Lawrence Livermore National Laboratory

Multi-actinide resonance ionization mass spectrometry (RIMS) for Nuclear Forensics Applications and Rapid Response

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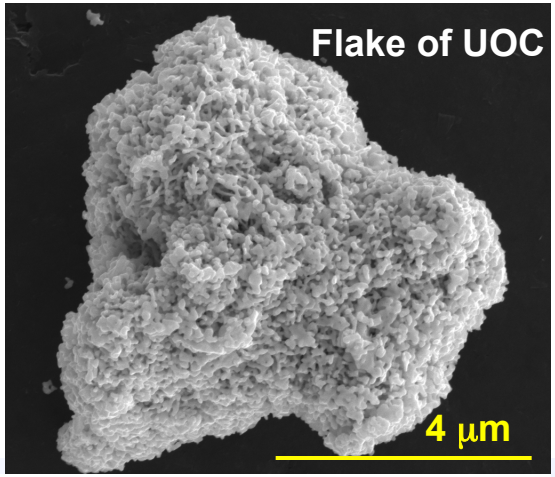
This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344

LLNL-PRES-516915

Nuclear forensics: What do we mean?

- analysis of interdicted illicit nuclear or radioactive material and associated materials to provide evidence for nuclear attribution
- the **goal** is identification of forensic indicators in interdicted nuclear and radiological samples or the surrounding environment, **linking people** to **places**, **materials**, **events**

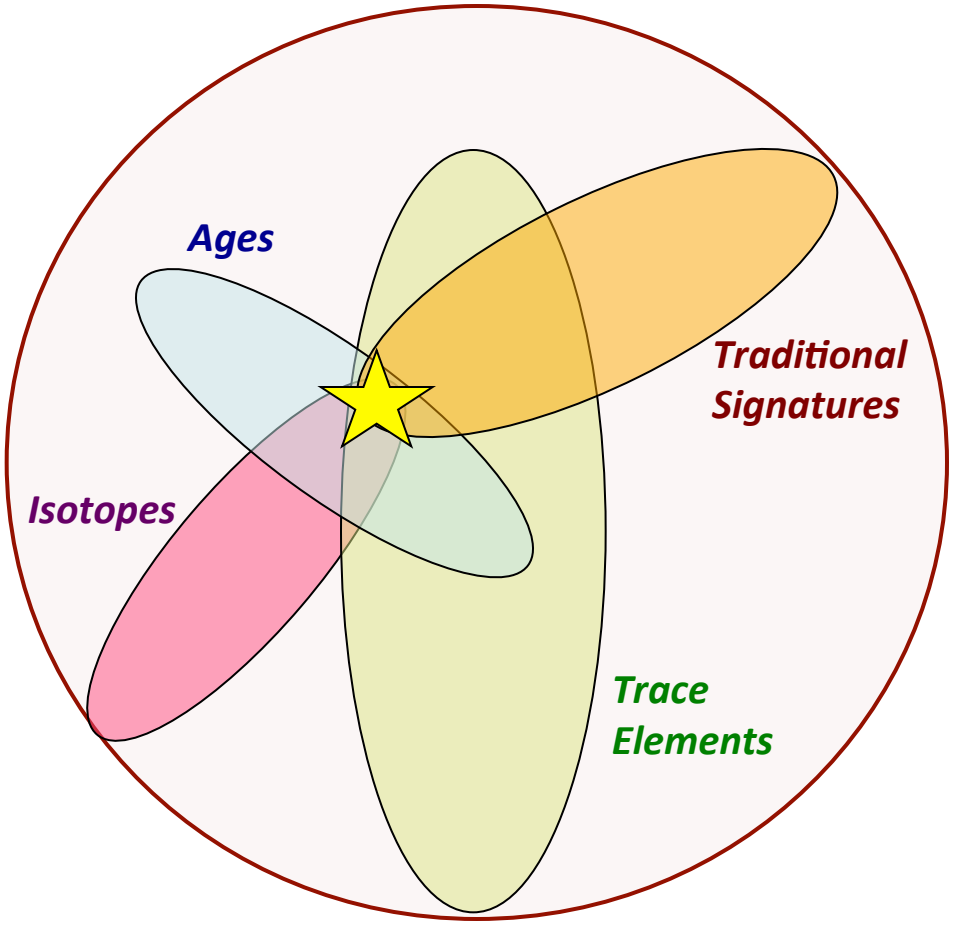
Measuring nuclear materials: One size does not fit all



Analytical considerations:

- Sample size?
- Spatial resolution?
- Homogenous or heterogeneous samples?
- Analytical location?
- Desired precision/ accuracy?
- Time frame?

