



Applications, Developments, and Recent Work with O⁻ Beams

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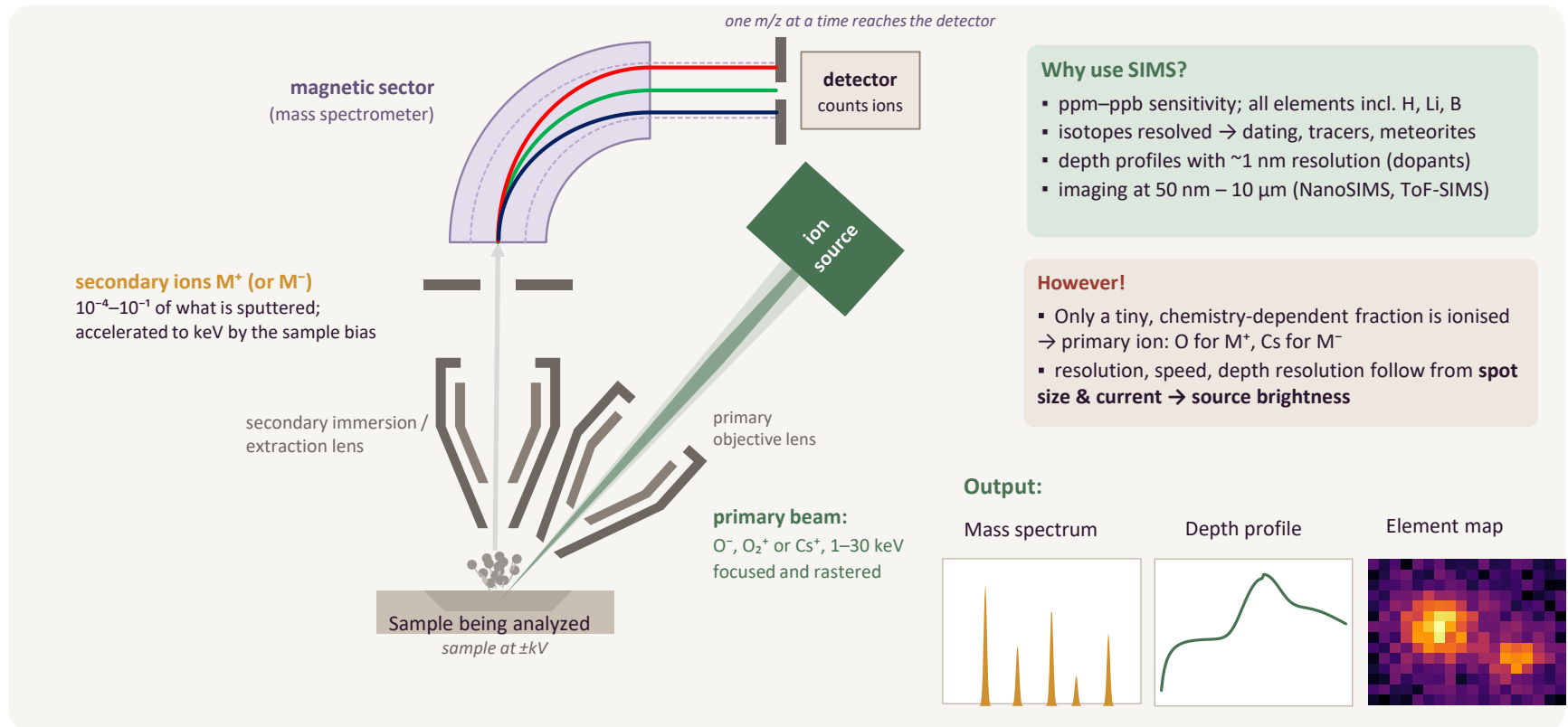
Overview

- What is Secondary Ion Mass Spectrometry (SIMS)
- Why Focused O^- beams are used.
- Ion Source requirements for Focused Ion Beams
- Evolution from Duoplasmatrons to ICP ion sources.
- Possible Future Developments



