Standoff Characterization of Nuclear Materials by Optical Techniques

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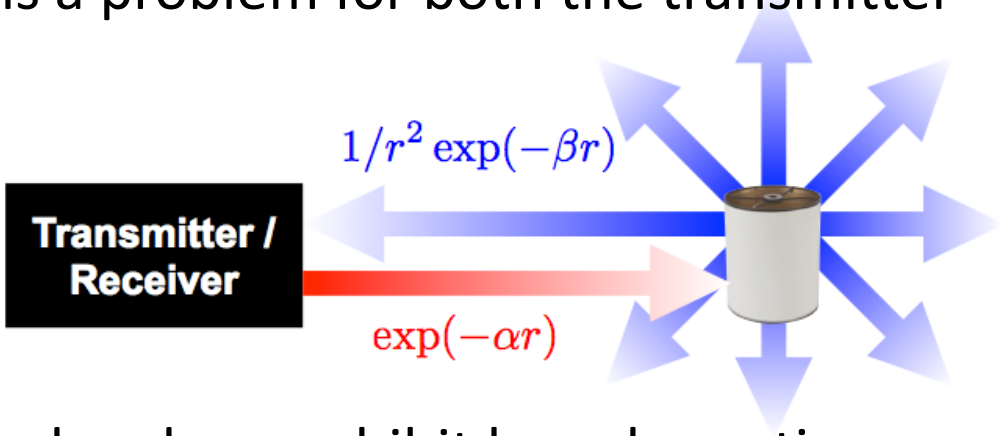
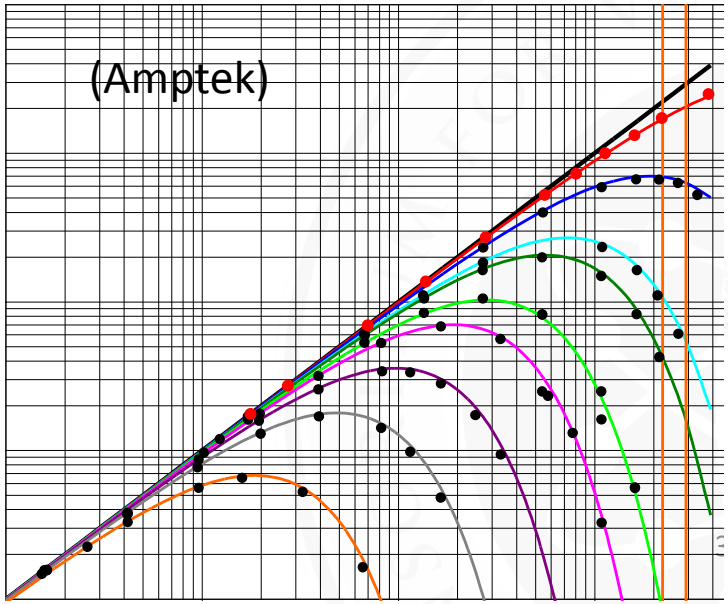
Challenges in standoff detection and characterization of nuclear materials

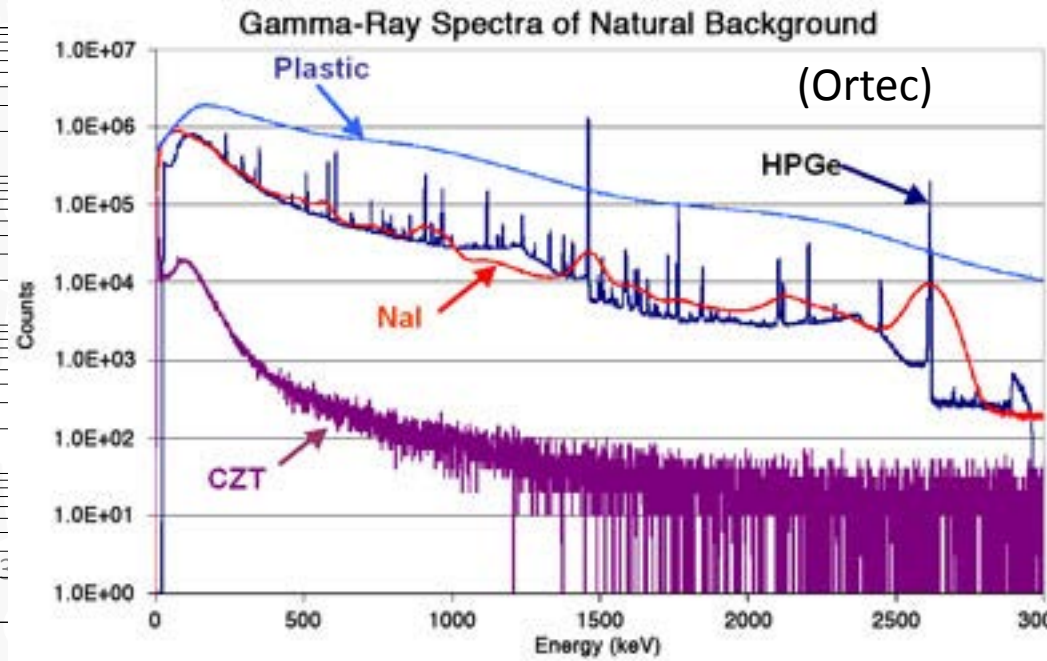
- Reasonably well collimated probes are available (accelerator-produced radiation sources and lasers)
- The return signal is invariably emitted nearly isotropically
- Atmospheric attenuation is a problem for both the transmitter and the receiver
- We wish we could produce a collimated return signal or guide it in some fashion



- Laser light is well collimated and can exhibit low absorption, but both fundamental and practical issues exist (Rayleigh length, air turbulence).

Detecting ionizing radiation can be problematic in high rate, complex radiation environments





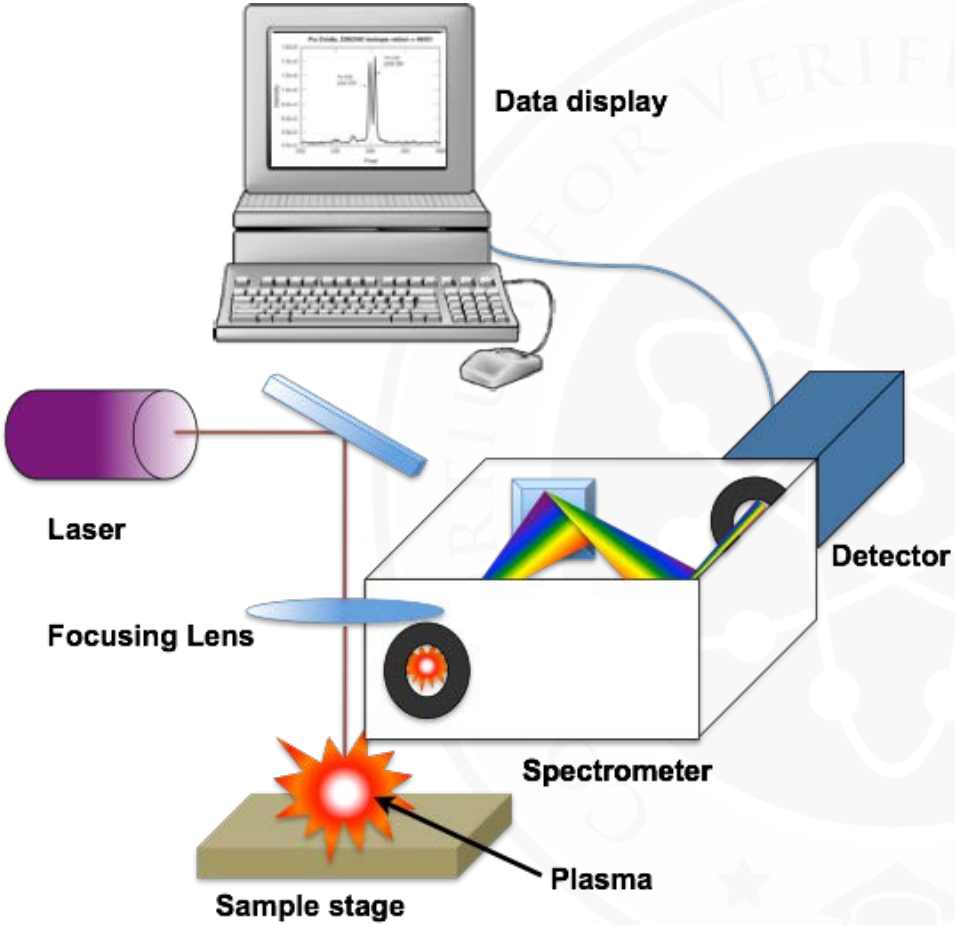
- Detector resolution
- Detector dead time and accidentals rate
- Particle ID in mixed radiation field

