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A World without Sample Preparation: Developing Rapid Uranium Isotope Measurement Capabilities by Resonance Ionization Mass Spectrometry (RIMS)

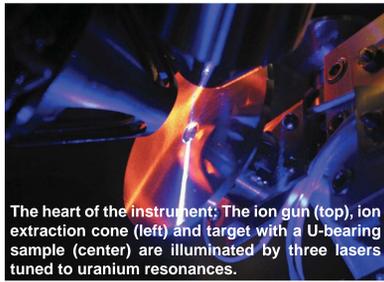
K. B. Knight, I. D. Hutcheon, B. H. Issehardt, M.
R. Savina, S. G. Prussin

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The heart of the instrument: The ion gun (top), ion extraction cone (left) and target with a U-bearing sample (center) are illuminated by three lasers tuned to uranium resonances.

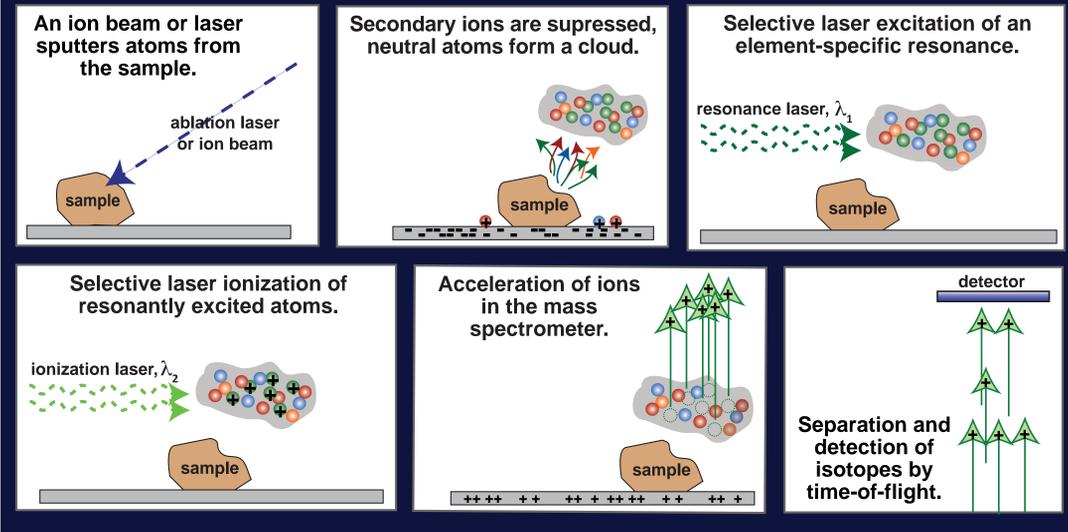
A World without Sample Preparation: Developing Rapid Uranium Isotope Measurement Capabilities by Resonance Ionization Mass Spectrometry (RIMS)

K.B. Knight¹ with I.D. Hutcheon¹, B.H. Isselhardt^{1,3}, M.R. Savina², S.G. Prussin³



We are developing highly sensitive, highly discriminating laser-based techniques for rapid determination of isotopic compositions. Rapid command of such information is critical to assessment of the origin and history of nuclear materials, particularly in post-detonation scenarios.

How does RIMS work?



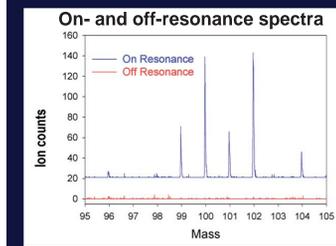
Collaborative Science

Actinide isotopic compositions provide insight into the origin and history of nuclear materials. Such information is critical in post-detonation scenarios, when rapid delivery of information is vital, as well as pre-detonation scenarios and environmental work, where trace concentrations of materials and/or large environmental backgrounds create challenging analytical conditions.

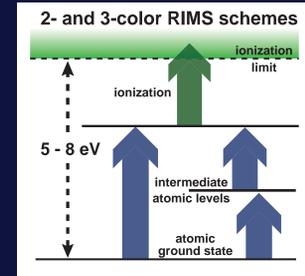
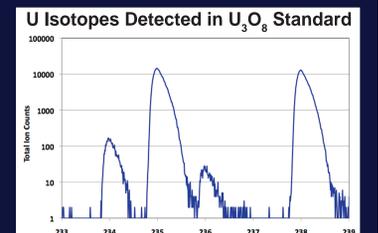
Using state-of-the-art RIMS instruments, designed and built at Argonne National Laboratory, alongside the nuclear forensics expertise of Lawrence Livermore National Laboratory, we are carving a fresh path forward for isotopic analyses of nuclear materials.

Our collaboration is developing analytical methods aimed at providing rapid uranium isotopic information from solid samples, and has completed several crucial proof-of-concept measurements.

Sensitive, Selective Detection of Uranium



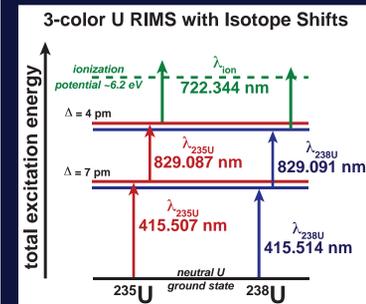
We have developed both 2- and 3-color RIMS to maximize uranium sensitivity and selectivity, while minimizing background (non-resonant) molecules and ions.



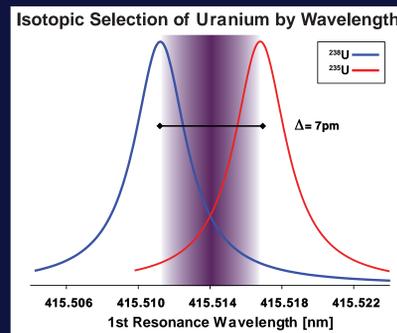
In RIMS, tuning lasers even slightly off-resonance (see Ru example, above) causes the signal to vanish. This provides unambiguous identification of the element of interest, and direct accounting of any residual background.

