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

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MANTRA

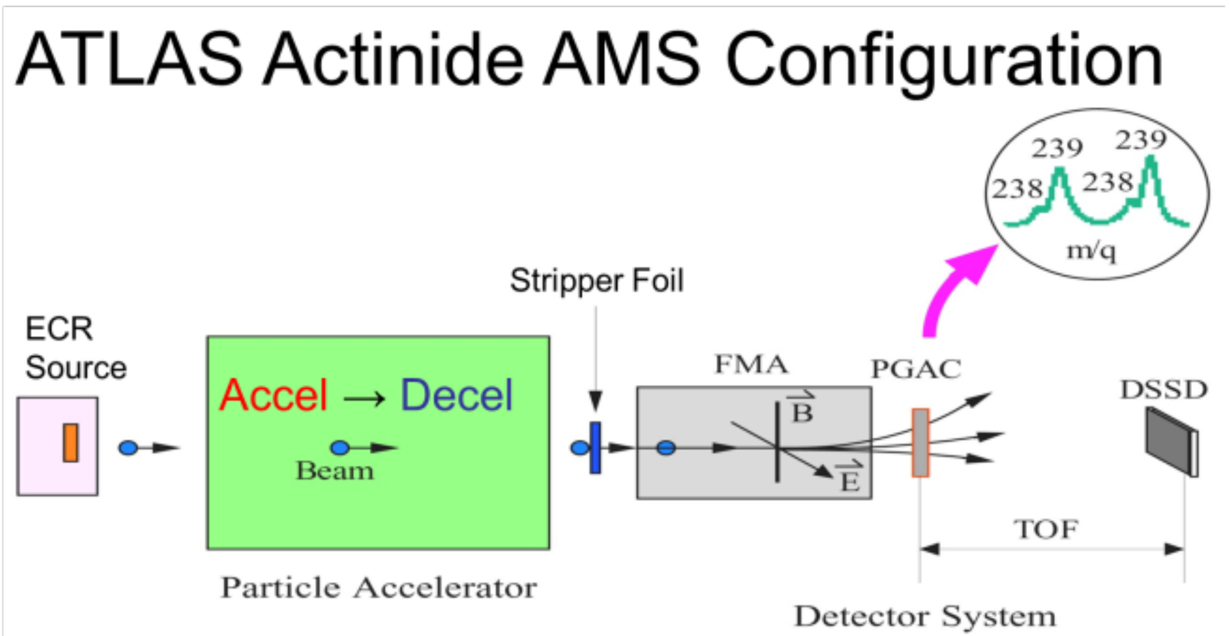
- Measurements of Actinide Neutron Transmission Rates with Accelerator mass spectroscopy.

- Joint project between Idaho National Laboratory and Argonne.
- Determine energy-averaged actinide neutron capture cross-sections.
- 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.
- Use accelerator mass spectroscopy to measure the nuclide production ratios of actinides produced in irradiation through sequential n-capture processes.
- 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



MANTRA

- We will use laser ablation at relatively low power levels to efficiently introduce solid materials into plasma. Expected benefits of laser ablation are:

- Efficient use of solids for AMS and enriched isotopes.
- Less sensitive to material chemical composition.
- Cleaner source operation.
- Decouples source operation from material insertion.

Laser ablation as a way to introduce solid material to ECR source.

- Laser Ablation– Removal of material by laser action. Distinguished from evaporation in equilibrium conditions

- To remove atom from solid $\varepsilon_{kin} = \varepsilon_{tot} - \varepsilon_b > 0$
  - Material parameters: Typical time for thermal equilibrium.
  - Laser parameters: Wavelength, Pulse duration, Intensity.
- Typical ablation fluence is of the order of 1 J/cm^2

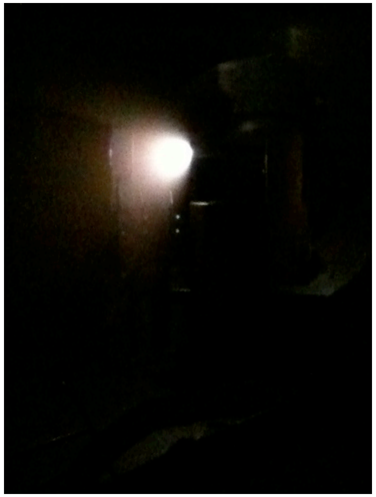
  - 100 fs → 10^{13} W/cm^2 Ionisation and formation of plasma non-equilibrium interaction. The extreme ablation mode, electrostatic ablation.
  - 10 ns → 10^8–10^9 W/cm^2 Heating melting and evaporation leads to large heat affected zones and throw out of a molten material. Equilibrium interaction. Thermal ablation.

  - Non equilibrium and sami-thermal mode. The majority of the atoms leaves the solid before the equilibrium is established.