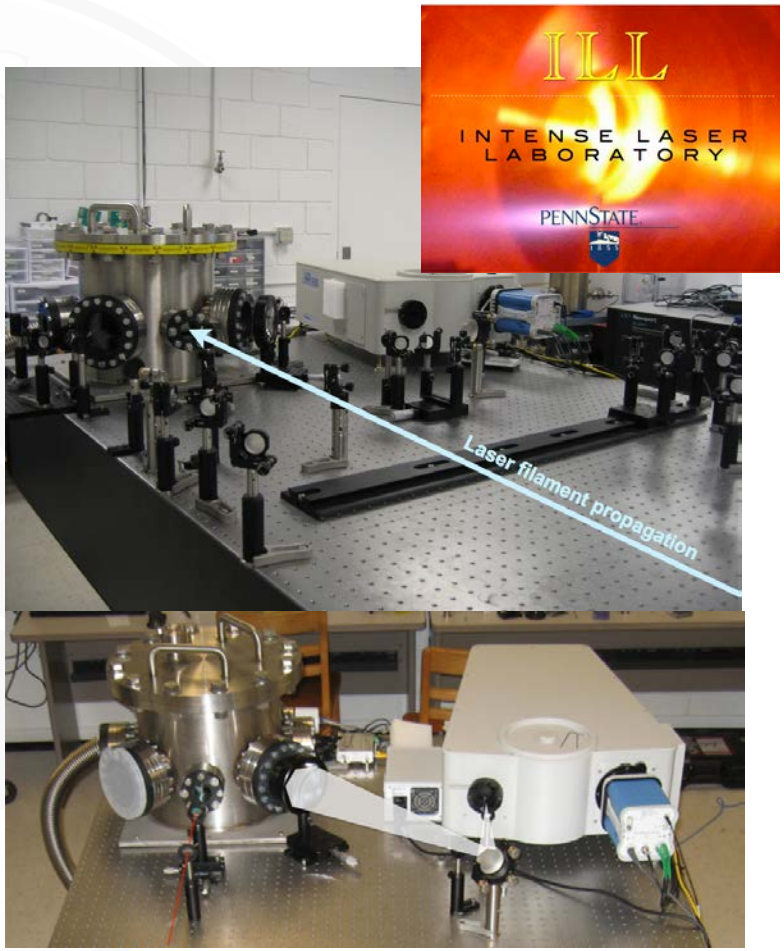
Why optical techniques?

- ☑ Availability of well-collimated, "zero-dose" optical probes
- ☑ Nearly lossless, physics-limited manipulation of optical photons available with COTS components
- ☑ High spatial (µm) and temporal (<fs) resolution
- ☑ Spectroscopic information available and typically not limited by statistics of information carriers
- ☑ Simple and powerful background rejection by imaging, temporal gating, spectral, and polarization filtering
- ☑ Detection of non-radioactive materials

- ✸ Direct line-of-sight access to material needed
- ✸ Limited to characterization of bare surfaces
- ✸ Susceptible to matrix effects and interferences

Laser-induced breakdown spectroscopy

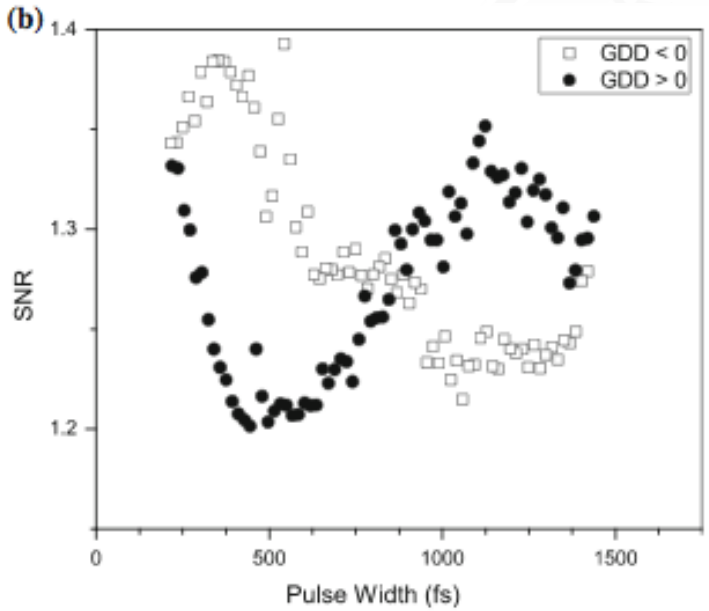




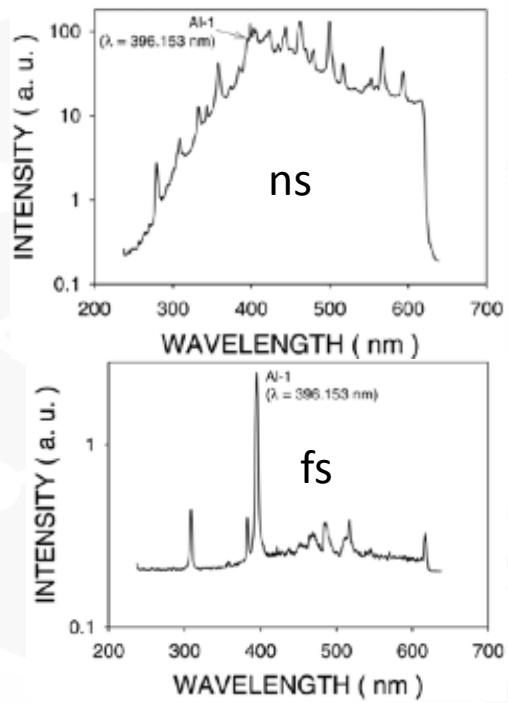
P. Ko, K. Hartig, K., R. Schur, T. Jacomb-Hood, and I. Jovanovic, I. (2013) Review of Scientific Instruments, 84, pp. 013104.

Femtosecond LIBS has unique properties

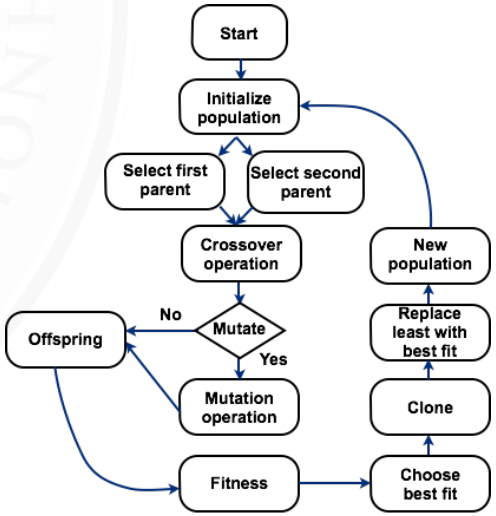
Signatures of coherent control in fs LIBS of U



(D.R. Alexander)

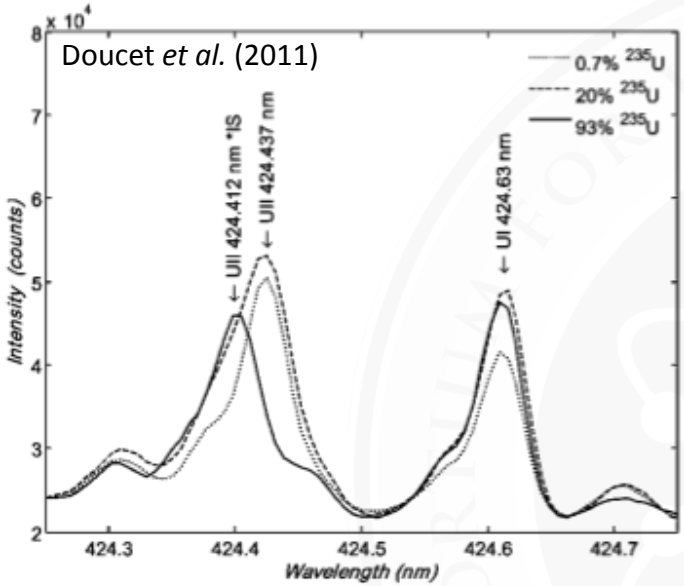


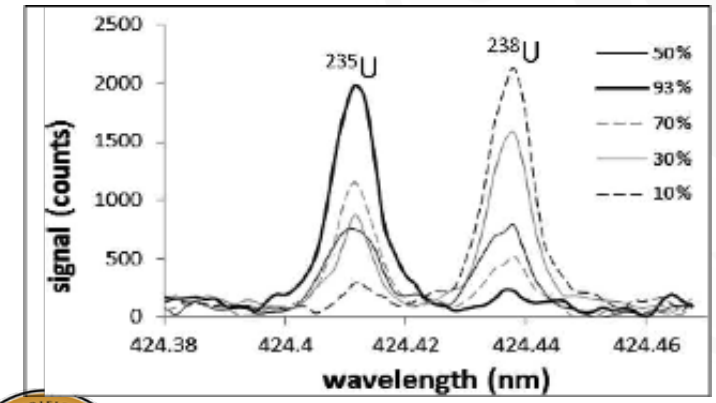
Optimization by use of a genetic algorithm



