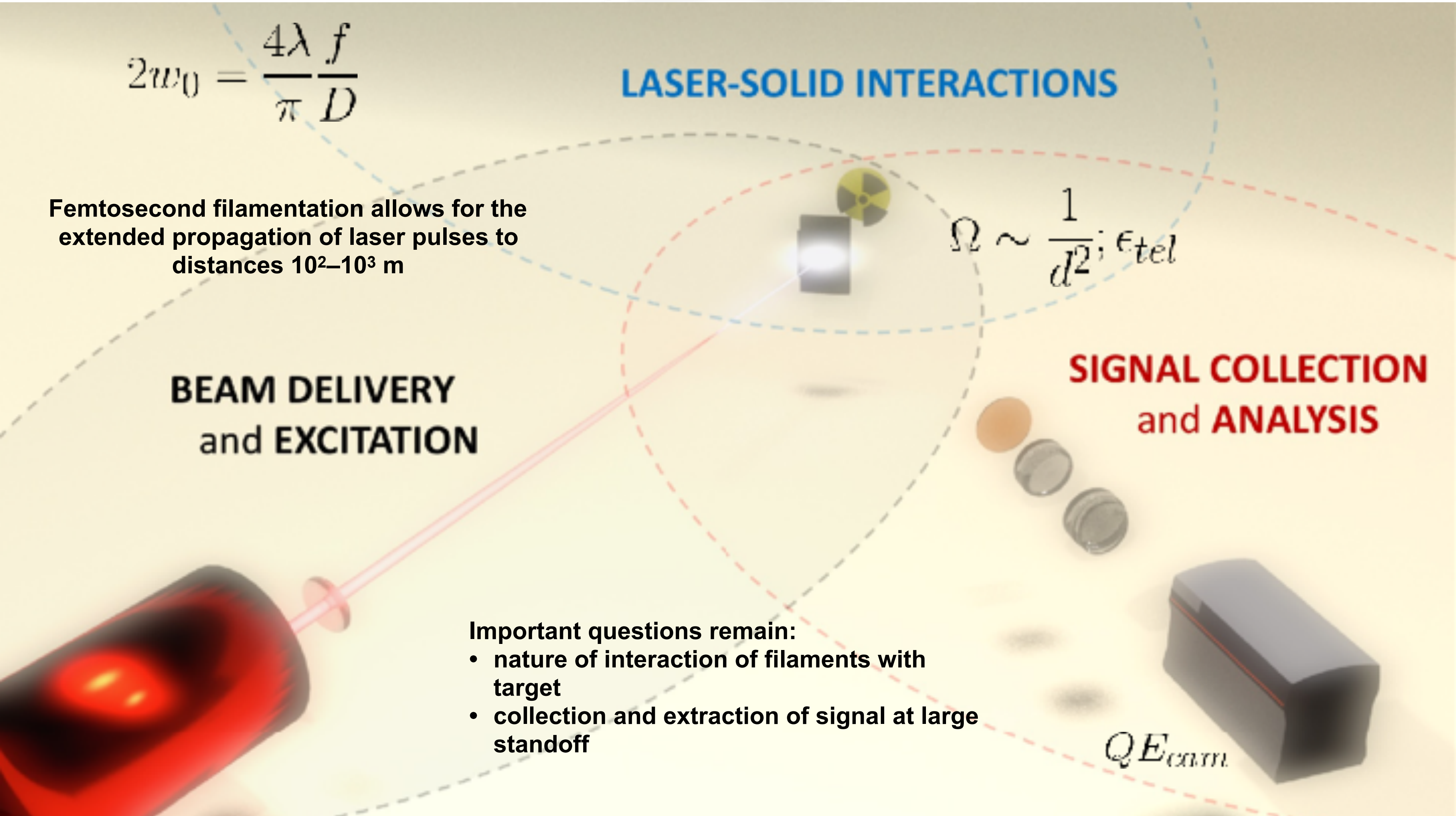


PNNL's top DNN R&D highlight, 2017

Pacific Northwest NATIONAL LABORATORY

M. Phillips, B. E. Brumfield, N. LaHaye, S. S. Harilal, K. C. Hartig, and I. Jovanovic, Sci. Rep. 2017

Remote sensing requires an understanding of beam delivery, signal production, and collection