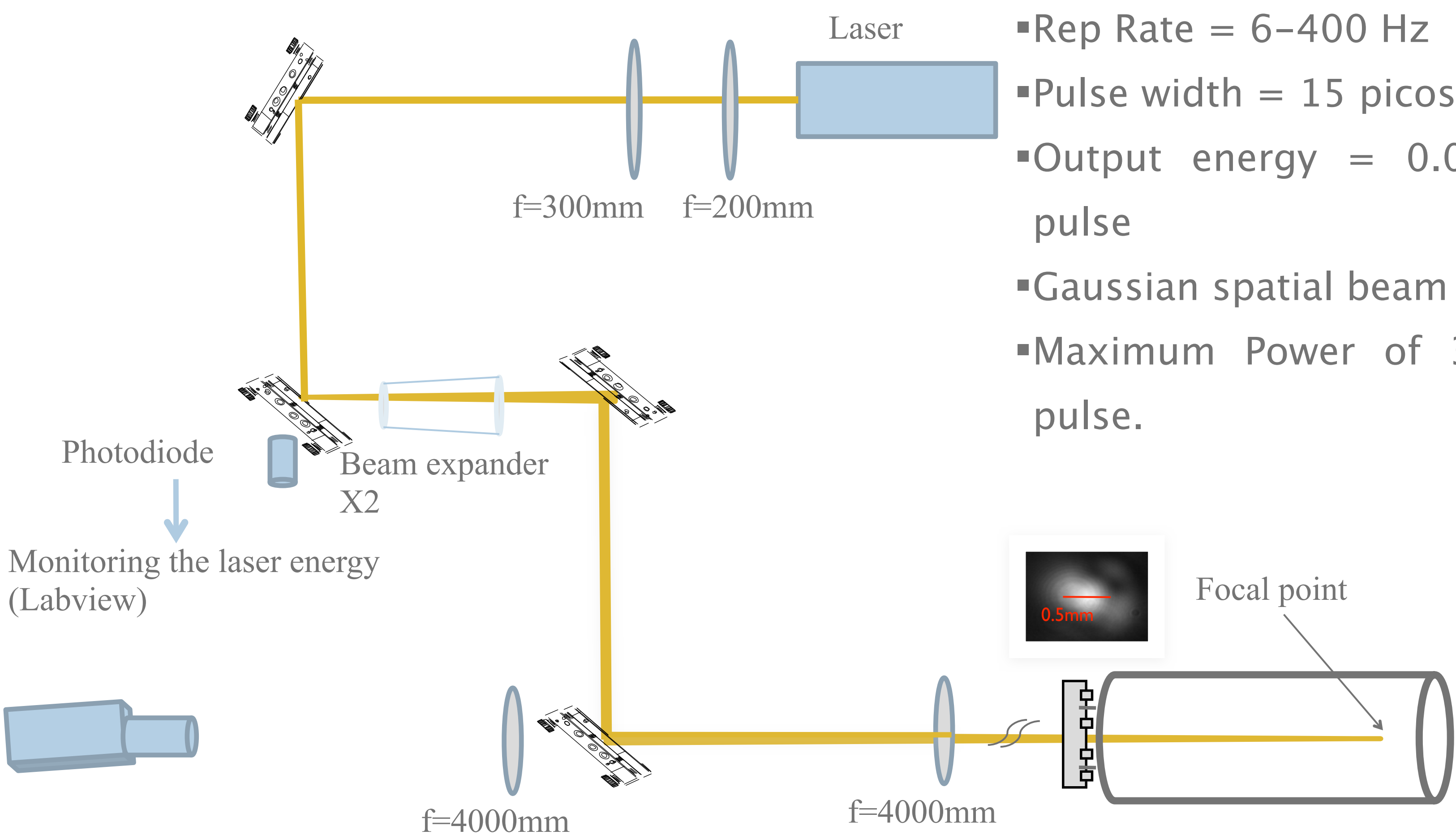
Laser ablation

- The ablation induces plasma expansion plume
- Plasma expansion speed of the order $1X10^6$ 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 = 6-400 Hz
- Pulse width = 15 picosecond
- Output energy = 0.01-5mj / pulse
- Gaussian spatial beam profile.
- Maximum Power of 3×10^8 W/ pulse.

Ablating Rates for different materials

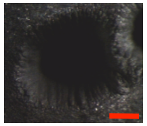
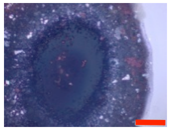
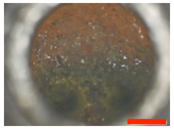
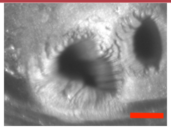
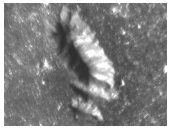
Laser Energy : 1.5-1.6 mJ