K.C. Hartig et al., J. Rad. Nucl. Chem. 296, 135 (2013).
K.C. Hartig et al., submitted

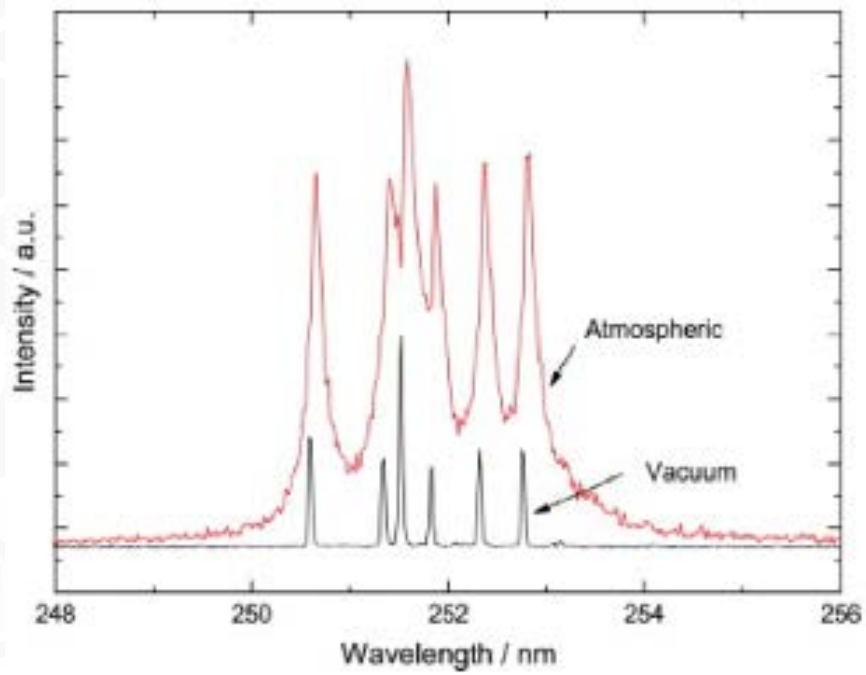
Isotopic measurements are needed





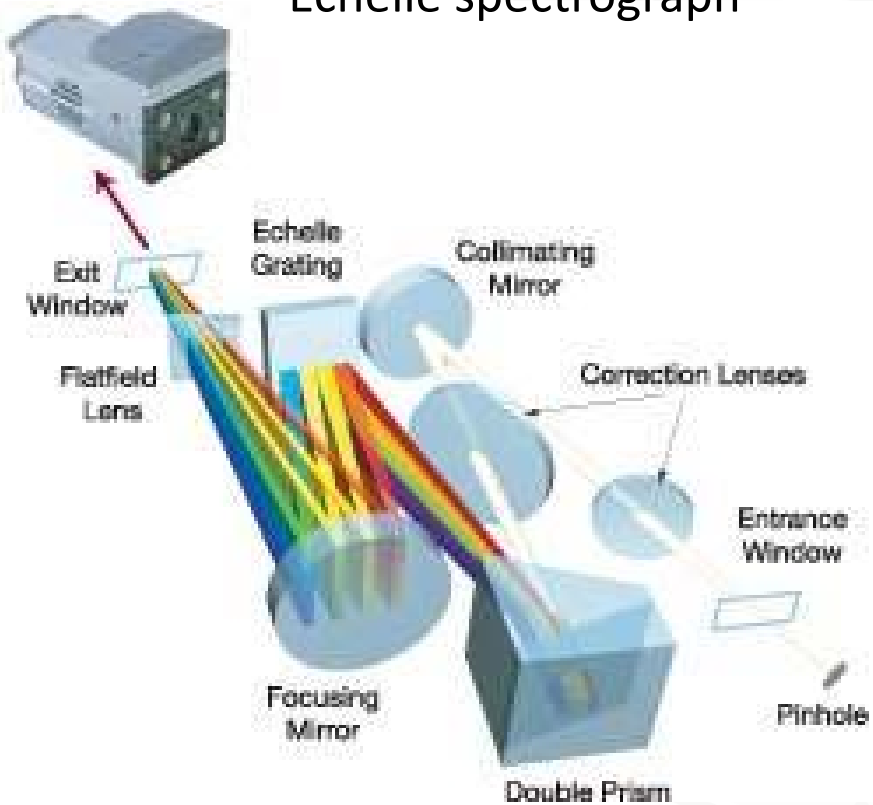
Cremers et al. (2012)

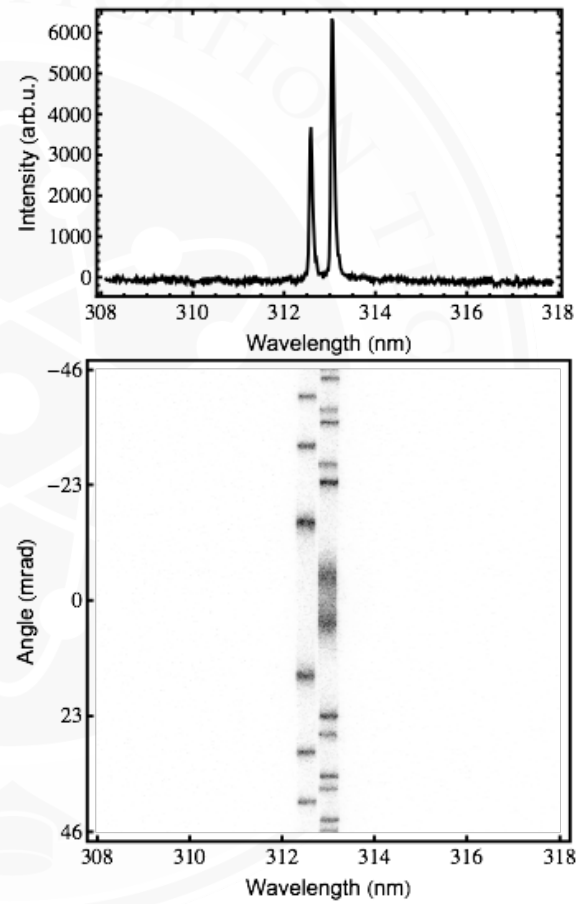
- Isotopic shift is small and can be affected by Stark and Doppler broadening



Compact and inexpensive technology to measure small isotopic shifts is desirable

Echelle spectrograph





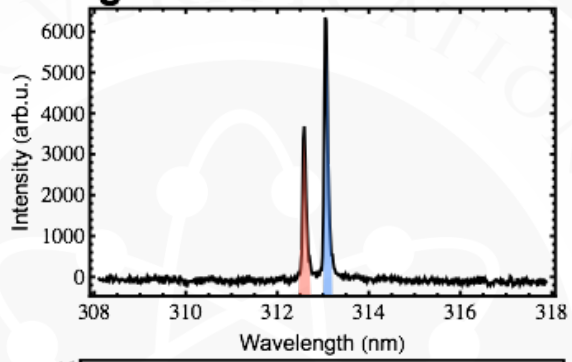
Hybrid Fabry-Perot – dispersive spectrometry

P. Ko, IMMM (2014)

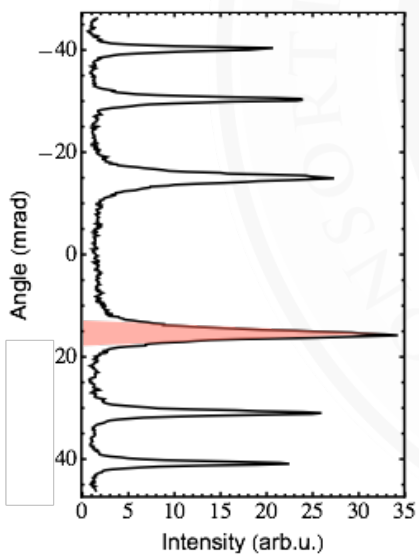
We are developing a hybrid interferometric/dispersive spectrometer