Femtosecond filamentation allows for the extended propagation of laser pulses to distances 10^2–10^3 m

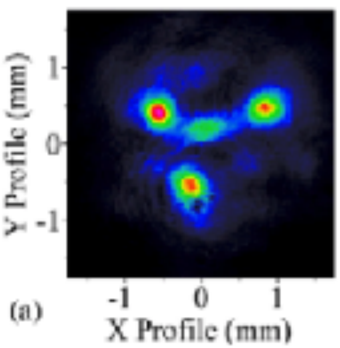
Important questions remain:

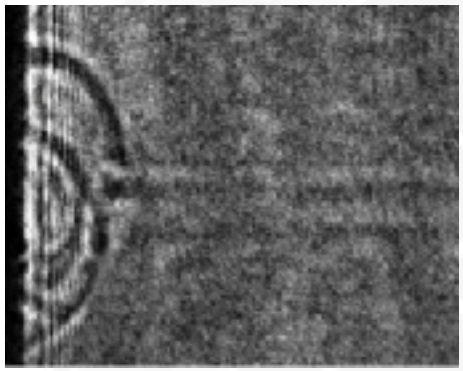
- **nature of interaction of filaments with target**
- **collection and extraction of signal at large standoff**

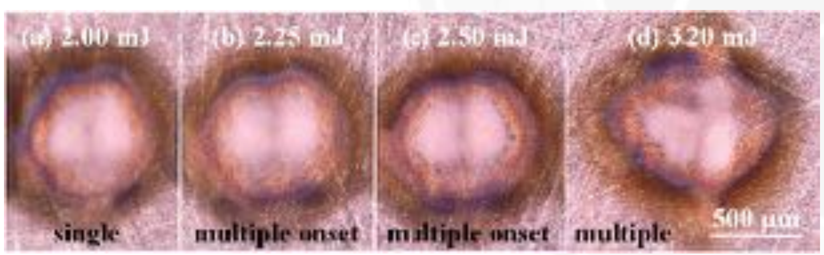
Effect of multiple filamentation on signal intensity

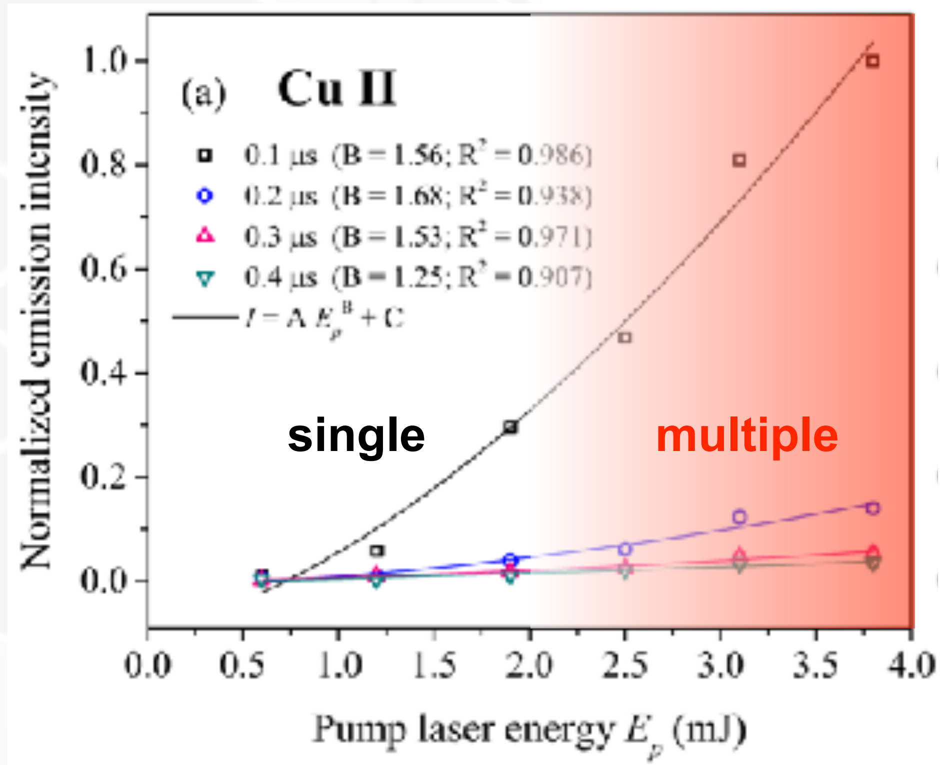
At high energies there is a break-up into multiple filaments.