400Hz repetition rate

Focal spot diameter: 0.5mm

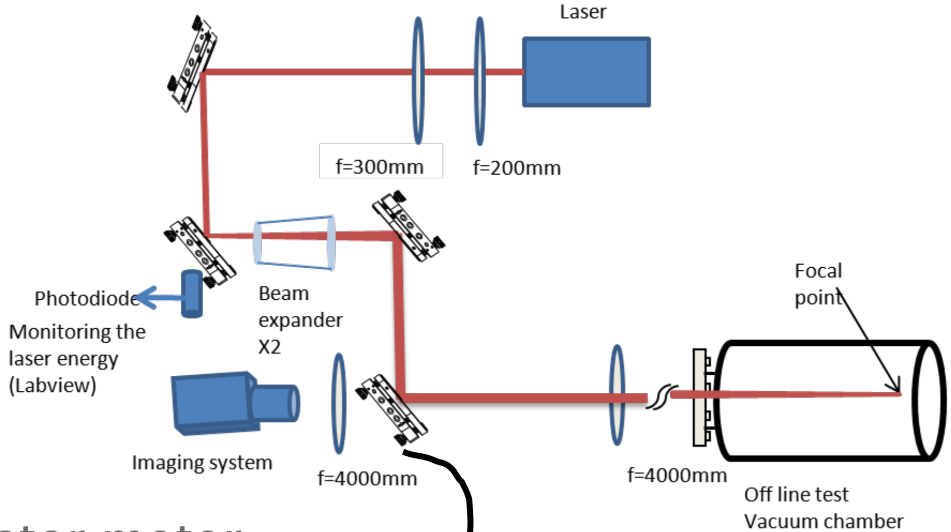
Peak fluence : 0.7 J/cm^2

Pulse duration : 15 ps

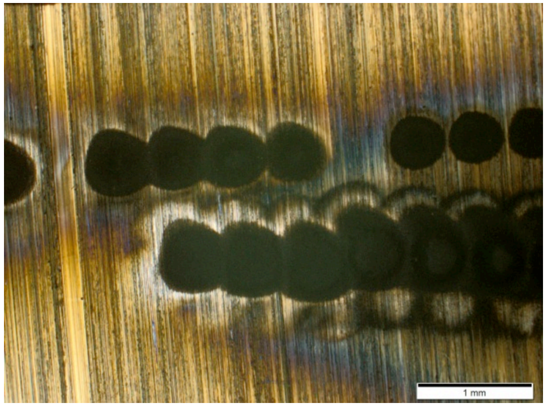
	Consumption rate	Hole depth	Image
Fe solid (1 location shooting for 39 min)	1.3mg/39min 0.033mg/min	1.2mm (for 39 min)	 0.2mm
Fe solid (3 locations 13 minutes on each location)	1.4mg/39min 0.035mg/min 3.7*10^17 atoms/min	1.19mm (for 13 min) 0.09mm/min	 0.5mm
Fe oxide powder– (3 locations 13 minutes on each location)	1.3mg/39min 0.033mg/min 1.24*10^17 atoms/min	1.07mm (for 13 min) 0.08mm/min	 0.2mm
Al oxide powder– (3 locations 10 minutes each)	0.1mg/30min 0.003mg/min 1.77*10^16 atoms/min	0.8mm (for 10 min) 0.08mm/min	 0.5mm
Tb oxide powder (2 locations 10 minutes each)	0.1mg/20min 0.005mg/min 8.2*10^15 atoms/min	0.57mm (for 10 min) 0.057mm/min	 0.5mm
U metal (3 locations 10 minutes each)	4mg/30min 0.13mg/min 3.289*10^{17} atoms/min		 0.5mm
U oxide (3 locations 10 minutes each)	0.5mg/30min 0.016mg/min 3.56*10^{16} atoms/min		

The Beam Manipulator

- Controlled motors mounted on the aligning knobs of the last mirror
- Can wobble the laser beam on the target sample
- Minimal step size on the target of 110μm



Manipulator motor

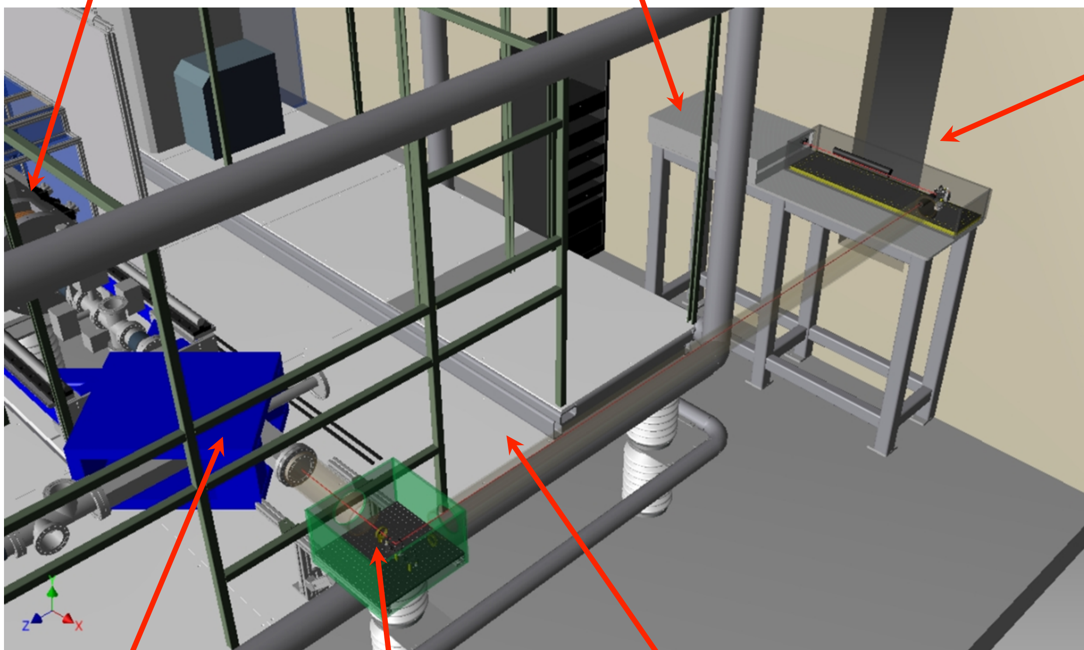


Installation at the source (ECR2)