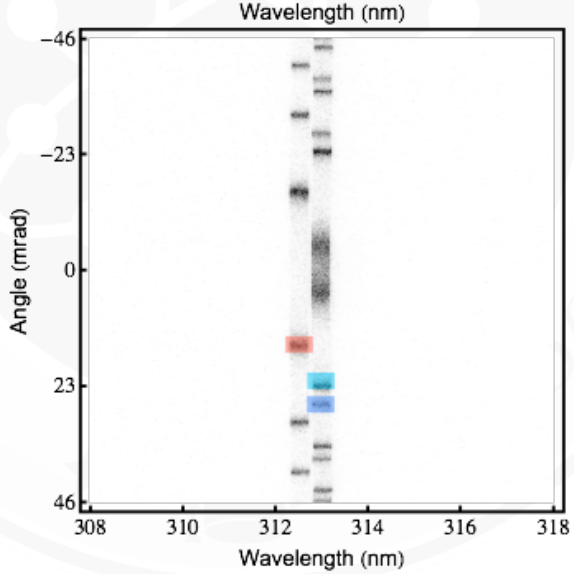
Measurement of Hg 312.57-nm line and 313.2-nm doublet

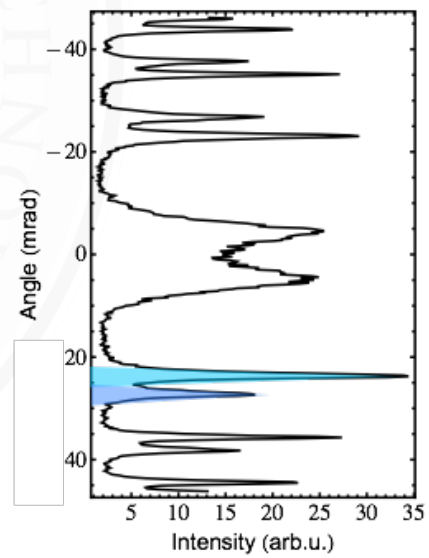
Hg(Ar) lines from a grating spectrometer.



The 313.2-nm doublet is resolved by the FP etalon.