There is no one path to an answer: Multiple signatures build attribution confidence

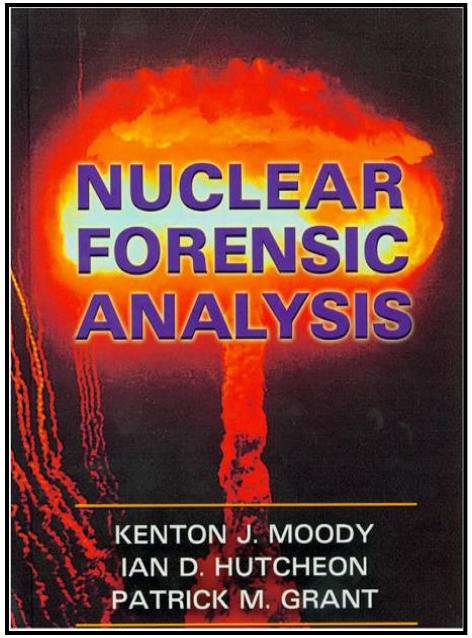


Primary tools for characterization of nuclear materials:

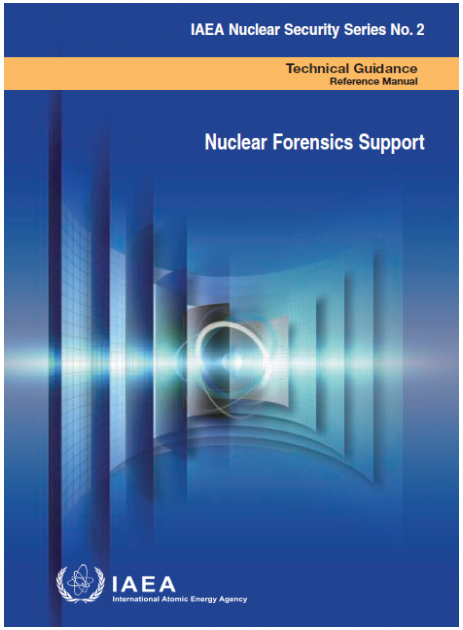
- **Radiation detection / counting**
  - Alpha (α), gamma (γ), beta (β^-)
  - Selective isotopic information
- **Excitation/stimulation**
  - X-ray generation
  - Atomic absorption and emission
  - Limited analysis of complex samples and trace concentrations
- **Ion formation (mass spectrometry)**
  - Any element and isotope
  - range of concentrations, spatial resolutions
  - **Generally higher precision, but time consuming sample preparation**

Moving capabilities forward: Nuclear Forensic Research

- Improve answers?
  - Precision and Accuracy
- Faster analyses?
- Smaller sample sizes?
- Fewer people needed?
- Reliable data processing?



*Reference text authored at LLNL, **2005***



*IAEA document by Kristo (LLNL), Smith (LLNL), Niemeyer (LLNL), and Dudder (PNNL), **2006***

U and Pu-isotopes contain critical clues about material origins, processes and intent

Actinide isotope measurements demand speed, precision and accuracy, rigorous conclusions

Resonance Ionization:
A Faster Path to a Better Answer

All Mass Spectrometers Work The Same Way:

- Make ions from the element(s) of interest
- Separate ions in space or time
- Detect the ions as separated masses