ECR plasma

Laser at ground potential

Optical elements



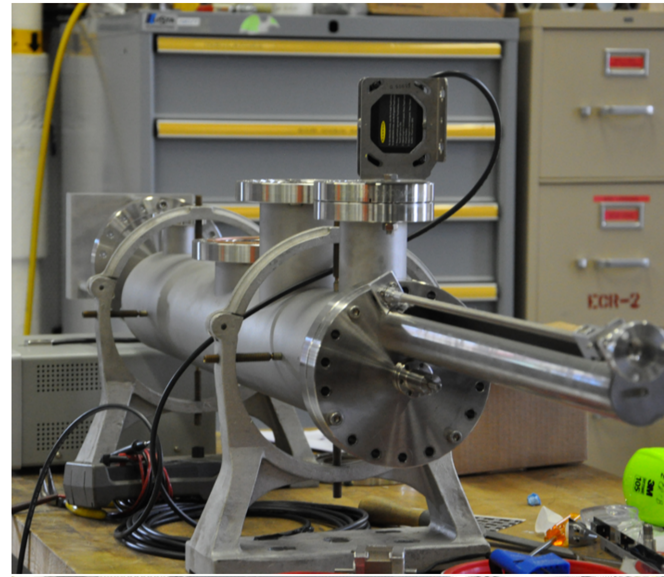
Bending magnet

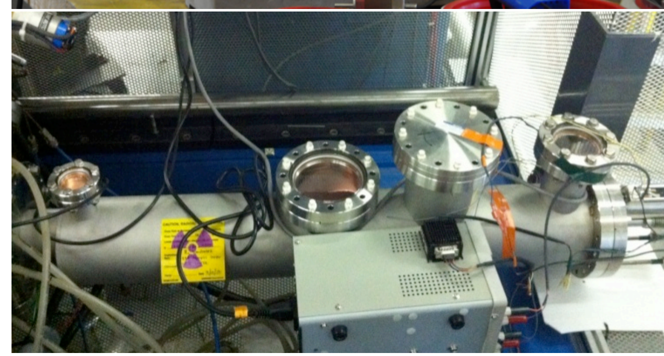
High voltage platform

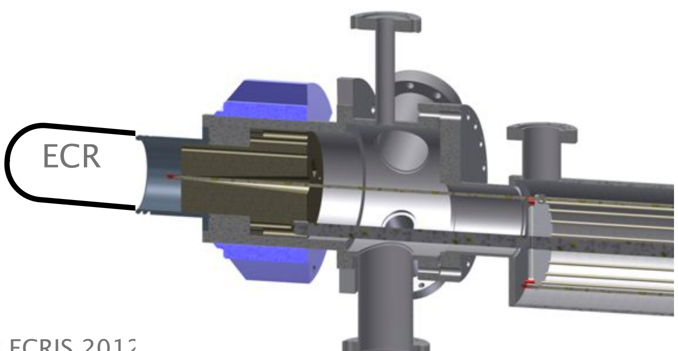
Focusing lens to target

Multisample changer for the source

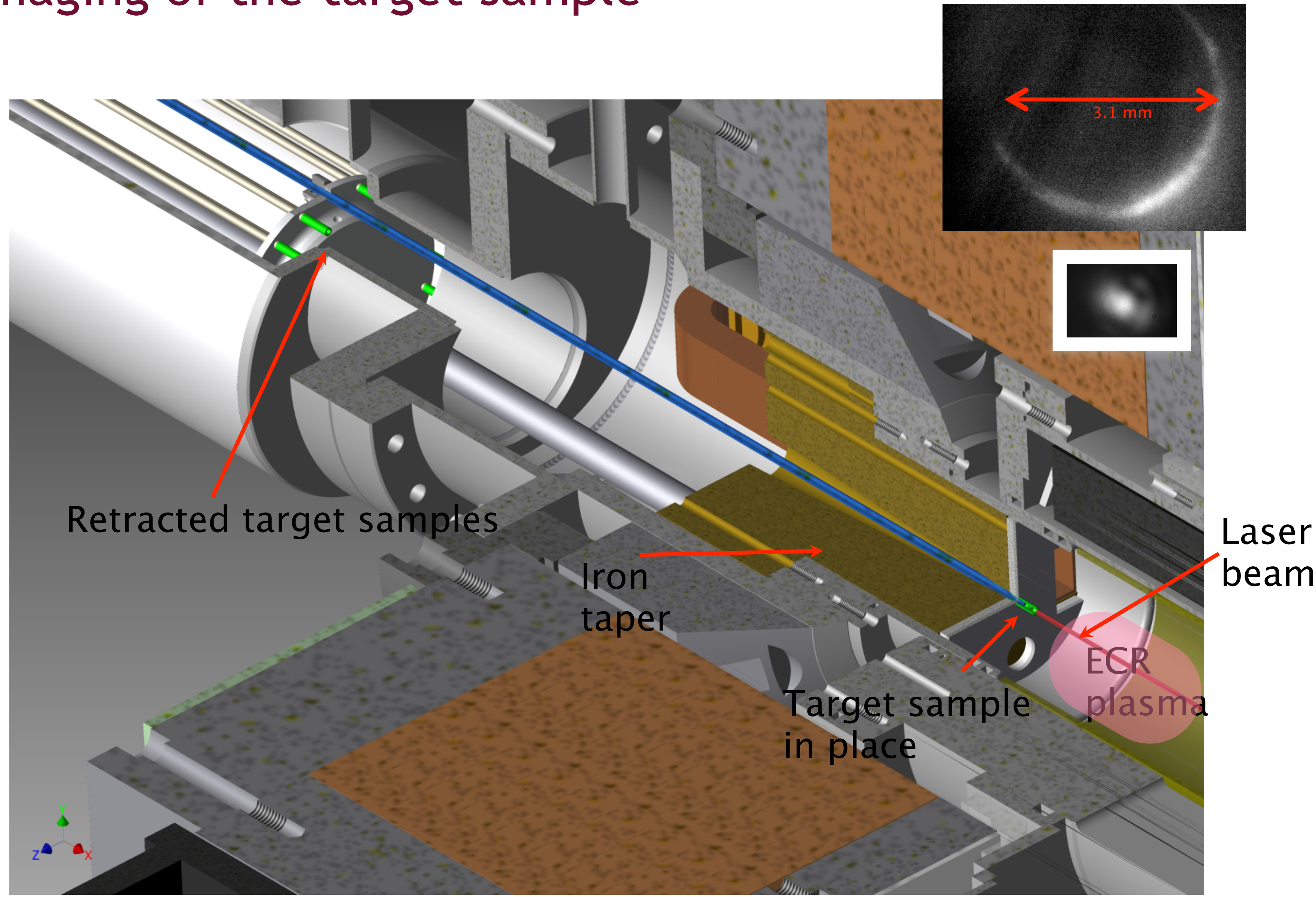
- Holds 20 samples
- Can change between samples in <1 minute
- Absolute encoder to maintain position information
- Size keeps operating mechanism out of high B field
- Laser sensor ensures sample is retracted before rotating
- Operation can be controlled by accelerator operator or experimental program (batch program)