Detected spectroscopic signal continues to increase despite multiple filamentation!





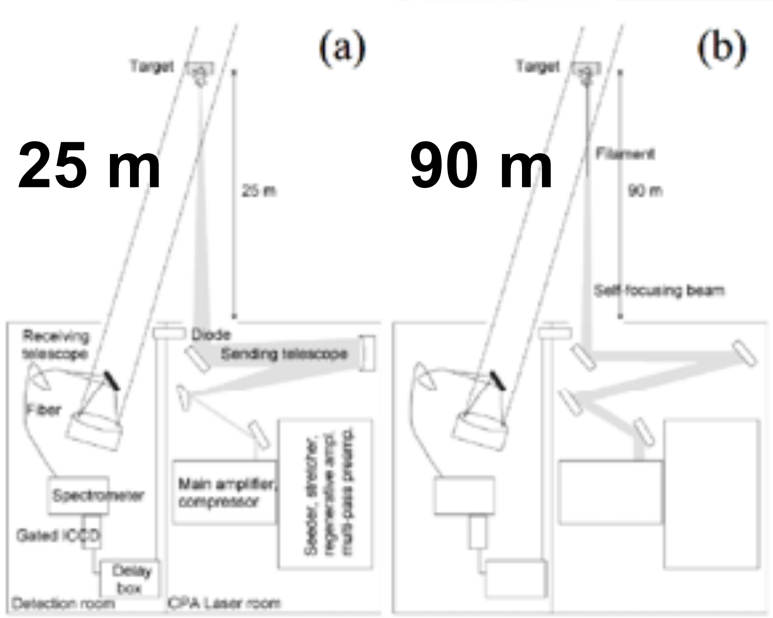


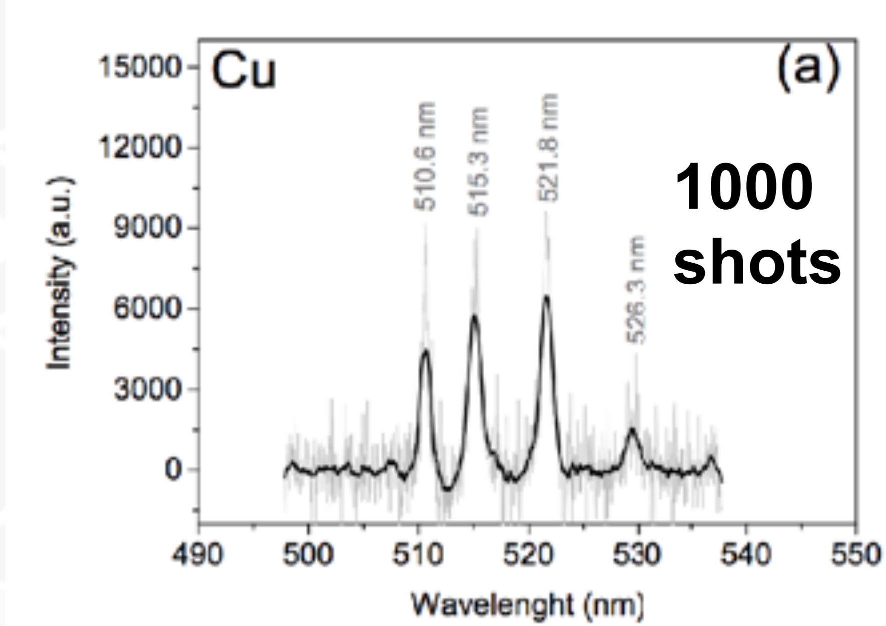


P. J. Skrodzki, M. Burger, and I. Jovanovic, Sci. Rep. (2017)