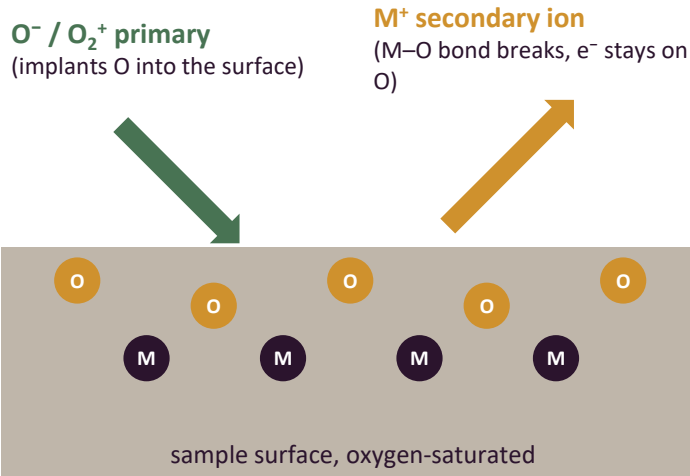
Secondary Ion Mass Spectrometry (SIMS)

Hit a surface with a focused keV ion beam, collect the few sputtered atoms that leave as ions, and mass-analyse them — the most sensitive surface analysis technique there is.



Instrument families: Magnetic Sector SIMS (Cameca NanoSIMS/1300/IMS Series — magnetic sector, high mass resolution and transmission – nuclear, biology, geology, etc..) ToF-SIMS (pulsed Bi⁺, Ar cluster — molecular surface info) Quadrupole SIMS (mostly semiconductor applications)

Why Oxygen Primary Beams for SIMS?



“Bond-breaking” picture of the oxygen matrix effect

Secondary ion yield of many electropositive species enhanced by implanted oxygen

- Sputtering removes atoms efficiently; the problem is that almost all leave **neutral**
- Oxygen in the surface raises the work function and forms M–O bonds; sputtered matrix atoms leave as M^+ with 10–1000× higher probability
- The beam itself implants oxygen, so the surface reaches a steady oxide state.
- Complement: Cs^+ lowers the work function → boosts **negative** secondaries (H, C, O, S, Si, halogens, As, P)

Oxygen beams ⇒ positive secondaries ⇒ the electropositive elements

Dopants and metals in semiconductors: B, Al, W, Cu, Fe, Ti

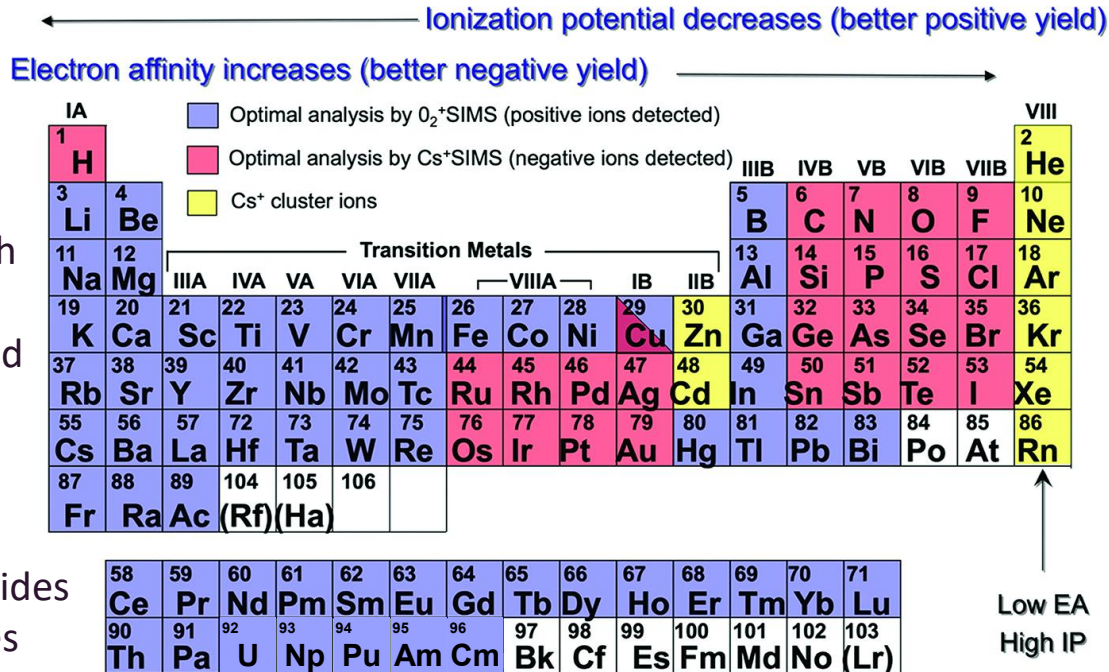
Geoscience, cosmochemistry, nuclear forensics: Mg, Al, Si, Ca, Ti, Cr, Fe, rare earths, U, Th, Pb