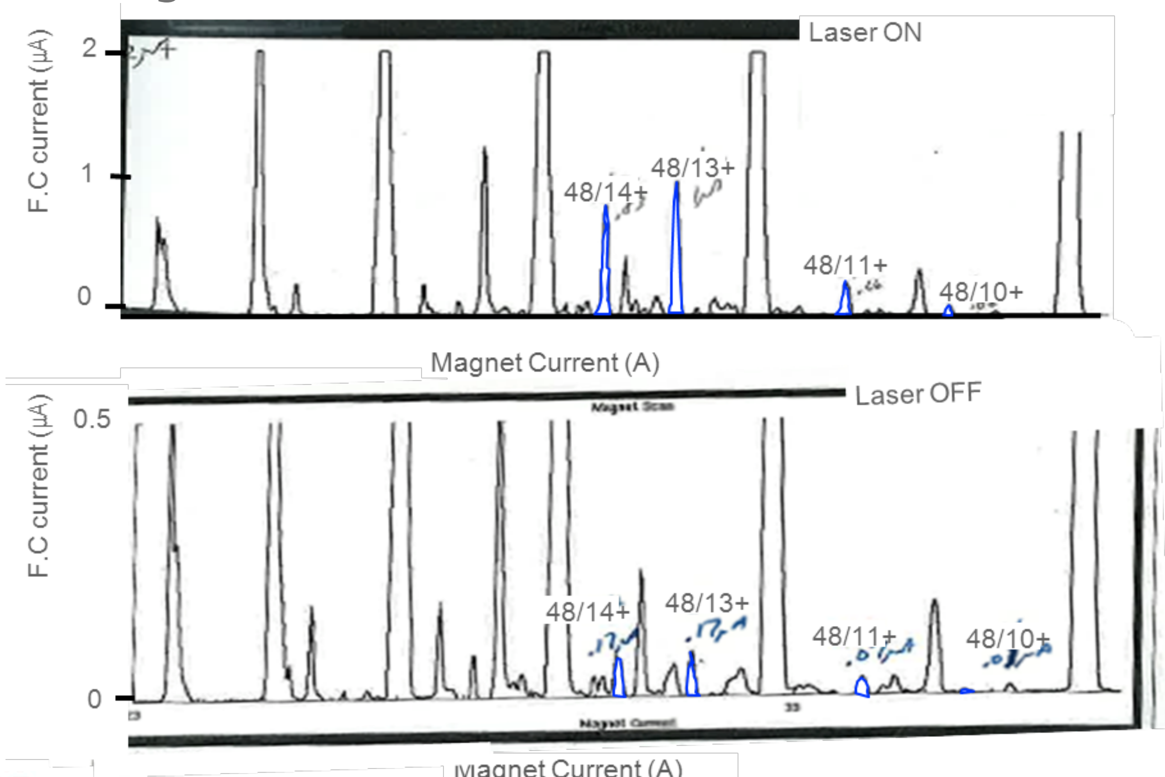


Imaging of the target sample



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 $5X10^{10}$ W/cm^2
- Charge State Distribution