Once we create the signal, from what distance can we measure the characteristic optical emission?

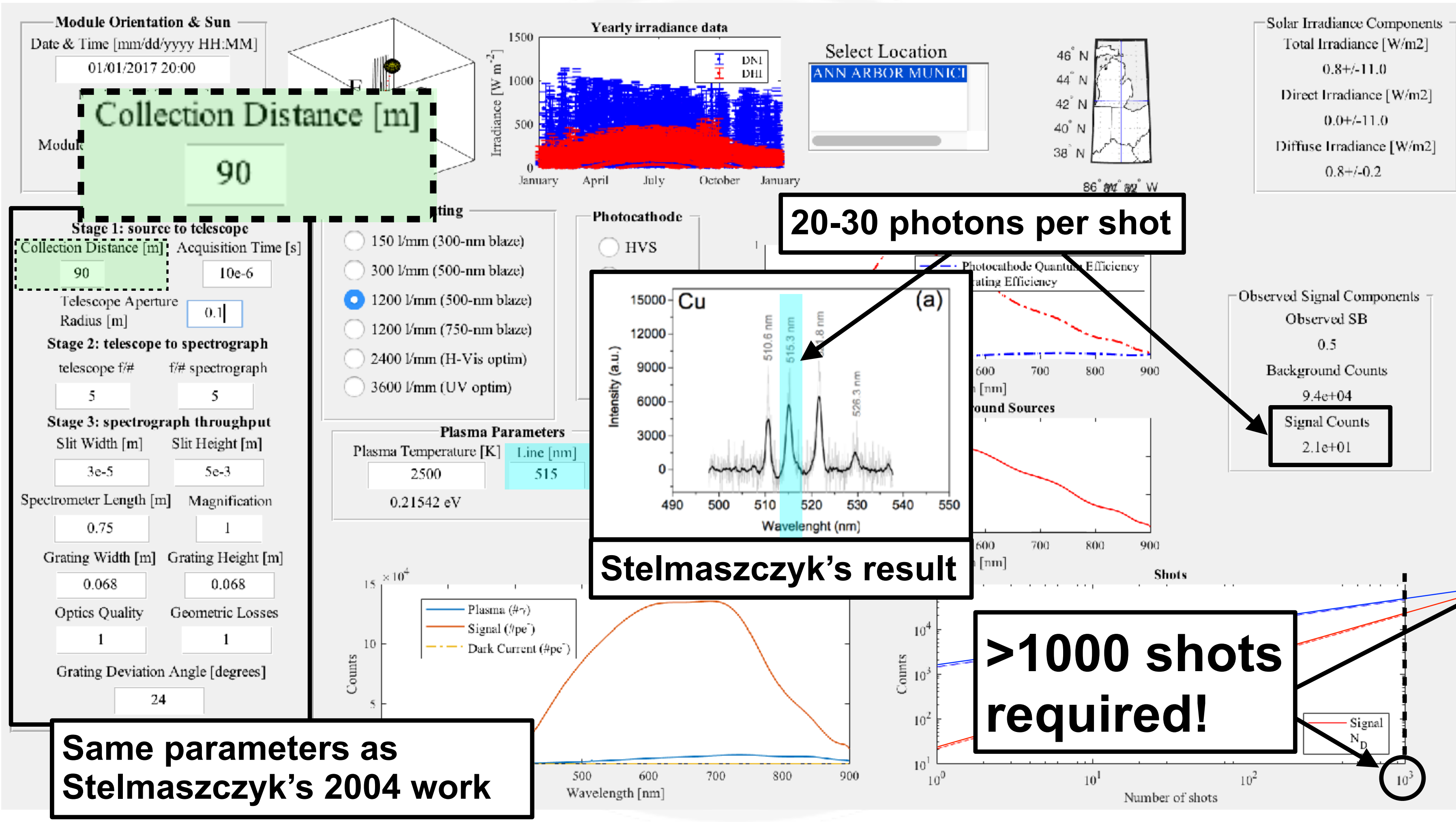
- Measured copper & steel emission from ~10^2 m
- counted ~20-30 signal photons per shot @ 90 m!