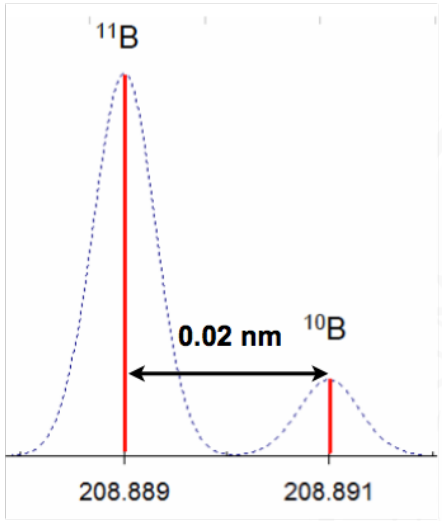




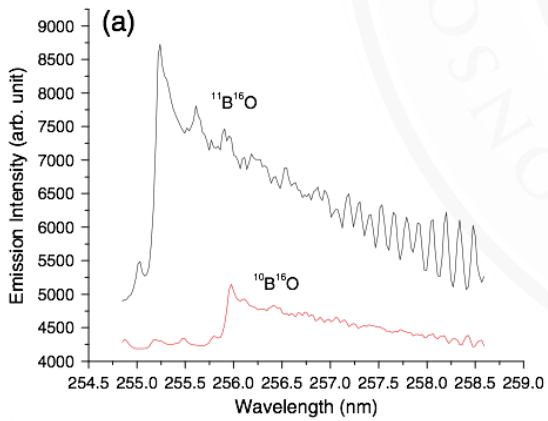


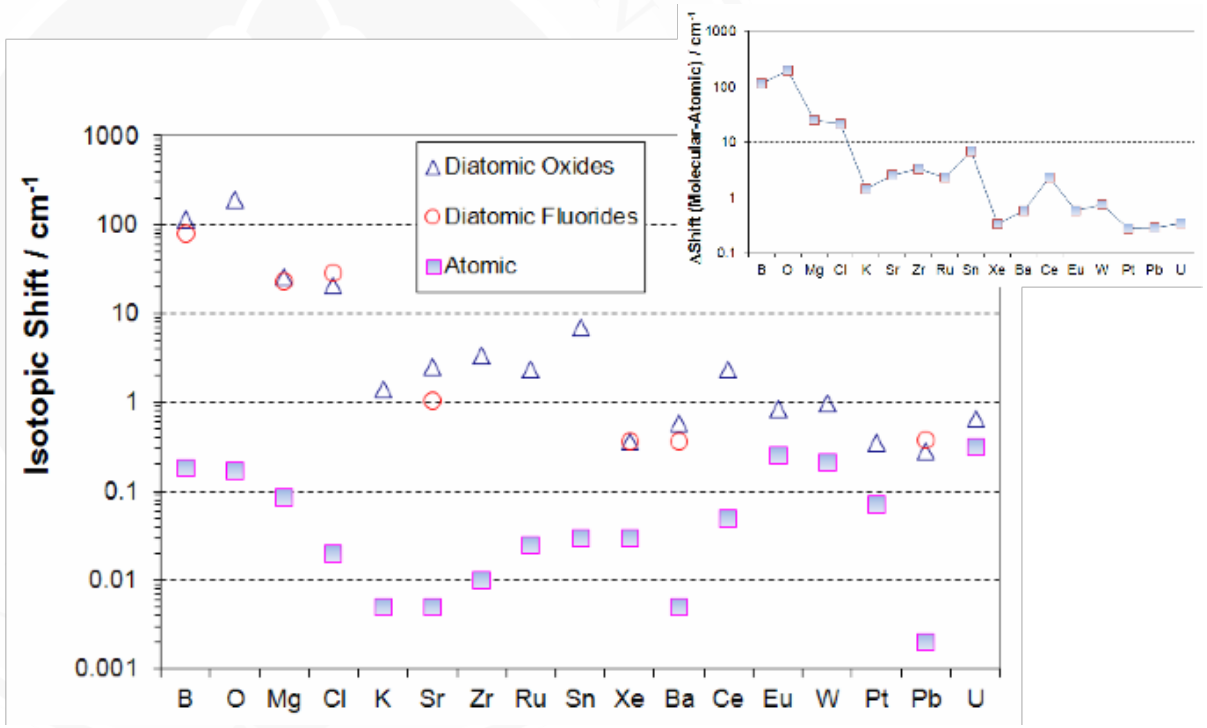
FP ring pattern

Atomic vs molecular spectroscopy for isotopic measurements

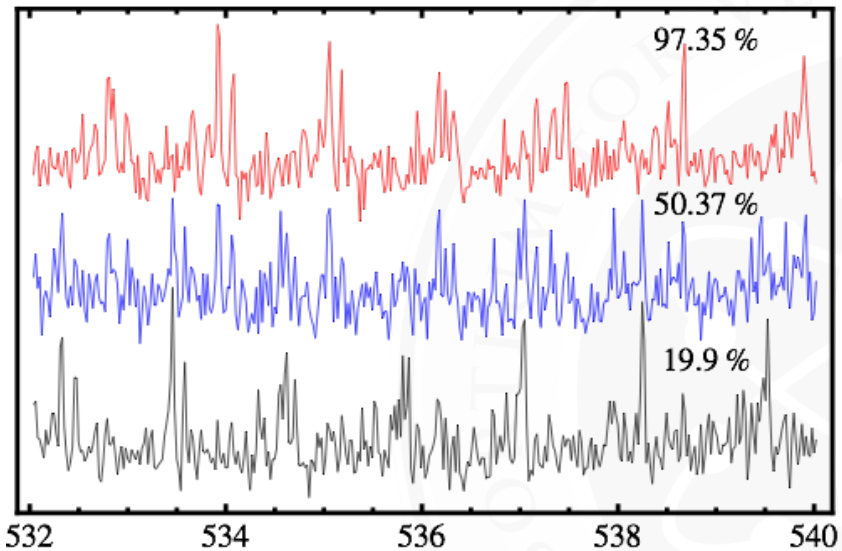


R. Russo, *et* al., Spectrochimica Acta B. 66, 99 (2011)

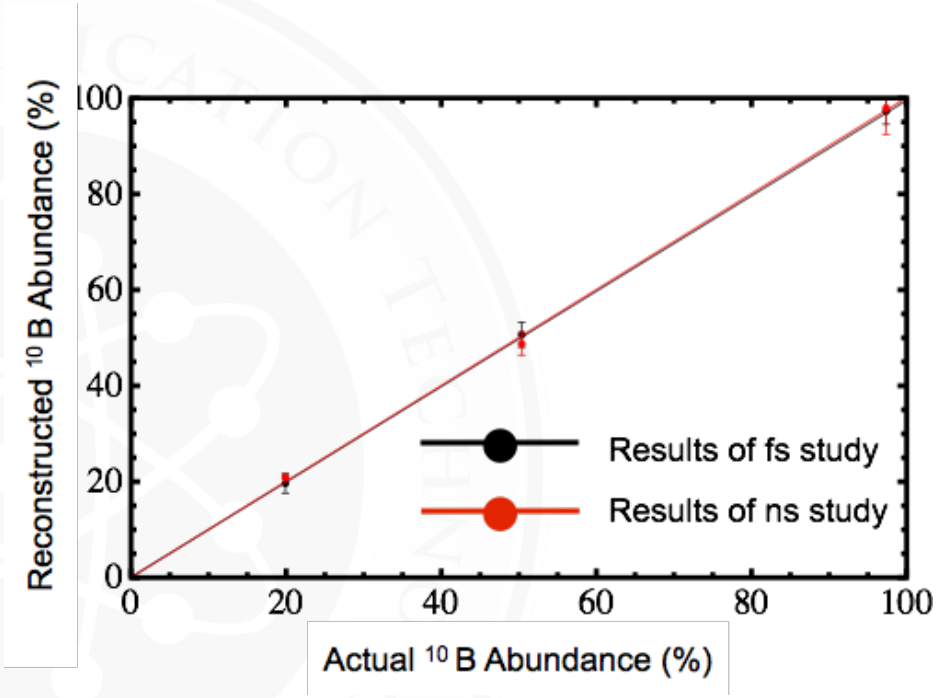




Molecular isotopic measurements are possible even without time gating



Emission spectra of boron oxide from ns-laser pulses

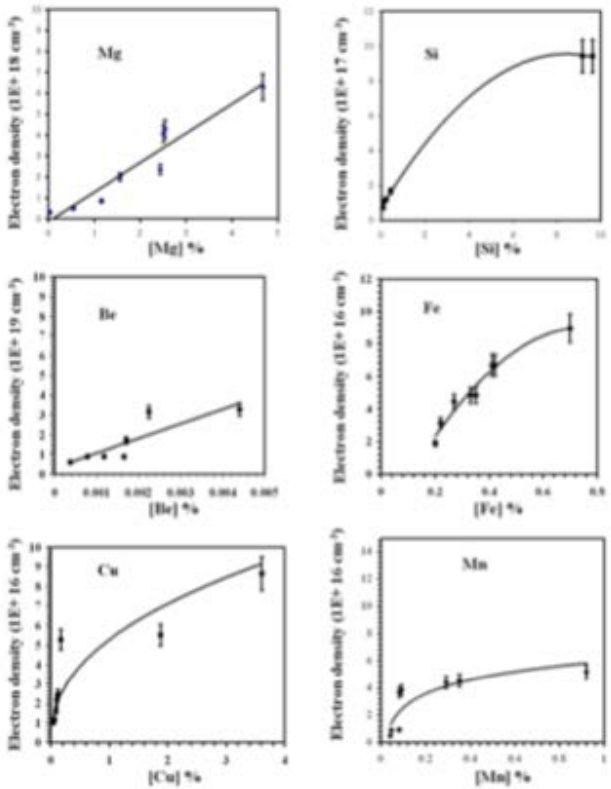


Reconstructed boron oxide abundance vs. actual abundance

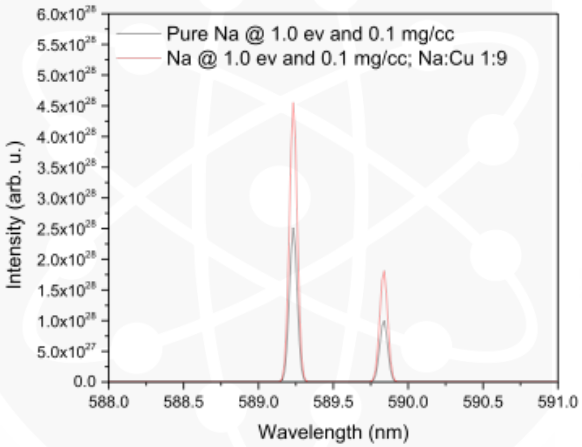
P. Ko and Jovanovic (2013) Spectrochimica Acta, Part B, 90, pp. 68-71.
B. Yee, K. C. Hartig, K. C., P. Ko, J. P. McNutt, and I. Jovanovic, I. (2013) Spectrochimica Acta, Part B, 66, pp. 72–76.

Matrix effects can be an obstacle for quantitative measurements

- Different elemental mixes produce plasmas with different emission properties

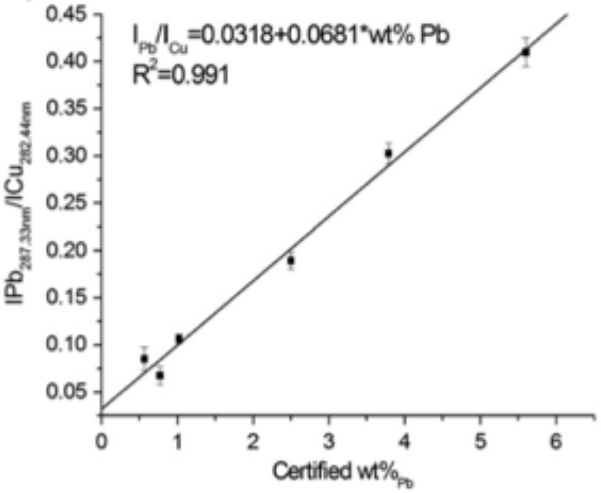


LANL Atomic Modeling



- Na matrix effect: adding Cu increases Na emission

- Quantitative analysis currently relies on matrix matched known samples



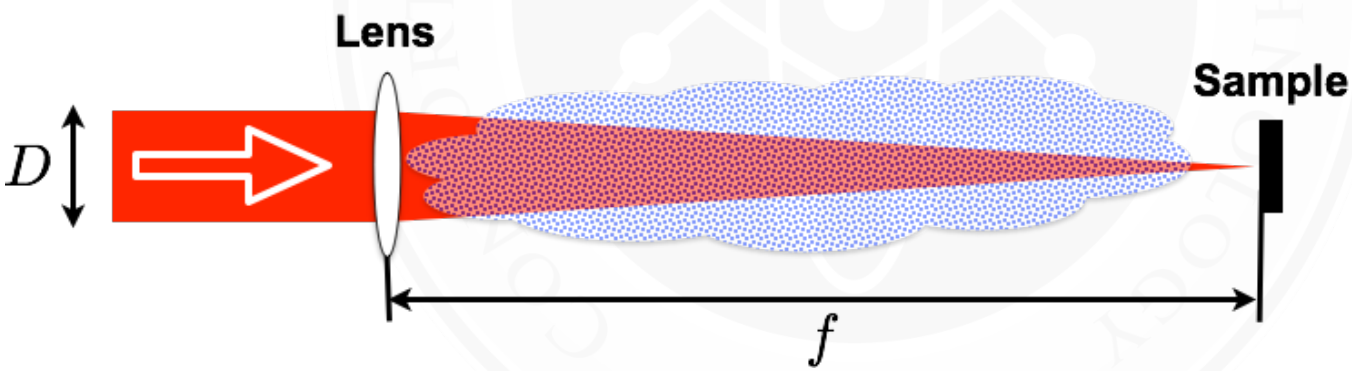
Walid Tawfik. The Matrix Effect on the Plasma Characterization of Six Elements in Aluminum Alloys

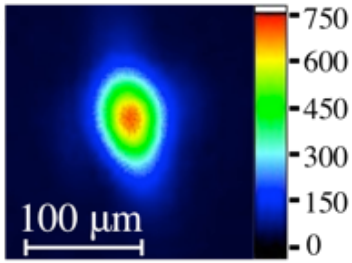
Large standoff distances present multiple challenges for LIBS

Performing LIBS at large standoff distances leads to two main challenges associated with beam delivery:

- prohibitively large optics
- absorption and atmospheric turbulence

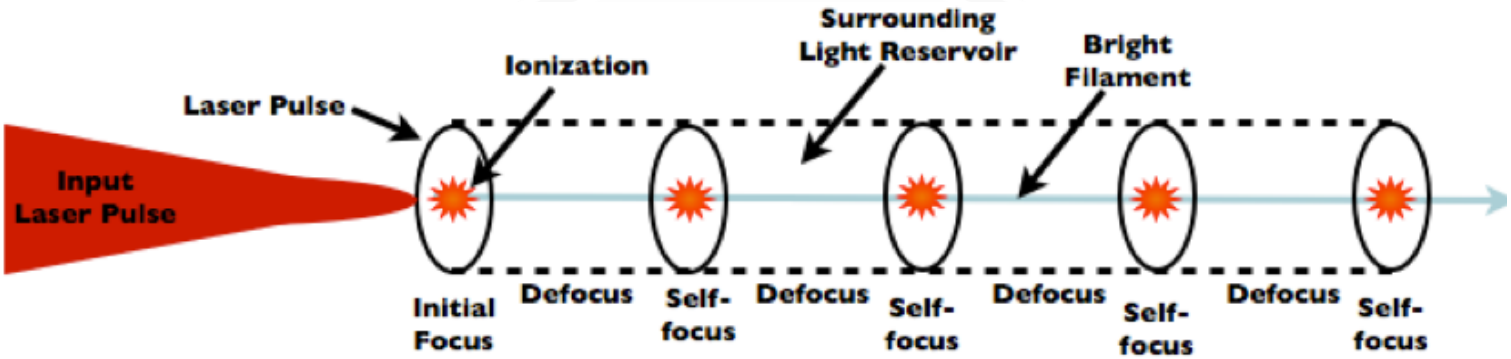
Spot size $w_0 \geq 1.22\ \lambda f / D$

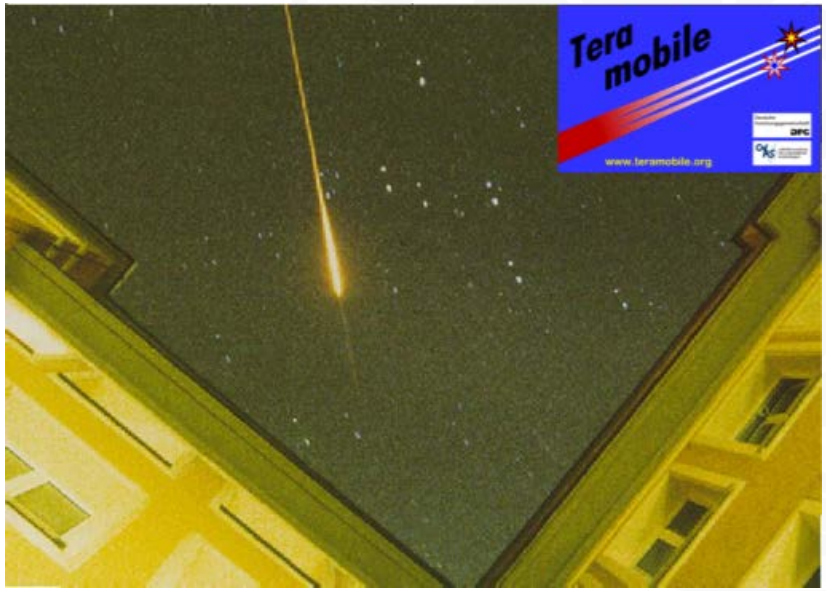




Filamentation induced by high-power ultrashort pulses can be used to overcome those limitations.

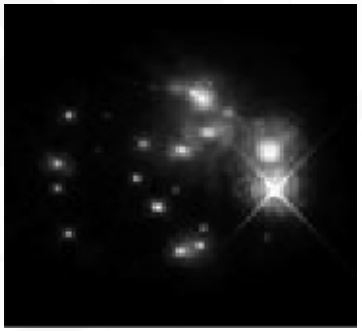
High-power femtosecond pulses can undergo self-focusing in air and induce plasma filaments





Self-focusing "lensing" effect

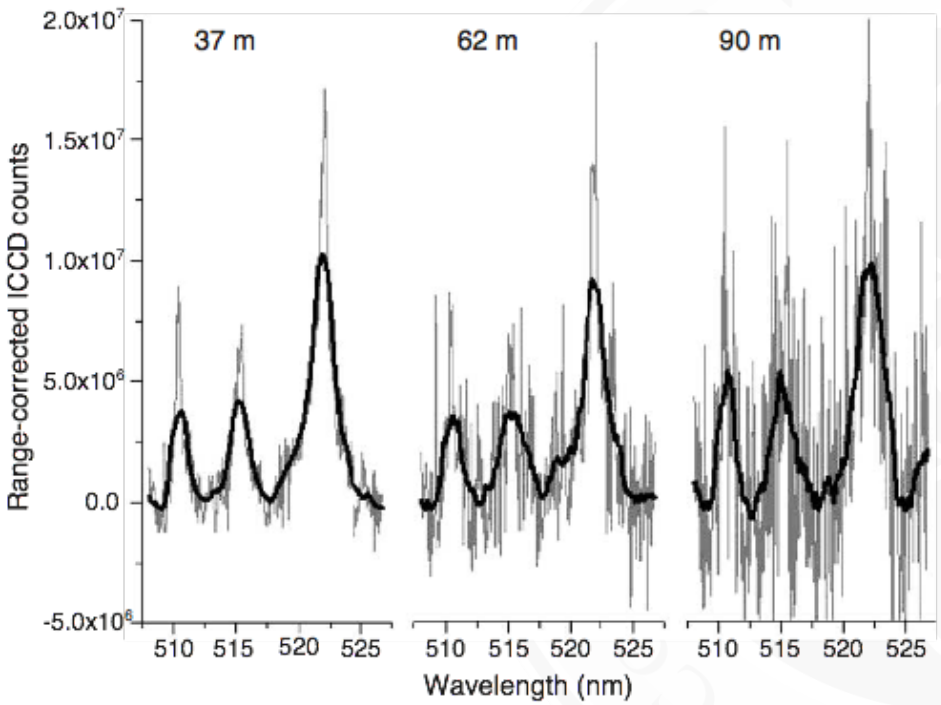
$$n = n_0 + n_2 I$$



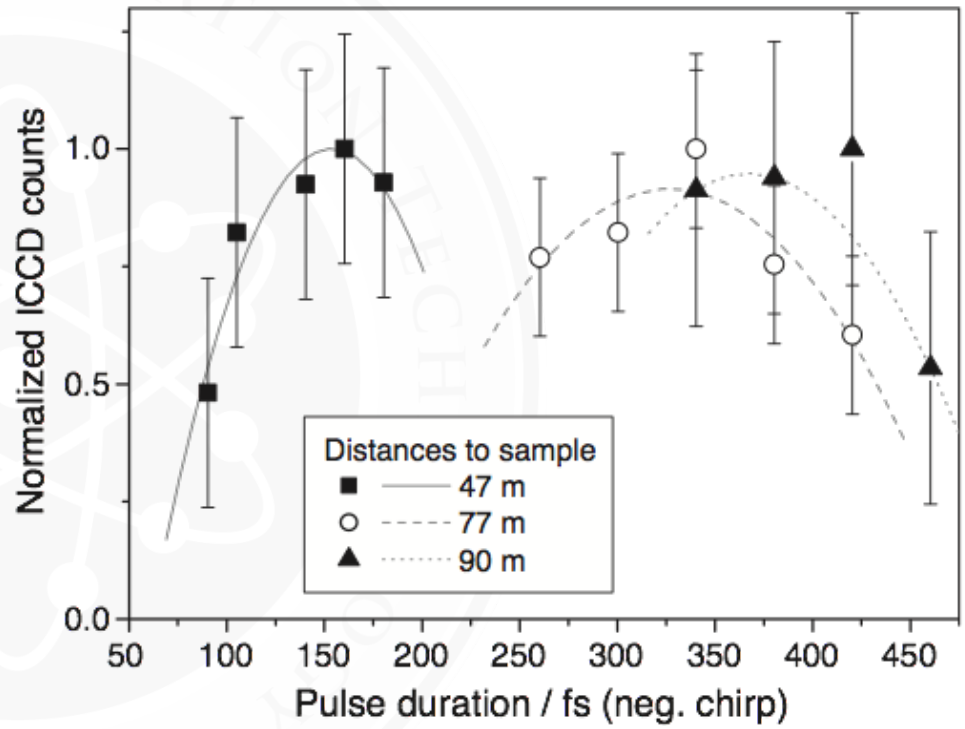
Multi-filamentation

Elemental detection and analysis capabilities of F-LIBS have been demonstrated on Cu