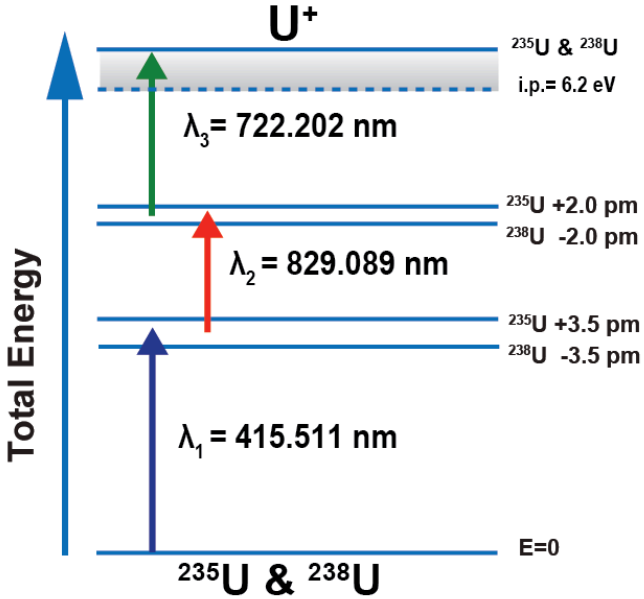
Resonance ionization uses intermediate electronic states specific to each element to form ions!

provide the energy

discrete photon energy

sufficient photon fluence

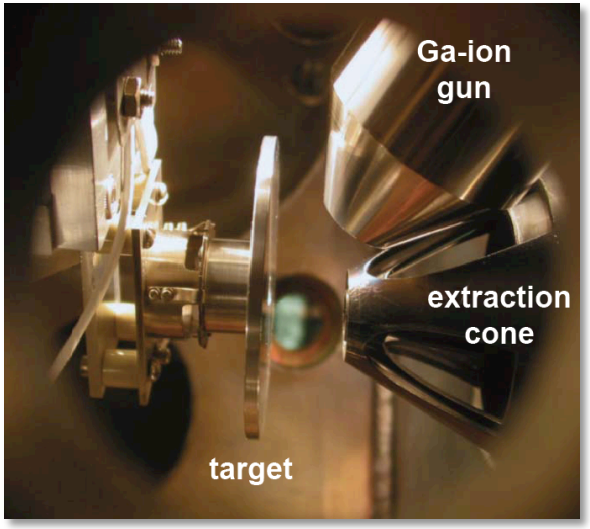
precision, timing, control

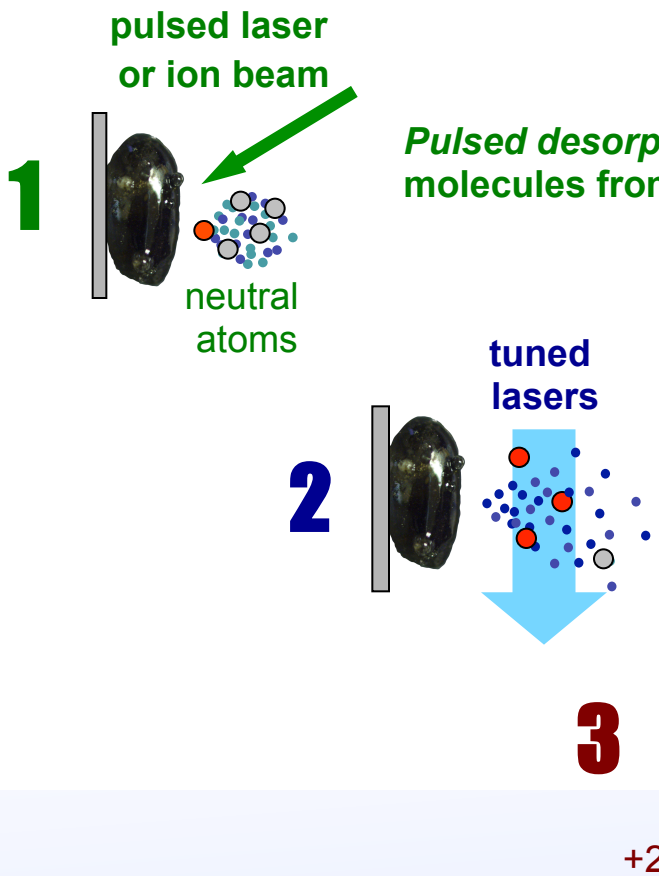


Almost any element can be ionized through this process!

