High Resolution Spectroscopy of Laser-Induced Uranium Plasma

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Introduction

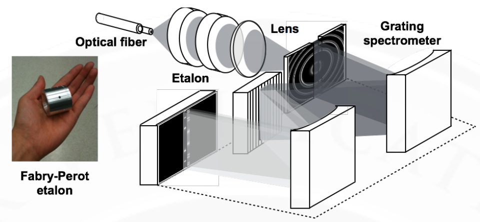
Abstract

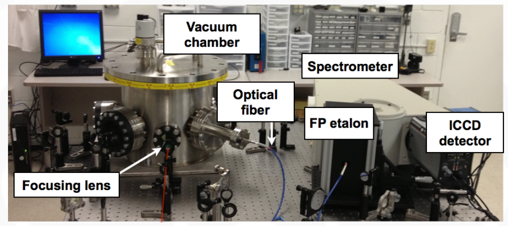
Laser-induced breakdown spectroscopy measures the optical emission spectra of laser-pulse generated plasma, and is capable of rapid analysis in the field. Isotopic measurements of nuclear material have been demonstrated using LIBS in recent years. Such high-resolution measurements usually require sizeable and costly components. Alternatively, a hybrid instrument consisting of an inexpensive, compact Fabry-Perot (FP) etalon coupled to a grating spectrometer is used to measure LIBS spectra. Reconstruction of the high-resolution spectra from the spatially encoded angular interference pattern of the etalon is experimentally demonstrated in this study. The results of this work could lead to the development of a new high-resolution LIBS instrument for isotopic measurements of nuclear material.

Objectives

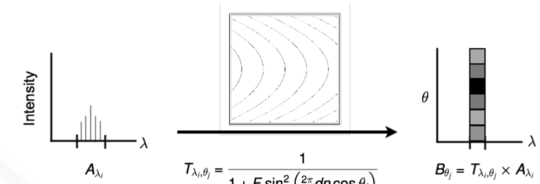
- **Improve laboratory instrumentation for physical and chemical analysis of nuclear materials.**
- **Develop a new analytical method for analyzing experimentally collected data.**
- **Design a high-resolution LIBS instrument for fingerprinting nuclear material for nuclear nonproliferation, safeguards, and verification applications.**

Principle of operation and experimental setup





Analytical model of the instrument



A_{λ_i} $\quad T_{\lambda_i,\theta_j} = \dfrac{1}{1 + F\sin^2\left(\frac{2\pi}{\lambda_i} dn\cos\theta_j\right)}$ $\quad B_{\theta_j} = T_{\lambda_i,\theta_j} \times A_{\lambda_i}$

$$F = \left(\frac{2r}{1-r^2}\right)^2 \qquad \Delta_{phase} = \frac{2\pi d}{\lambda} n\cos\theta \qquad T_{\lambda,\theta} = \frac{1}{1 + F\sin^2 \Delta_{phase}}$$

Coefficient of finesse $r^2 = R$ (reflectivity) — **Phase difference** — **Transmission equation of the etalon (Airy function)**

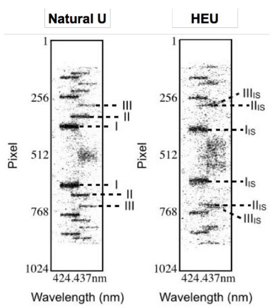
$$\begin{bmatrix} B_{\theta_1} \\ B_{\theta_2} \\ B_{\theta_3} \end{bmatrix} = \begin{bmatrix} T_{\lambda_1,\theta_1} & T_{\lambda_2,\theta_1} & T_{\lambda_3,\theta_1} \\ T_{\lambda_1,\theta_2} & T_{\lambda_2,\theta_2} & T_{\lambda_3,\theta_2} \\ T_{\lambda_1,\theta_3} & T_{\lambda_2,\theta_3} & T_{\lambda_3,\theta_3} \end{bmatrix} \begin{bmatrix} A_{\lambda_1} \\ A_{\lambda_2} \\ A_{\lambda_3} \end{bmatrix}$$

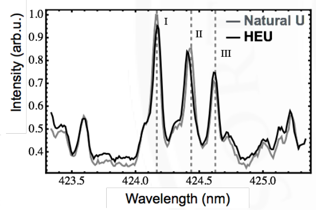
2D transmission function maps wavelengths to angles.

$$B_{\theta_1} = T_{\lambda_1,\theta_1}A_{\lambda_1} + T_{\lambda_2,\theta_1}A_{\lambda_2} + T_{\lambda_3,\theta_1}A_{\lambda_3}$$

Measurements of uranium have been performed

Naturally (0.7% ^{235}U) and highly enriched (93% ^{235}U) uranium measured using grating spectrometer with (below) and without (right) FP etalon

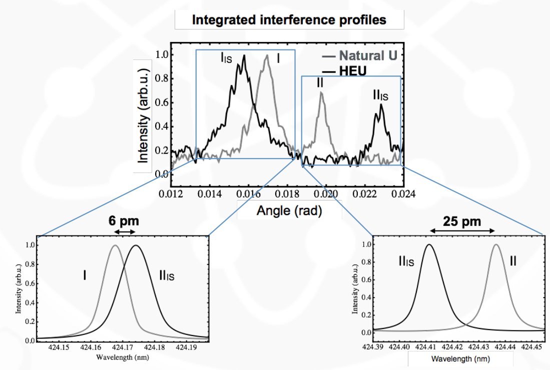




U spectral lines and isotope shifts (IS)

^{238}U	λ (nm)	^{235}U	λ (nm)	IS (pm)
I	424.167	I_{IS}	424.173	-6
II	424.437	II_{IS}	424.412	25
III	424.626	III_{IS}	424.621	5

Isotopic selectivity has been demonstrated

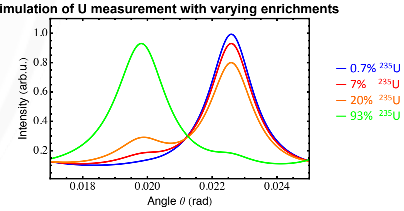


Reconstructed spectra of highly enriched 93.2% ^{235}U (black) and natural 99.3% ^{238}U (gray) uranium, distinguishing the isotope shift

Discussion and future work

- **There is an ongoing need to distinguish among uranium isotopes, which enables the measurement of uranium enrichment.**
- **A method of data analysis was developed that enables rapid reconstruction of high-resolution spectra measured using a hybrid interferometric/ dispersive spectrometer.**
- **The device was capable of resolving the isotope shift of uranium atomic lines not resolvable by the grating spectrometer alone.**

Simulation of U measurement with varying enrichments



Future Work

- **Determine sensitivity of detection method using calibration standards.**
- **Test hybrid spectrometer with filamentation LIBS measurements.**
- **Simulate multiple U lines for optimizing isotopic analysis.**
- **Design a compact prototype of a high-resolution instrument.**

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National Nuclear Security Administration