Background and range-corrected spectra



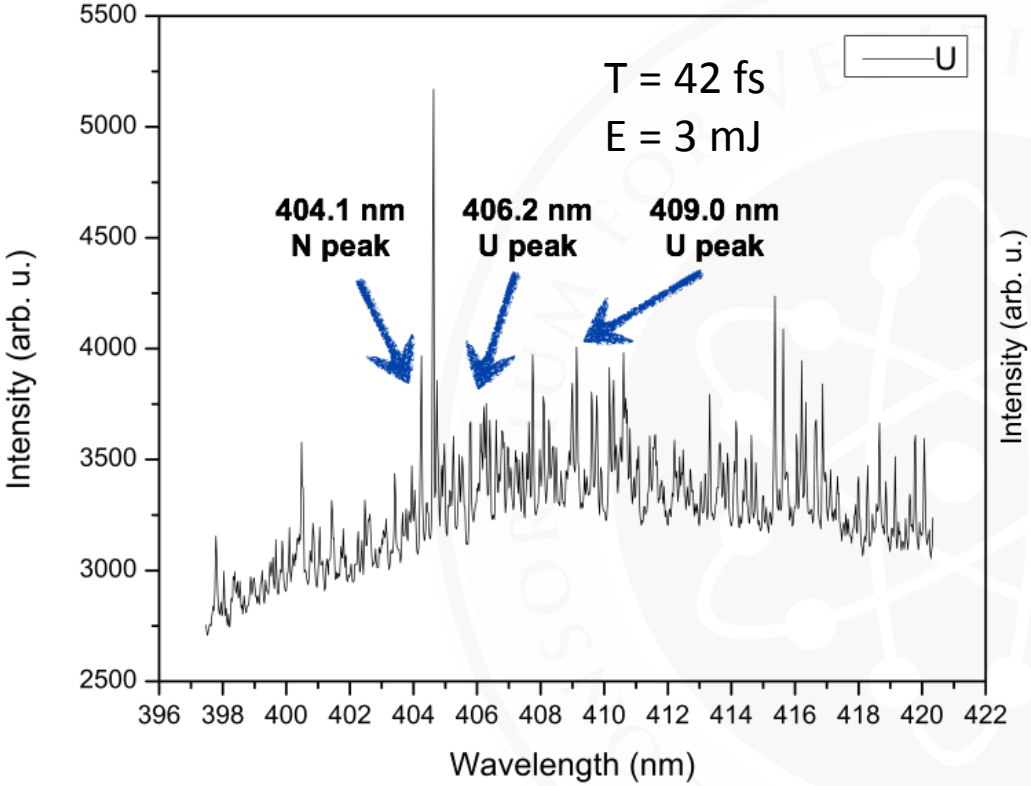
Chirp optimization

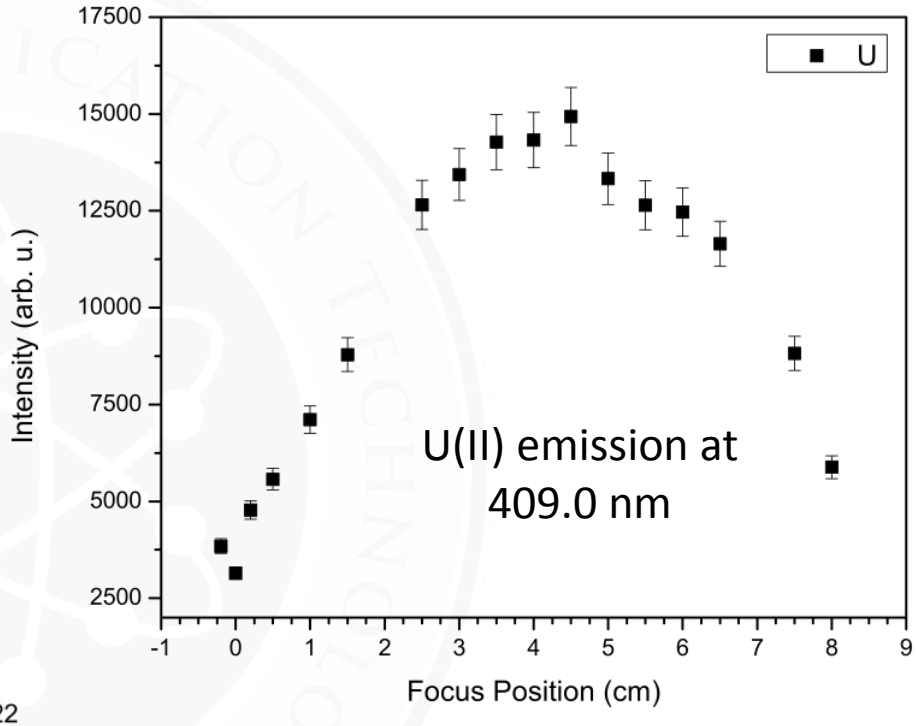


Ph. Rohwetter *et al.*, Spectrochimica Acta Part B 60,1025 (2005)

We will study the potential of this technique for measurements performed on nuclear materials.

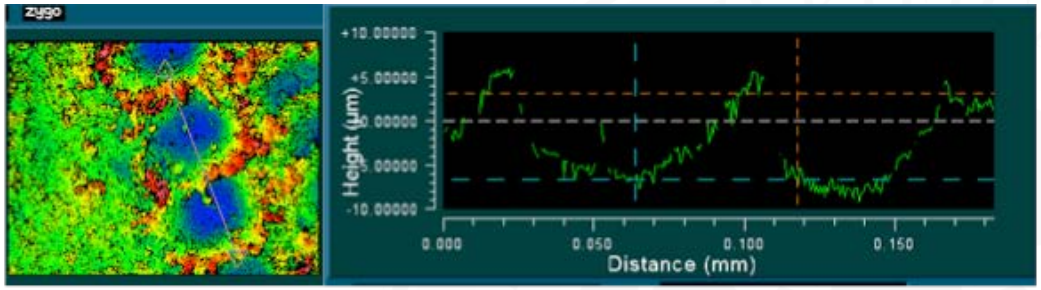
Optimization of filamentation LIBS



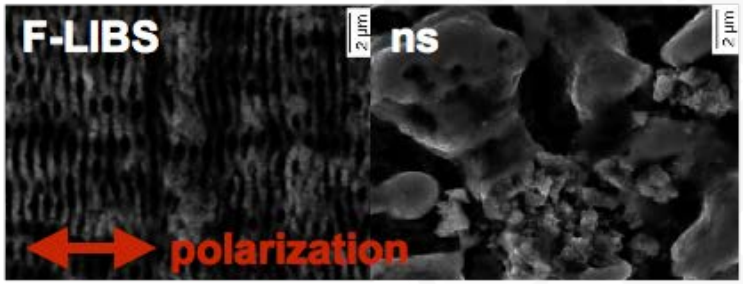


- up to 4x improvement in LIBS signal intensity can be obtained by use of filamentation compared to conventional fs-LIBS
- optimal filament length that produces maximum emission: 50-60% of the maximum filament length

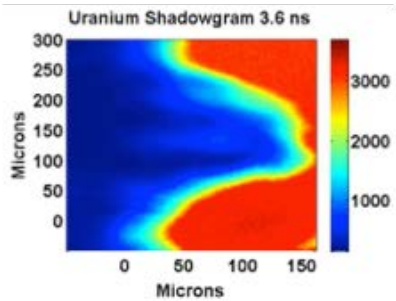
Interactions of filaments with surfaces are not well understood and will be studied

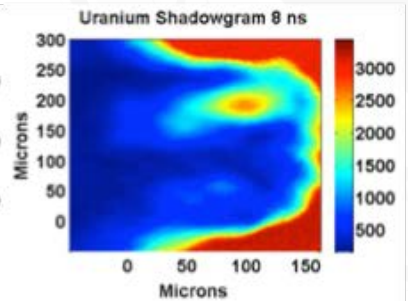


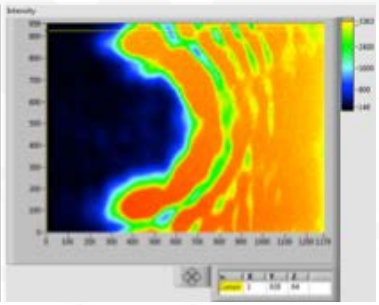
Uranium surface ablated by a series of 100 fs laser shots per spot. Characterization was performed using a white light profilometer (Zygo).