Depth 200μm

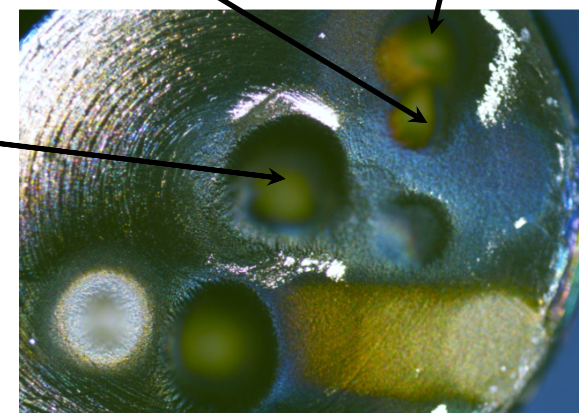
Diameter 0.18mm

Depth 350μm

Diameter 0.24mm

Depth 930μm

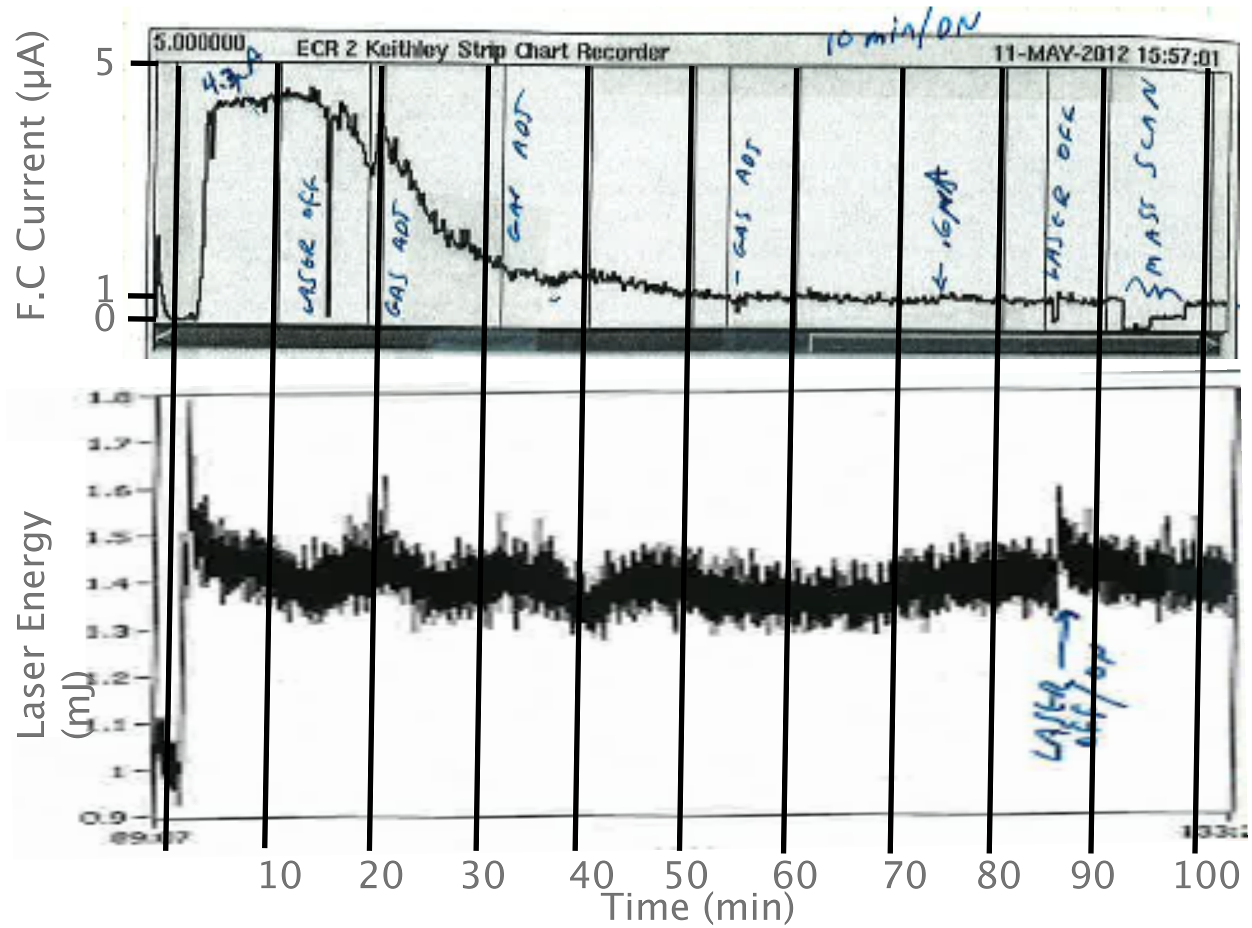
Diameter 0.42mm



Long-term beam output from ablated Ti sample

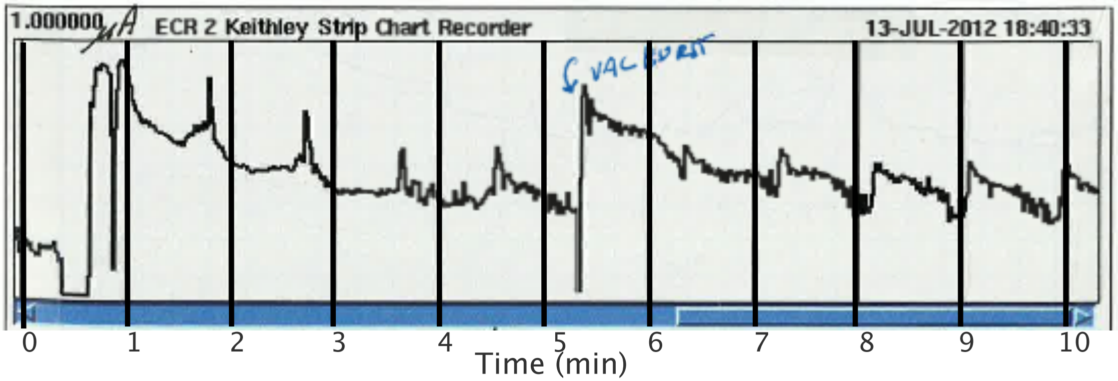
- Laser repetition rate: 25 Hz
- Laser Energy: ~ 1.5mJ
- Charge state: $48/13^{+}$

- stable for the first 10 min
- drops 80% in the next 20 min
- stay stable for 65min

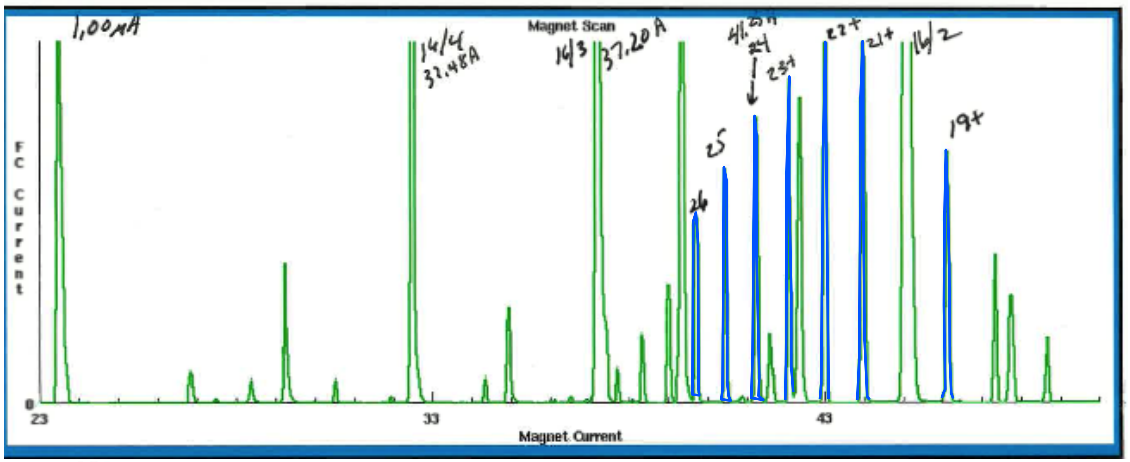


Terbium oxide sample at the ECR source

- Moving the laser beam on the sample in a constant rate using the beam manipulator
- Consumption rate: 0.32mg/hour
- Beam output for 159/24+. Laser parameter: 100Hz Rep. Rate 2.3mJ Energy/ pulse.