RIMS performs chemical separation in the instrument

Resonance ionization mass spectrometry measures actinide isotopes using selective laser ionization for chemical separation during analysis





Pulsed desorption event **removes atoms & molecules from solid materials**

One element absorbs the *resonance and ionization lasers*, the rest are transparent

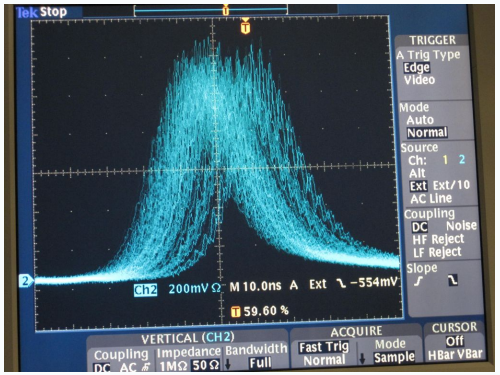
The photo-ions are accelerated to the *mass analyzer / detector*

Laser Performance

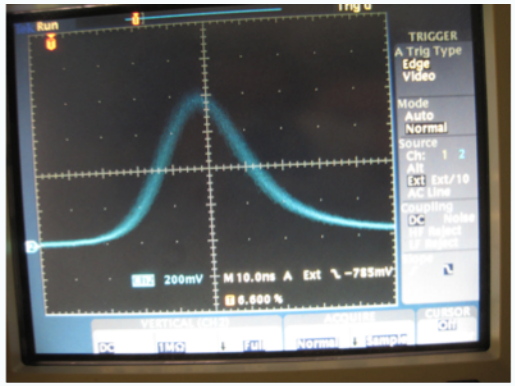
Real lasers vary in:

- Wavelength
- Position
- Power
- Time

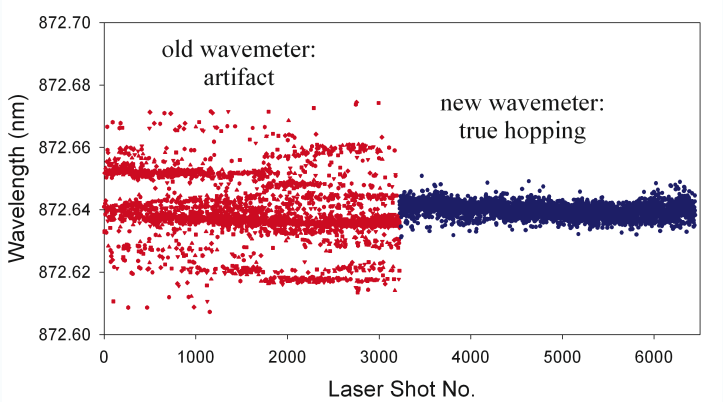
Time Variation from Pulse to Pulse:



