

Laser spectroscopy and ultra-trace analysis on lighter actinides- from Uranium to Curium

N. Kneip¹, F. Weber¹, Ch. E. Düllmann^{2,3,4}, C. Mokry^{2,3}, S. Raeder⁴, J. Runke^{2,4}, D. Studer¹, N. Trautmann² and K. Wendt¹

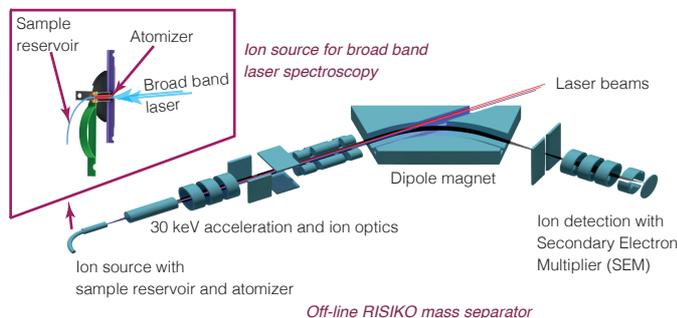
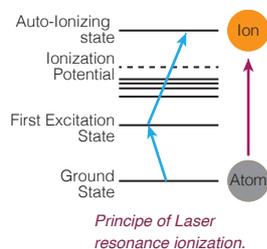
¹Institut für Physik, Johannes Gutenberg University, 55099 Mainz, Germany
²Department Chemie - Standort TRIGA, Johannes Gutenberg University, 55099 Mainz, Germany
³Helmholtz-Institut, 55099 Mainz, Germany
⁴GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt



Resonant laser spectroscopy in the actinides

Resonance Ionization Mass Spectrometry (RIMS) combines highly efficient and element-selective laser ionization with mass separation for the detection of single isotopes. Pulsed Ti:Sapphire lasers, developed at the Mainz University, are used for the stepwise excitation of valence electrons followed by the ionization of the atoms at the off-line RISIKO mass separator.

The RIMS is used for spectroscopy on rare isotopes and ultra-trace analysis due to its high ionization efficiency and element selectivity. Exemplary laser spectroscopy on curium and ultra-trace analysis on a multi-element mixture are shown.



Broad band laser spectroscopy on curium

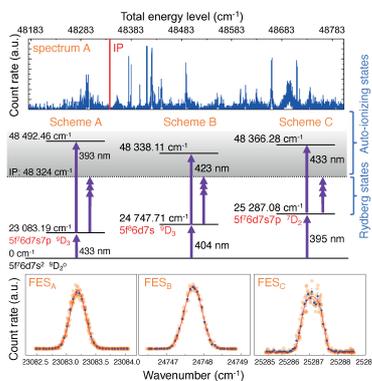
Ionization scheme development

A standard Ti:Sapphire laser is used to populate a first excited step (FES). In addition, an intracavity second harmonic generation (IC-SHG) grating Ti:Sapphire laser is used for the spectroscopic investigation of the ionization region above the ionization potential (IP).

Properties of suitable excitation schemes:

- Ionization efficiency: isotopes in small amounts
- Element-selectivity: multi-element determination

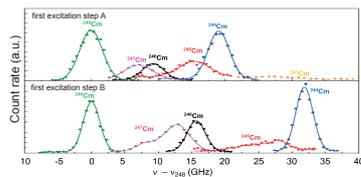
Top: Measured ionization spectrum of scheme A in the range of 48183 cm⁻¹-48783 cm⁻¹.
Bottom: Line profile of the three FES.



Characterization of the isotope shift for homogeneous isotope ionization

The spectral lines of an element differ from isotope to isotope in their relative position and is caused by the nucleus specific properties [5].

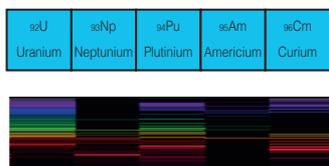
- Small: homogeneous excitation of several isotopes
- Large: isotope selection already by laser excitation



Measured line profile of the FES_A (top) and FES_B (bottom) for ²⁴³⁻²⁴⁸Cm. The line width is reduced by a YAG ethalon.

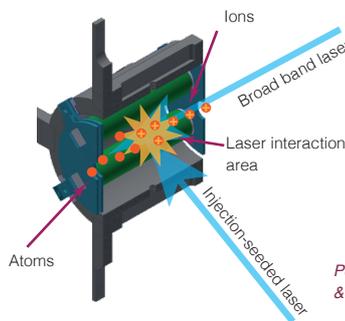
Resonant laser spectroscopy in actinide mixtures

Two-step ionization schemes are developed for U, Pu and Am [9]. Wide spectral ranges are investigated in each of the three elements. Only energy levels, which are not matched in their spectral position with resonances of other elements, are used as excitation steps.



