Laser Ablation of Solids into an Electron Cyclotron Resonance Ion Sources for Accelerator Mass Spectroscopy

MANTRA

- Measurements of Actinides Neutrons Transmission Rates with Accelerator mass spectroscopy.
- Joint project INL and ANL.
- Determine energy-averaged actinide neutron capture cross-sections.
  a. Preparation and Irradiation of pure actinide samples. $^{232}$Th, $^{235}$U, $^{236}$U, $^{238}$U, $^{237}$Np, $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{242}$Pu, $^{244}$Pu, $^{241}$Am, $^{243}$Am, $^{244}$Cm and $^{248}$Cm.
  b. Use accelerator mass spectroscopy to measure the nuclide densities of actinides produced in irradiation through sequential n-capture processes.
  c. Infer capture cross-sections from these ratios.
MANTRA

AMS Challenges:

- Small sample size (few mg total, actinide component <1mg)
- Large number of samples desired to reduce errors
- Minimize cross-talk between samples
- Stable, repeatable transmission between source and ion detector
- Limited “Z” element resolution in detectors

ATLAS Actinide AMS Configuration
We will use laser ablation at relatively low power levels to efficiently introduce solid materials into plasma. Benefits of laser ablation expected are:

- Efficient use of solids for AMS and enriched isotopes.
- Less sensitive to material chemical composition.
- Cleaner source operation (yet to be proven).
- Decouples source operation from material insertion.
Laser ablation

- Laser Ablation - Removal of material by laser action. Distinguished from evaporation in equilibrium conditions
- To remove atom from solid $\varepsilon_{\text{kin}} = \varepsilon_{\text{tot}} - \varepsilon_b > 0$
  - Material parameters: Typical time for thermal equilibrium.
- Two regimes of Laser Ablation
  - Thermal ablation - Heat conduction and hydrodynamic. Large heat affected zones and throw out of a molten material.
  - Non equilibrium - Electrostatic ablation

Source: The experimental points are from Stuart et al. (1996). Theory (dotted line) is from Gamaly et al. (2002).
Laser ablation

- The ablation induces plasma expansion. Plume
- Plasma expansion speed of the order $1 \times 10^6$ cm/sec.
- Laser plumes contain ions, atoms, macroscopic particles, and liquid droplets
  - Spatial intensity across the focal spot of the laser
  - Condensation of vapor during the plume expansion

- The number of ejected atoms for picosecond laser $10^{13}$ atoms/pulse. The ion flux is about 1%.
Off line experimental set up

- λ = 1064 nm
- Rep Rate = Up to 400 Hz
- Pulse width = 15 picosecond
- Output energy = 0.01-4.5mj per pulse
- Gaussian spatial beam profile
- Maximum Power of 3 x10^{10}W per pulse.

Monitoring the laser energy (Labview)

Imaging system

Focal point

Off line test Vacuum chamber

Photodiode

Beam expander X2

f=300mm f=200mm

f=4000mm f=4000mm

Laser
The ions energy was extracted from time of flight measurements using a Faraday cup. The cup was at a distance of 6.875 inch from a solid Ta target.
# Ablating Rates for different materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Consumption rate</th>
<th>Hole depth</th>
<th>Image</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe solid (1 location shooting for 39 min)</td>
<td>1.3mg/39min, 0.033mg/min</td>
<td>1.2mm (for 39 min)</td>
<td><img src="image" alt="Image" /></td>
</tr>
<tr>
<td>Fe solid (3 locations 13 minutes on each location)</td>
<td>1.4mg/39min, 0.035mg/min, 3.7*10^17 atoms/min</td>
<td>1.19mm (for 13 min)</td>
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</tr>
<tr>
<td>Fe oxide powder-MANTRA target (3 locations 13 minutes on each location)</td>
<td>1.3mg/39min, 0.033mg/min, 1.24*10^17 atoms/min</td>
<td>1.07mm (for 13 min)</td>
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<tr>
<td>Al oxide powder-MANTRA target (3 locations 10 minutes each)</td>
<td>0.1mg/30min, 0.003mg/min, 1.77*10^16 atoms/min</td>
<td>0.8mm (for 10 min)</td>
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<tr>
<td>Tb oxide powder-MANTRA target (2 locations 10 minutes each)</td>
<td>0.1mg/20min, 0.005mg/min, 8.2*10^15 atoms/min</td>
<td>0.57mm (for 10 min)</td>
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<tr>
<td>U metal (3 locations 10 minutes each)</td>
<td>4mg/30min, 0.13mg/min, 3.289*10^17 atoms/min</td>
<td></td>
<td><img src="image" alt="Image" /></td>
</tr>
<tr>
<td>U oxide (3 locations 10 minutes each)</td>
<td>0.5mg/30min, 0.016mg/min, 3.56*10^16 atoms/min</td>
<td></td>
<td><img src="image" alt="Image" /></td>
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</tbody>
</table>

**Ablating Rates**

**Samples from INL**

**Laser Energy:** 1.5-1.6mJ

**400Hz repletion rate**

**Focal spot diameter:** 0.5mm

**Peak flounce:** 0.7 J/cm^2

**Pulse duration:** 15ps
The Beam Manipulator

- Controlled motors that placed on the aligning knobs of the last mirror.
- Can wobble the laser beam on the target sample.
Installation at the source (ECRII)

ECR plasma  Laser at ground potential  Optical elements

Bending magnet  Focusing lens to target  High voltage platform
Multisample changer for the source

holds 20 samples can change between samples in <1 minute absolute encoder so position information is preserved size keeps operating mechanism out of high B field laser sensor to ensure sample is retracted before rotating operation can be controlled by accelerator crew or experimental program (batch program)
Back light

Retracted target samples
Iron taper
Target sample in place
Laser beam
ECR plasma

3.1 mm
Ti sample at the ECR source

- Consumption rate 0.3 mg/hour
- Laser parameters:
  - Repetition rate 25Hz
  - Energy 0.5-1.5 mJ
  - Peak intensity of $5 \times 10^{10}$ W/cm$^2$
Beam from titanium sample ablated into ECRIS

- Charge State Distribution

Laser ON

Laser OFF

Charge State Distribution

- 48/14+
- 48/13+
- 48/11+
- 48/10+
Long-term beam output from ablated Ti sample

- Laser repetition rate 25 Hz
- Laser Energy ~ 1.5mJ
- Charge state 48/13$^+$
  - stable for the first 10 min
  - drops 80% in the next 20 min
  - stay stable for 65 min
Long-term beam output from ablated Ti sample

- Laser repetition rate 25 Hz
- Laser Energy ~ 0.5mJ
- Charge state 48/13$^+$

  - drops 36% for the first 20 min
  - drops 15% for the next 20 min
  - stay stable for 20 min
Ti sample at the ECR source

- New target
- Changed the focal spot to be bigger
- Consumption rate 0.35 mg/hour
Long-term beam output from ablated Ti sample

- Laser repetition rate 25 Hz
- Laser Energy ~ 1.5mJ
- Charge state 48/13⁺
  - drops 37% in the first 2 min
  - stay stable for the next 20 min
  - drops 43% for 15 min
Conclusion

- Demonstrated beam production at moderate intensities.
- Most of the beam loss is due to the drilling.
- Only part of the beam loss is due to laser instabilities.

What next

- Improving the stability of the beam
  - Adjusting the focal spot of the laser
    - bigger focal spot.
    - change the spatial profile of the laser beam to a hat top.
  - Moving the laser beam on the sample in a constant rate using the beam manipulator.
- Next test July 12.