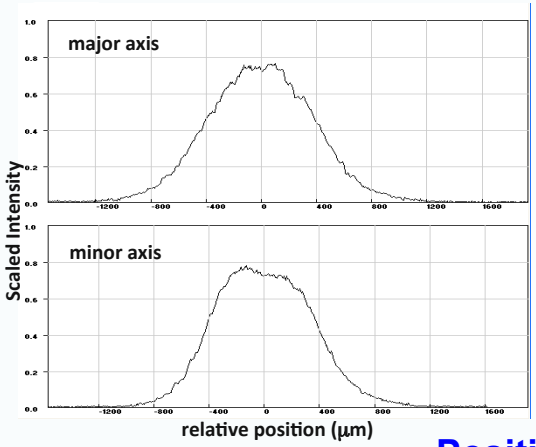
Old laser: 14 ns FWHM

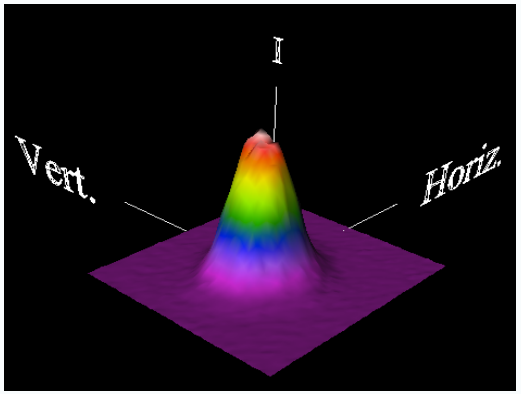


New laser: 4 ns FWHM



Wavelength Variation

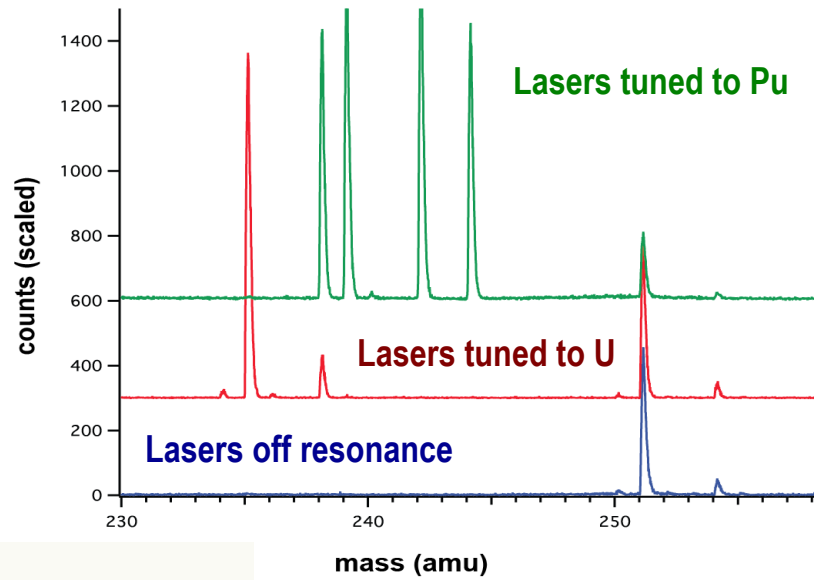


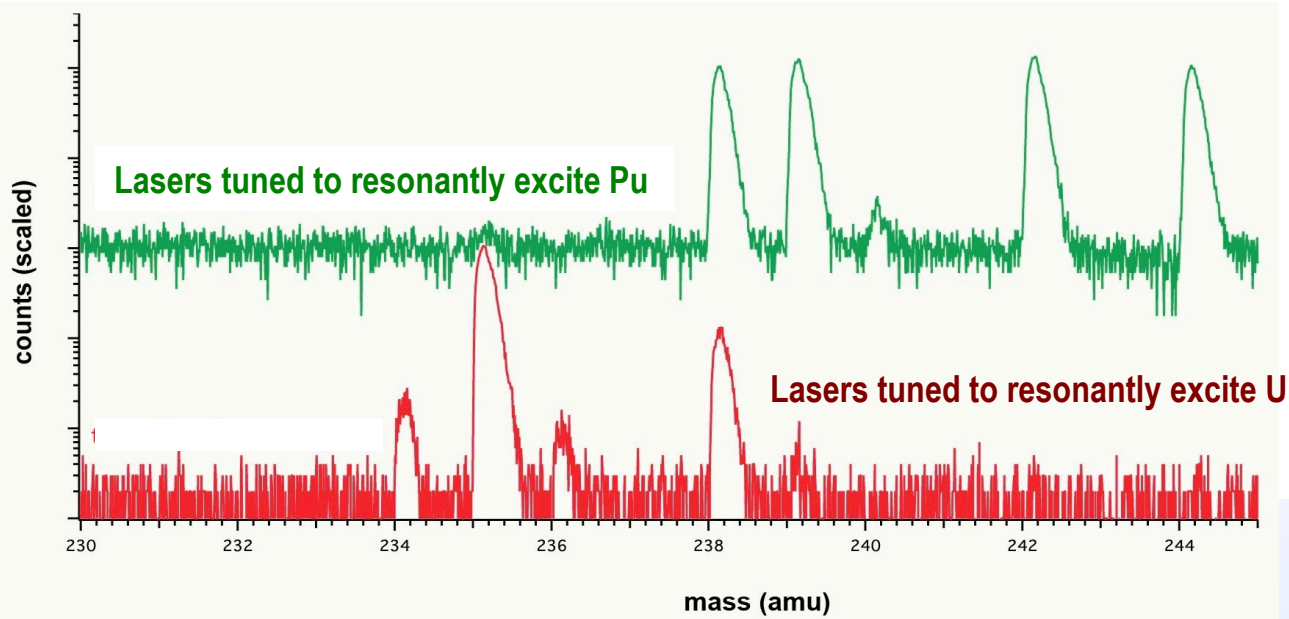


Position

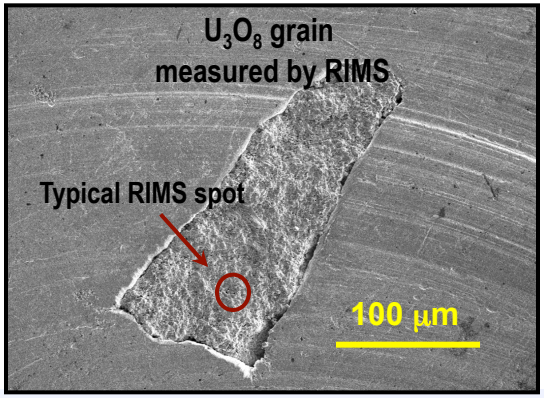
RIMS demonstrates discrimination between U and Pu from the same sample