Spectral lines of the lighter actinides. U with the highest spectral line density as Am with the lowest.

High-resolution spectroscopy on curium



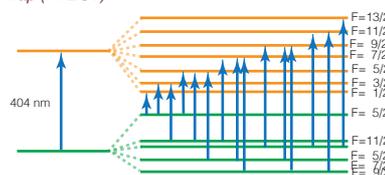
High-resolution laser spectroscopy

The hyperfine spectrum for the 404.073 nm ground state transition in ²⁴⁵Cm is measured (FES_B). All expected 15 lines could be resolved. The fit parameters obtained from the spectrum can be used to calculate core-specific quantities of ²⁴⁵Cm.

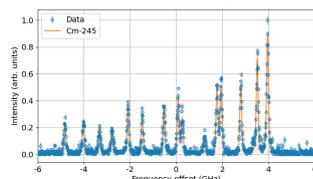
Ion source

The PI-LIST is used as ion source for high-resolution spectroscopy to reduce spectral Doppler broadening. The injection-seeded Ti:Sapphire laser (20-40 MHz linewidth) is used for the spectroscopy transition. The high-power, broadband laser (5-10 GHz linewidth) conducts the ionization step.

Perpendicularly Illuminated Laser Ion Source & Trap (PI-LIST)

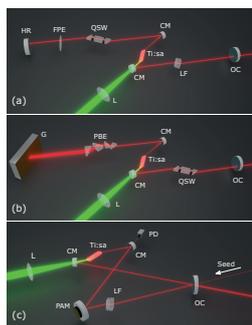


Hyperfine levels of the ground state and of the first excitation step B (FES_B).



Measured hyperfine structure of FES_B. All expected 15 lines could be resolved.

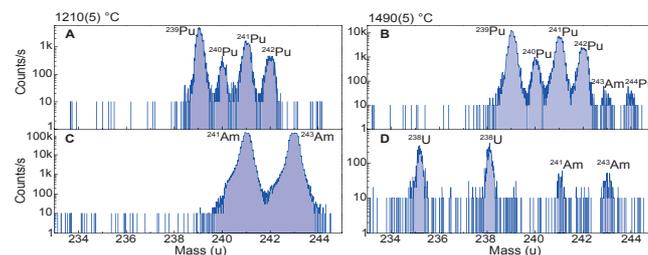
Ti:sapphire lasers for resonant laser spectroscopy



- (a) Broad band laser:
 - High power laser (3 to 50 W)
 - Ultra-trace analysis and multi-element characterization
- (b) Grating-tuned laser (computer controlled):
 - Wide tuning range (700 to 1020 nm)
 - Ionization scheme development, Rydberg analysis
- (c) Injection-seeded laser:
 - Spectral band-width (20 MHz)
 - High-resolution spectroscopy, hyperfine structure and isotope shift

Intra Cavity Doubling Unit for Frequency Doubling (Second Harmonic Generation SHG).

Sketch of the laser types.



Measured mass spectra at different source temperatures (1490(5) °C and 1210(5) °C). The lasers are optimized for the resonances of the isotopes ²⁴¹Pu (A, B), ²⁴¹Am (C), and ²³⁸U (D).

As the actinides have different evaporation curves, the mass spectra are measured at different source temperatures corresponding to the optimal release of an element into an atomic gas.

The suppression factor *S* specifies the selectivity due to resonant laser ionization

$$S = \frac{N_{\text{resonantly-ionized}}}{N_{\text{contamination}}} \cdot X_{\text{sample composition}}$$

N_{contamination}: correspond to the respective cps for signal and selected interference
X_{sample composition}: ratio of the atomic number of isotopes in the initial sample

and is determined for the element ratio ²³⁸U : ²⁴¹Pu with 8 orders of magnitude and ²⁴³Am : ²⁴¹Pu with 3 order of magnitude.

Sample composition:

- 10¹⁶ U-atoms - 1:1 ratio of ²³⁵U:²³⁸U
- 10¹³ Am-atoms - 1:1 ratio of ²⁴¹Am:²⁴³Am
- 10¹² Pu-atoms - 1 : 0.13 : 0.56 : 0.15
²³⁹Pu : ²⁴⁰Pu : ²⁴¹Pu : ²⁴²Pu

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