SEM characterization of Cu
(Ph. Rohwetter et al, 2006)
Quasi-periodic sub-wavelength structure (interference between incoming and anisotropically scattered light from surface)



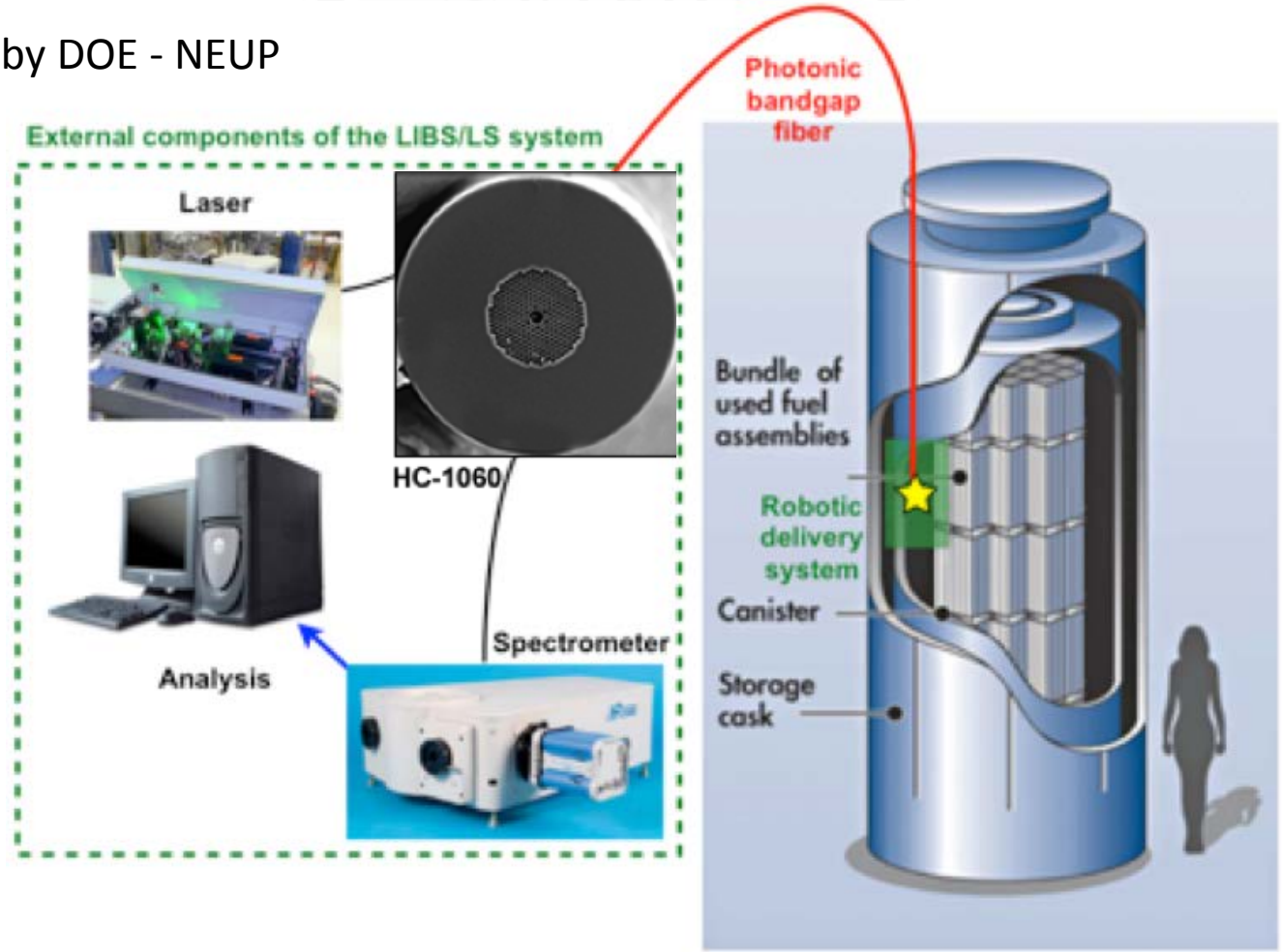




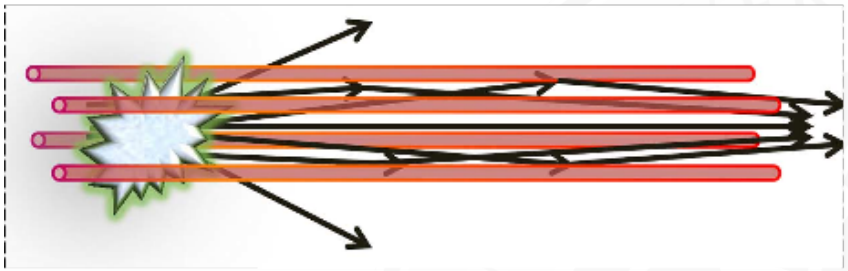
Picosecond shadowgraphy and ex-situ surface characterization will be used to help understand surface interactions.

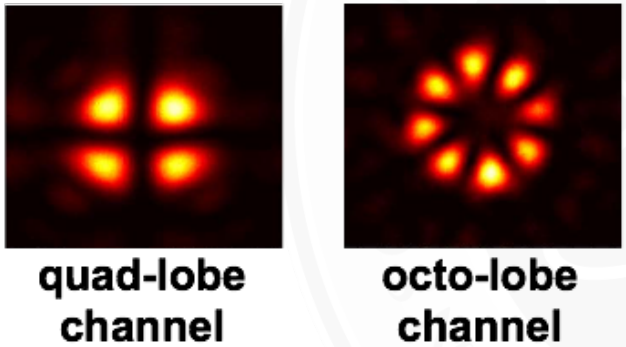
Standoff measurements utilizing a non-intrusive optical probe

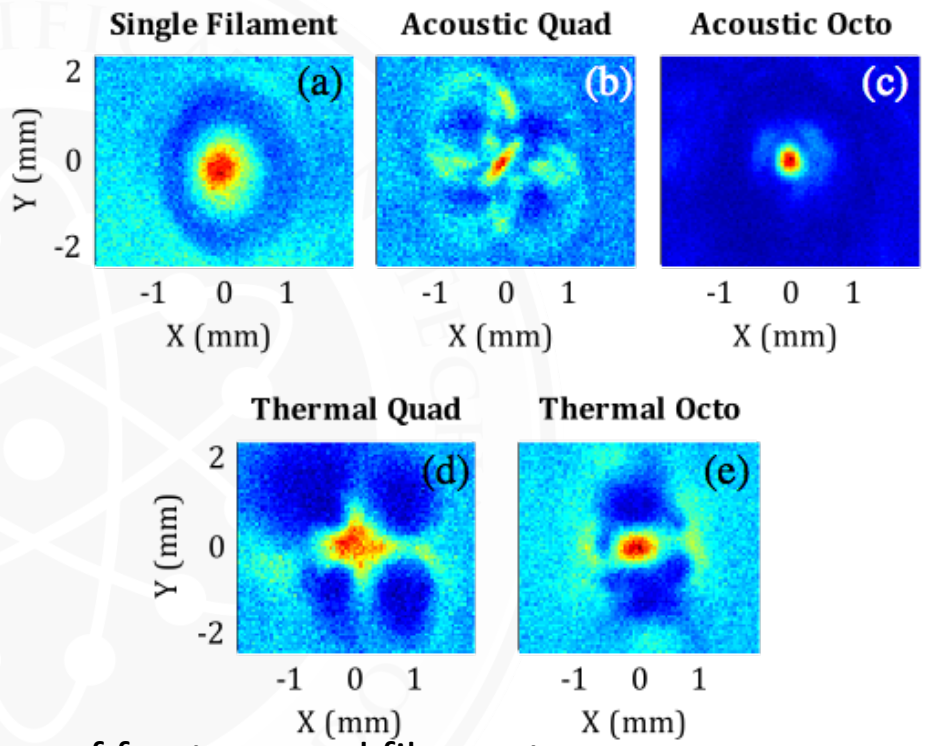
Supported by DOE - NEUP



The problem of isotropic return signal persists, but new solutions may be on the horizon







- Long-lived air guide is formed by an array of femtosecond filaments.
- Increase of collected signal by 50% over 1 m demonstrated, implying a potential for 10^4 enhancement over 100 m.

E. W. Rosenthal et al., Optica 1, 5 (2014)

We have already secured material access for experiments at PSU

- Recently licensed by the NRC to posses and use SNM and other hard-to-get materials in LIBS experiments

Inventory

- 9.8 g 93.2% HEU metal in 4 Zr-U fuel plates
- 0.3 g 7.2% U-235 in UO2 Pathfinder fuel pellets
- Np-237 electroplated metal sample
- Multiple Th and natural U metal strips

Fuel Plates

Np metal

Natural U metal

Fuel Pellets

Conclusion

Primary research topics to be addressed during this program:

- Feasibility and characteristics of isotopic measurements at standoff via filamentation LIBS
- Improved understanding of matrix effects via experiments and first-principles modeling (LBNL, LANL, INL collaborations).

Please be sure to check out the poster presented by PSU graduate student Kyle Hartig.

Acknowledgements:

- Prior related work and infrastructure development supported by DHS/DNDO, DOE/NNSA, and DTRA.
- Penn State ES&H, Radiation Science and Engineering Center
- Nuclear Regulatory Commission