- ^{238}U and ^{238}Pu would normally have to be separated chemically, prior to mass spectrometry
- **RIMS does this separation in the instrument**
- Tuned lasers ionize the desired element for analysis from a multi-actinide sample

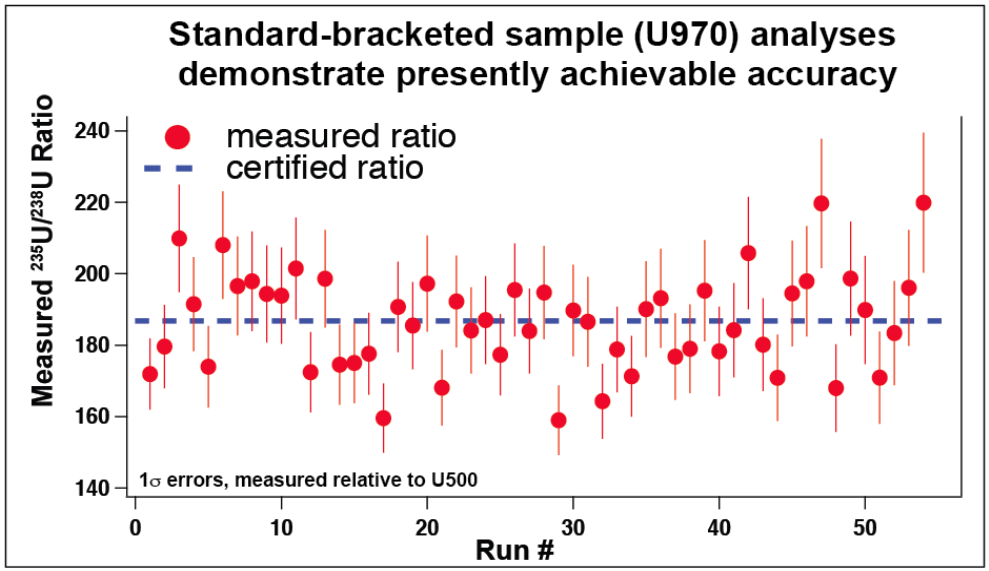




Secondary electron image



Actinide isotope ratios have been measured accurately in metal, oxide and glass samples



CRM970 (U_3O_8)