K. Stelmaszczyk et al., Phys. Rev. E, Vol. 60 (2004)
Ph. Rohwetter et al., J. Anal. At. Spectrom., Proc. SPIE 6158 (2004)

We simulate a detection system to help guide upcoming experiments in which we extend the collection distance

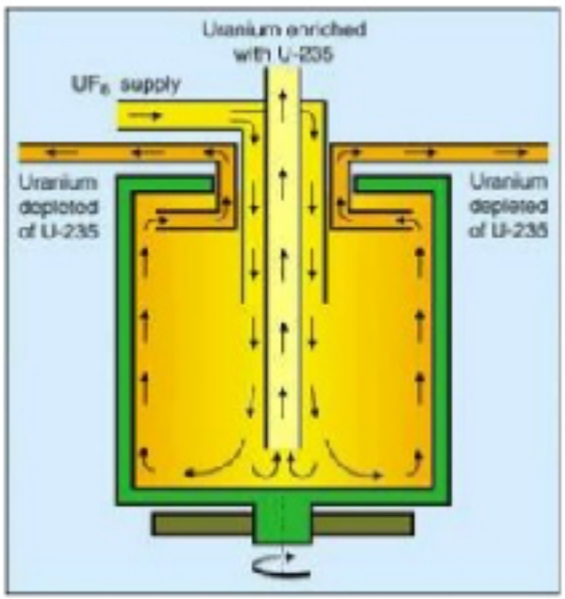


Uranyl fluoride is a unique signature of enrichment activity which may be effectively detected by the optical techniques

UO_2F_2 aerosols are produced by hydrolysis of UF_6.

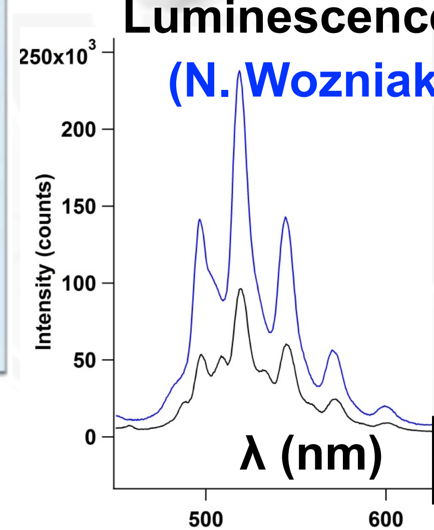
$$\boxed{UF_6 + 2H_2O \rightarrow UO_2F_2 + 4HF}$$