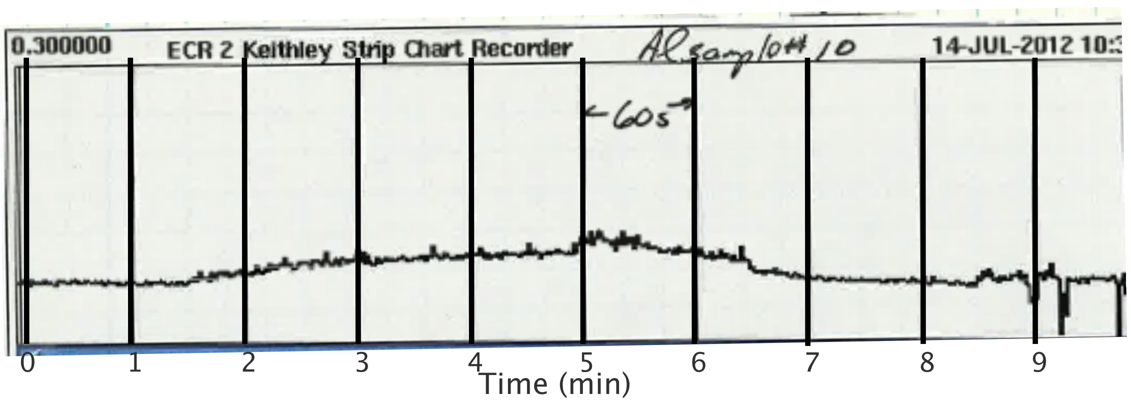


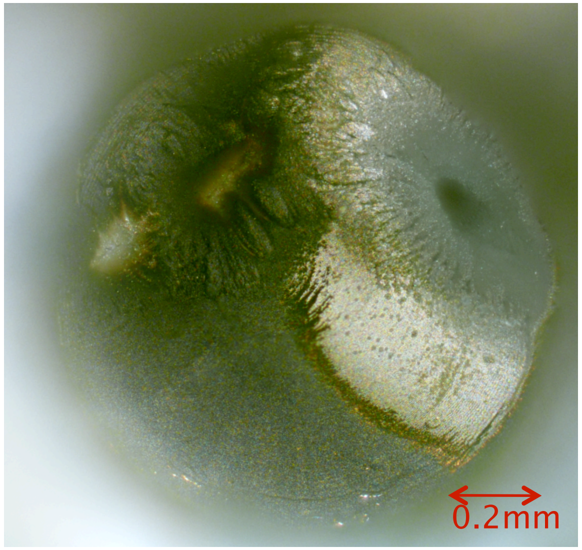
Charge state distribution



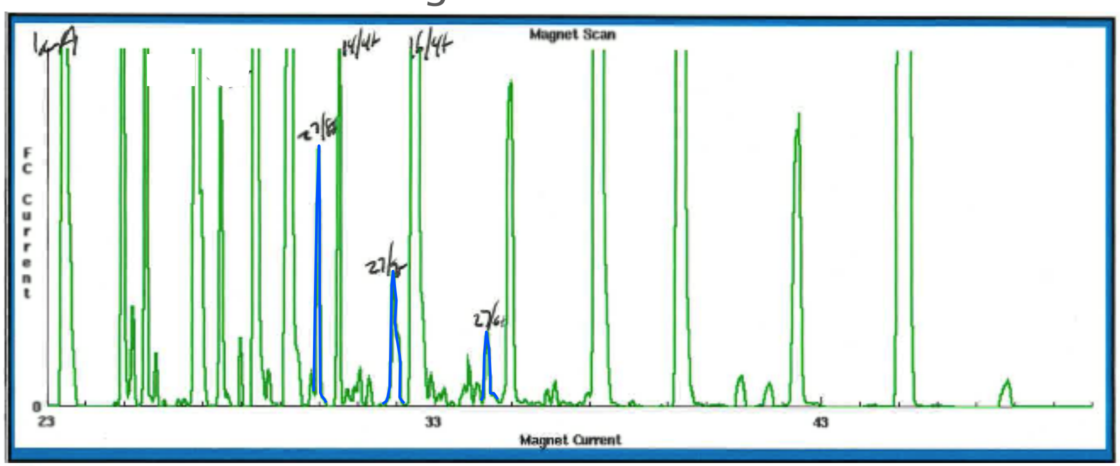
Aluminum sample at the ECR source

- Consumption rate: 0.45mg/hour
- Beam output for 27/8+. Laser parameter: 100Hz 2.2mJ.