$^{235}U/^{238}U_{cert}$ = 186.77

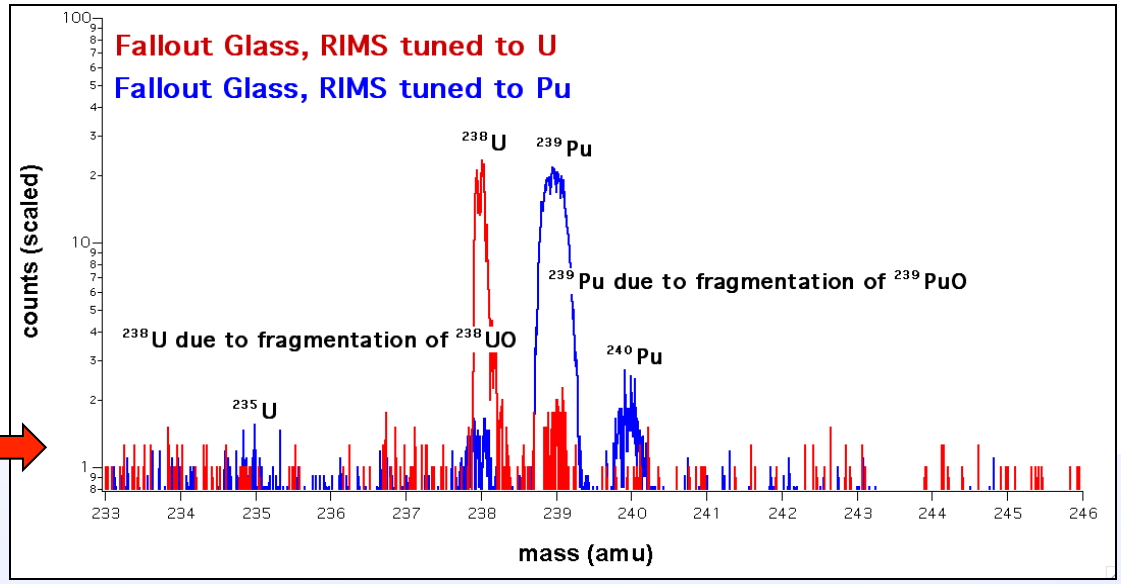
$^{235}U/^{238}U_{meas}$ = 185.16

(1σ) ± 1.76

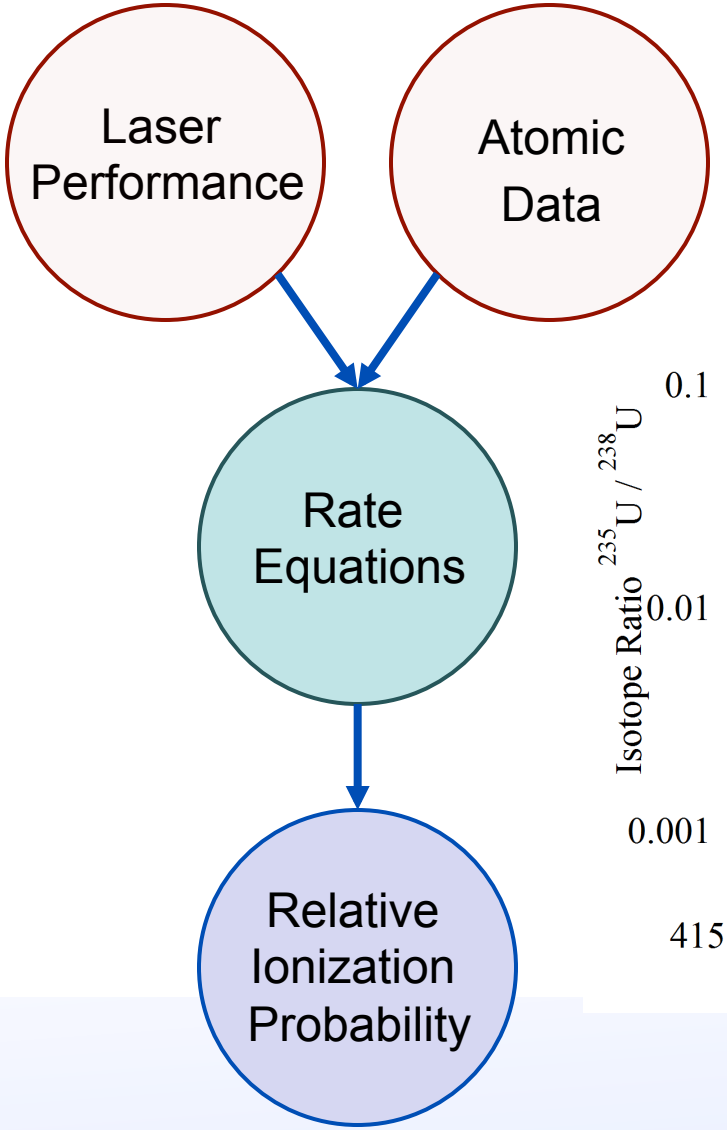
χ^2 = 1.10

- We have measured isotope ratios over seven orders of magnitude
- Completed complementary U and Pu analysis in ~2 hours or less