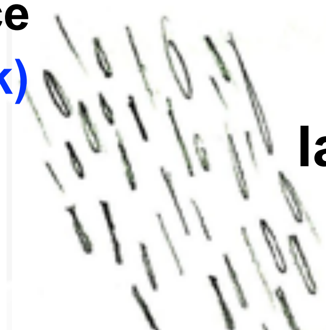
Hydrolysis



Enrichment process

Cavity ringdown
Filamentation LIBS



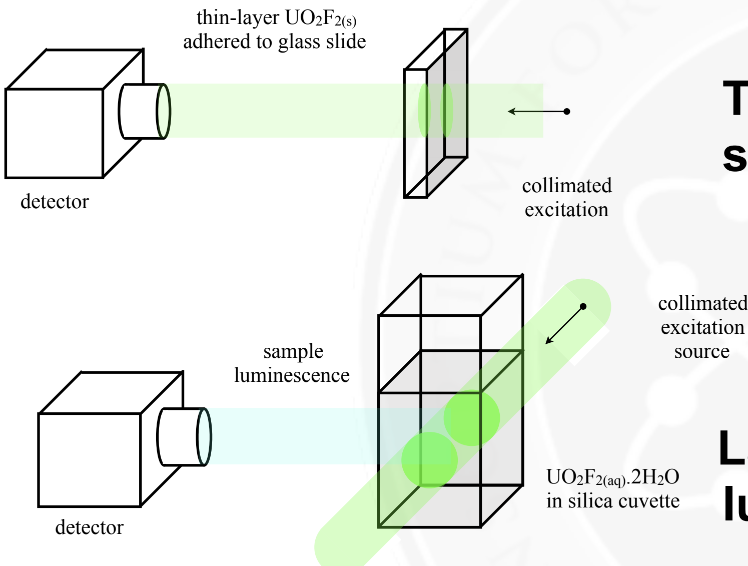


large particles - ground deposits

Laser induced fluorescence LIBS

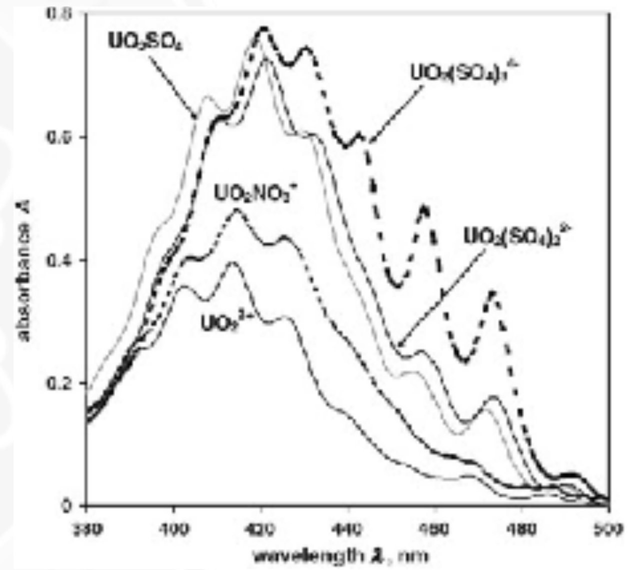
We are investigating the signatures and optical methods for detection of uranyl fluoride

We recently partnered with K. Czerwinsky (UNLV) to obtain UO_2F_2 samples, which will be used in experiments at Michigan.



Transmission spectroscopy

Laser-induced luminescence



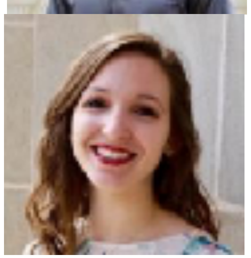
We also established a collaboration with A. Hero (EECS) to develop a unified method to understand the time-dependent spectral signatures of UO_2F_2 and other compounds.