We have been successful in detecting four U isotopes in uranium metal, UO_2 , and U_3O_8 over a dynamic range of three orders of magnitude. Though preliminary, we have had similar success on U-bearing ores and silicates.

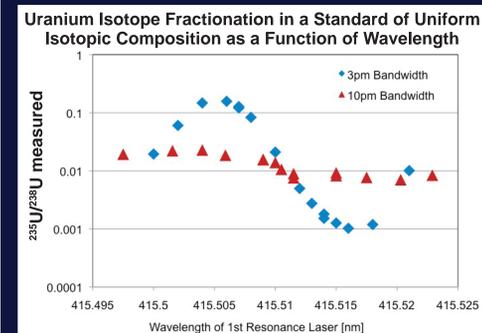
Laser Control is the Key!



Large isotope shifts, characteristic of deformed nuclei including the actinides, mean that very small shifts in wavelength create large isotopic fractionations!



All lasers 'jitter' in wavelength at some scale. Even 2-3 pm control of laser wavelengths, however, is not sufficient to achieve stable measurement of uranium isotope ratios with narrow bandwidth lasers.



Broadening the laser bandwidth from 3 pm (left, blue data) to 10 pm (left, red data) makes measured isotope ratios much more robust.

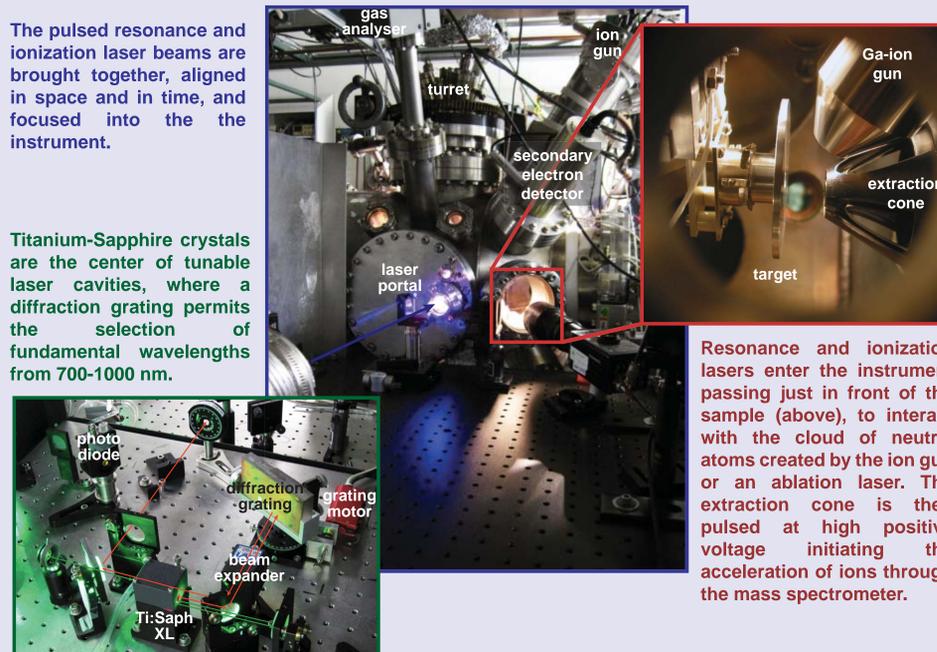
Further stabilization of narrow bandwidth lasers may allow preferential enhancement of minor isotopes, advantageous for sensitive detection of trace isotopes in samples with dynamic range issues.

CHARISMA

CHicago Argonne Resonance Ionization Spectrometer for Mass Analysis

The pulsed resonance and ionization laser beams are brought together, aligned in space and in time, and focused into the instrument.

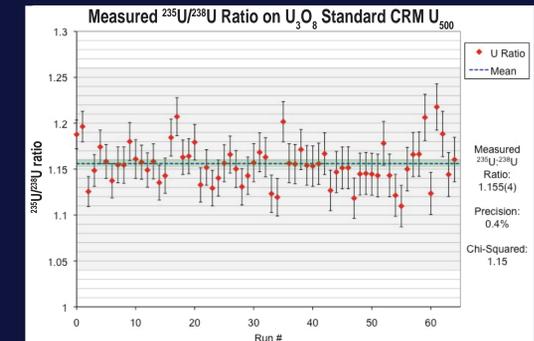
Titanium-Sapphire crystals are the center of tunable laser cavities, where a diffraction grating permits the selection of fundamental wavelengths from 700-1000 nm.



Resonance and ionization lasers enter the instrument passing just in front of the sample (above), to interact with the cloud of neutral atoms created by the ion gun or an ablation laser. The extraction cone is then pulsed at high positive voltage initiating the acceleration of ions through the mass spectrometer.

First Proof-of-Concept Measurement

Our initial exploration of uranium resonance ionization processes has enabled us to measure the $^{235}U/^{238}U$ ratio in a U_3O_8 standard to better than 0.5% precision, compared with a precision of ~10% errors just one year ago. We are continuing these experiments, and upgrading our lasers and equipment to make this measurement robust and routine.



Our next steps include plans to explore and optimize laser bandwidth schemes, the development of resonance ionization methods for Pu, and expanding measurements to "real world" samples such as complex oxides, debris and waste glasses.

Our 3-year goal is to provide U and Pu isotopic compositions to better than 1% precision in less than 4 hours from the time of sample receipt.

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