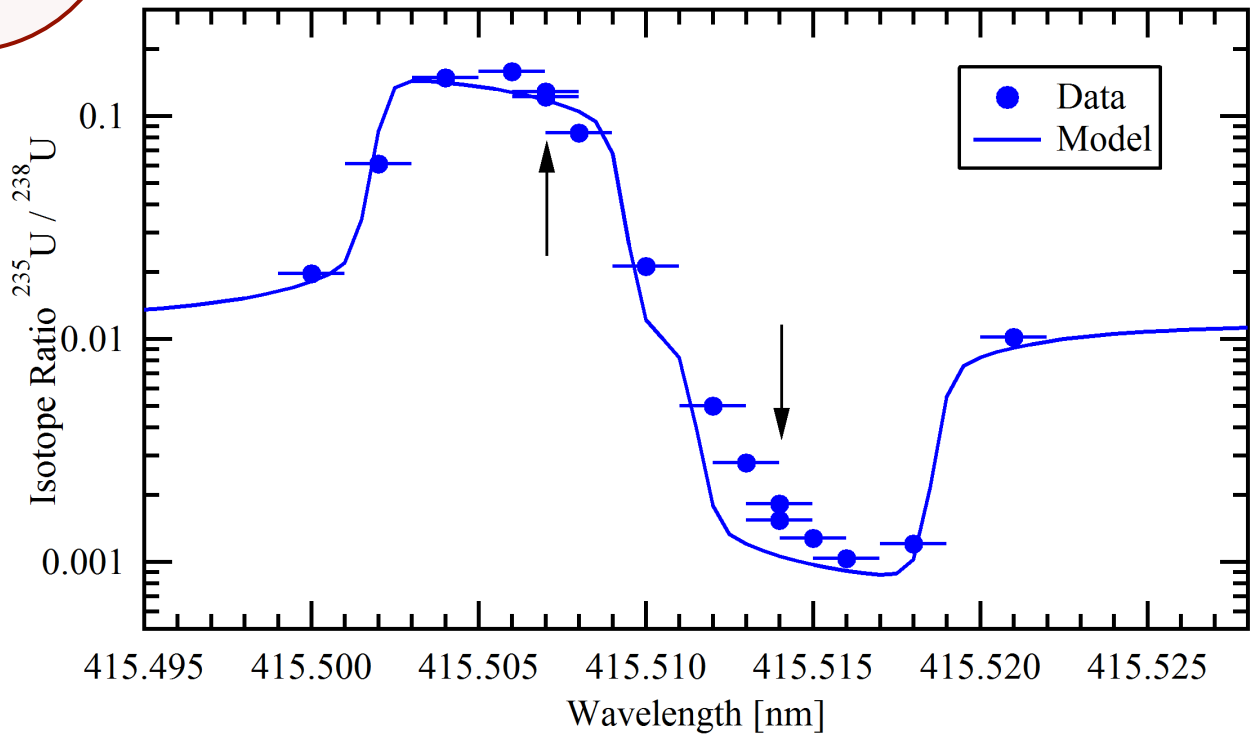
Preliminary results demonstrate detection of actinides in fallout debris samples with part-per-million concentrations of actinides



Model: Predicting Ionization Behavior of Uranium



Variation of Measured Isotope Ratio as a Function of Laser Wavelength in the First Transition



Sustaining and advancing nuclear forensic capabilities

Nuclear forensics is an emerging science

By necessity, it draws on a team of people to research and innovate, provide process knowledge and materials experience, and perform technical analyses

Research → **Application** ← **Experience**

Over 5 years, this application of the RIMS methodology has advanced from concept to demonstrated multi-element technique for measuring isotope ratios in actinides without chemical separation.