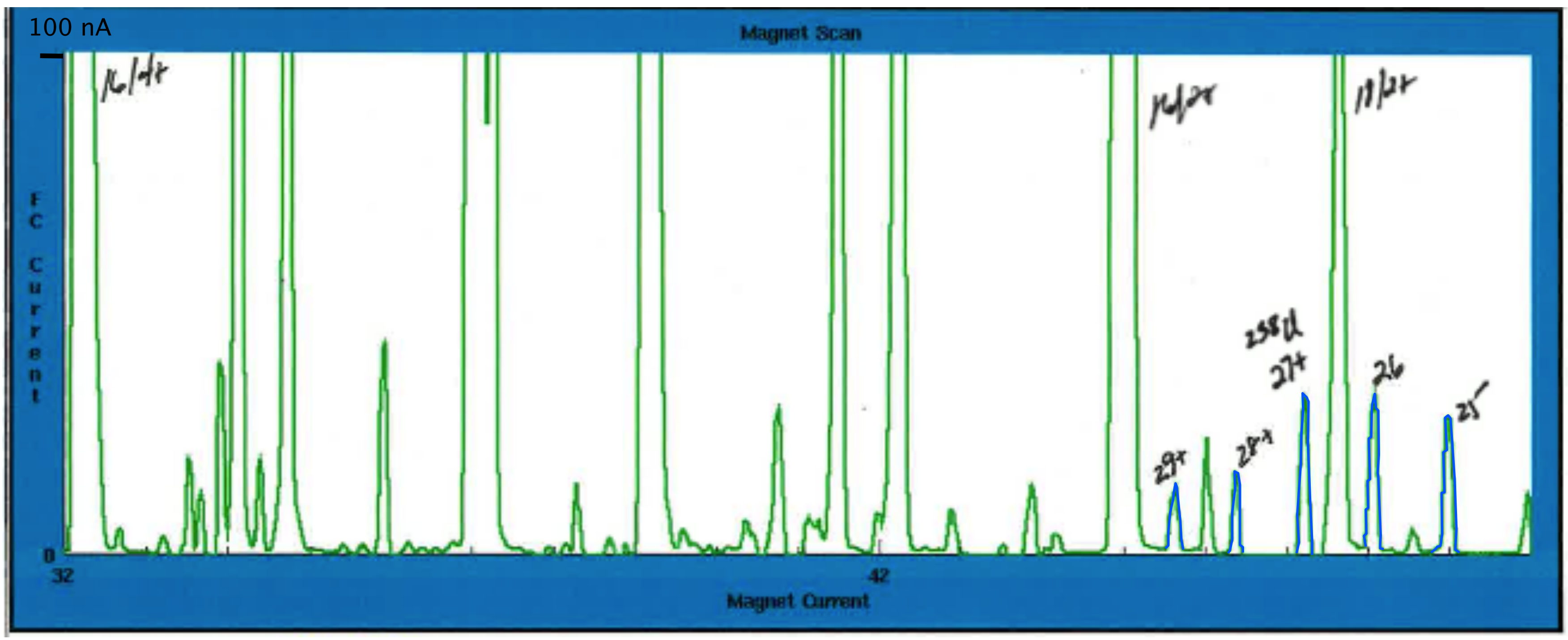
Charge state distribution



Uranium oxide sample at the ECR source

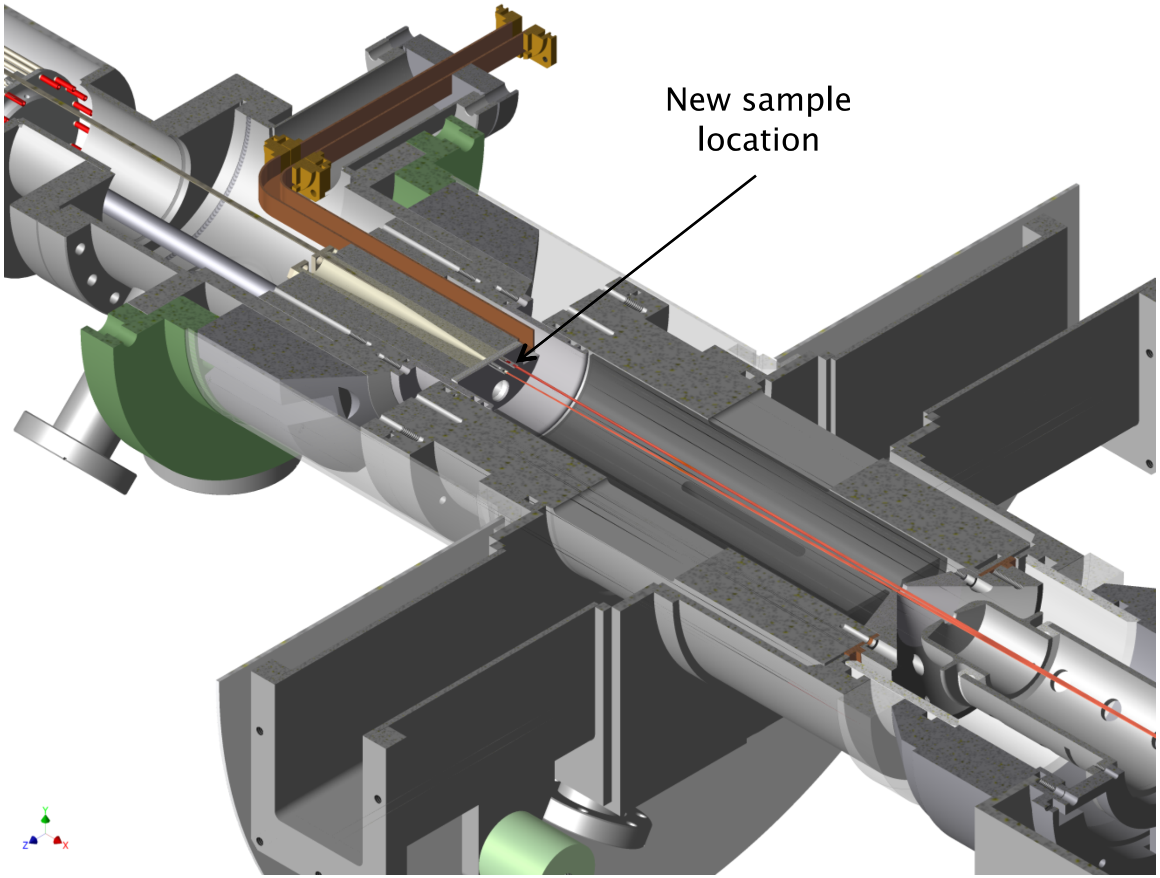
- Consumption rate: 0.7mg/hour
- Laser parameter: 200-400Hz, 5mJ.

Charge state distribution



A new - off axis location of the sample

- Poor performance of the ECR ion source using the on axis geometry with a hole in the middle of the bias disc (16/6+ at 35μA)
- The performance of the ECR ion source are recovered once the hole in the middle of the bias disc is filled with Al (16/6+ at 160μA).
- New design with an off axis location of the target sample.



Conclusion

- Demonstrated beam production at moderate intensities.
- Most of the beam Instability is due to drilling and low source performance.

What next

- Improving the stability of the beam
- Moving the sample to be off-axis.
  - Adjusting the focal spot of the laser
    - bigger focal spot.
    - change the spatial profile of the laser beam to a flat top profile.