Research team

Igor Jovanovic
faculty

Milos Burger
postdoctoral fellow

Patrick Skrodzki
PhD student



Lauren Finney
PhD student

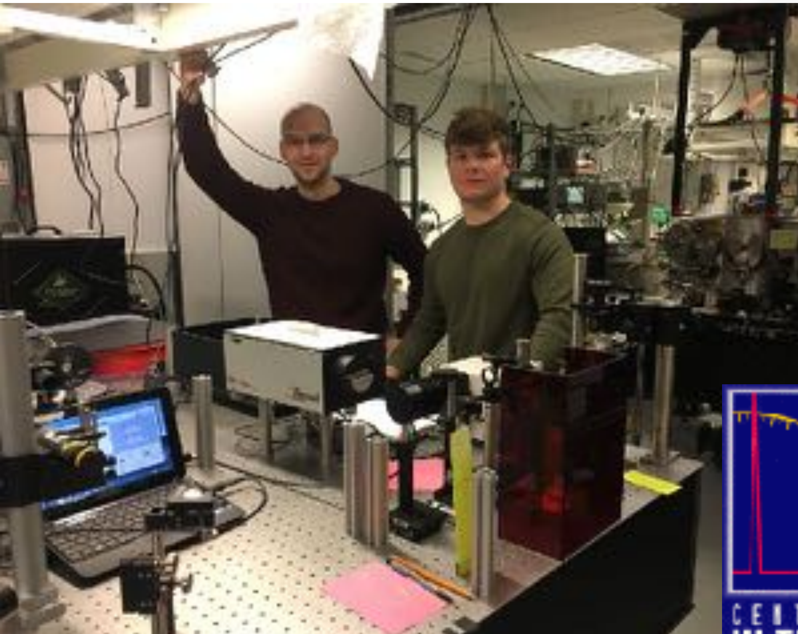
Recent Graduates

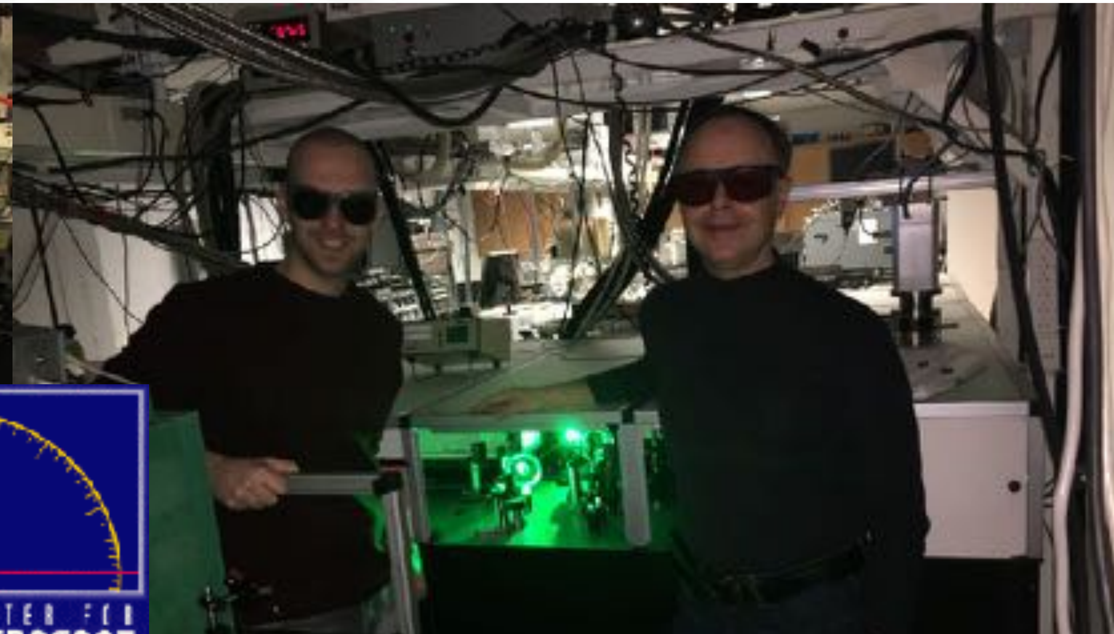
Phyllis Morgan
Ph.D. 2015
DTRA

Kyle Hartig
Ph.D. 2016
PNNL & University of Florida

Ian Baughman
Undergraduate student

Thank you and please attend our lab tour





CENTER FOR ULTRAFAST OPTICAL SCIENCE

This work was funded by the Consortium for Verification Technology under Department of Energy National Nuclear Security Administration award number DE-NA0002534.