Biology: Ca, Mg, Na, K and Lanthanides

Secondary ion yield enhancement of electropositive ions with oxygenated surface

Alkali metals and alkali earth metals give highest ion yield

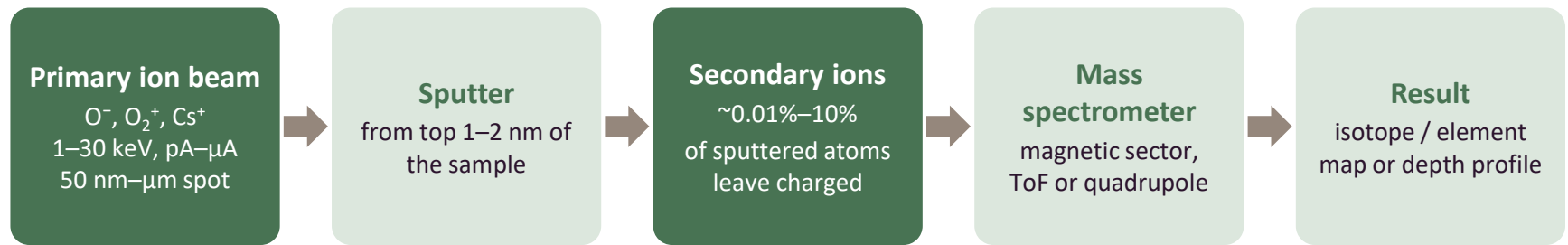
Lanthanides
Actinides



U–Cm shaded here (unshaded in the original only because no RSF data existed): the actinides have low ionisation potentials (U 6.2 eV, Pu 6.0 eV) and form very polar oxides, so O^- / O_2^+ is the standard beam for U and Pu isotopics — measured as U^+ , UO^+ and UO_2^+ .

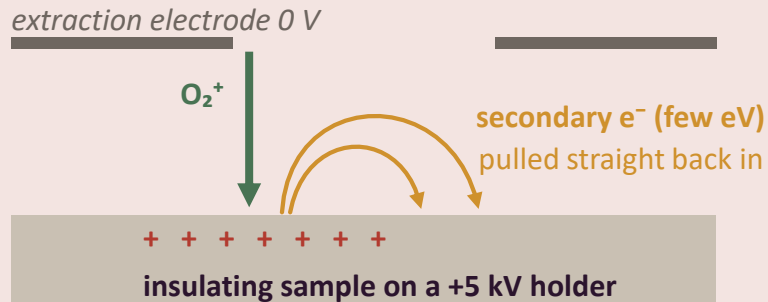
Ref: Secondary Ion Mass Spectrometry: Fundamentals, Advancements and Applications, 2025

SIMS for Trace Element Detection



Why O^- for Insulating Samples?

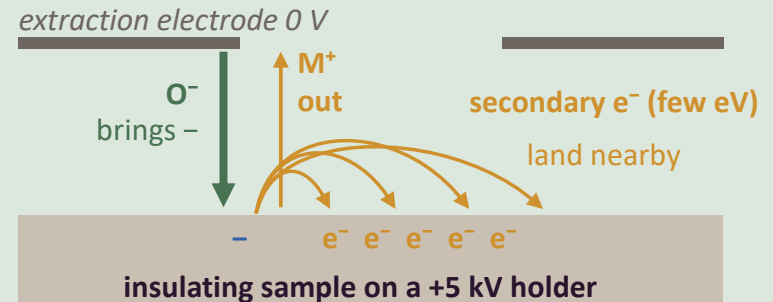
Positive primary (O_2^+)



→ surface potential rises without limit

- primary beam is deflected and defocused → image degrades
- secondary ions gain energy → shifted out of the spectrometer's energy window

Negative primary (O^-)



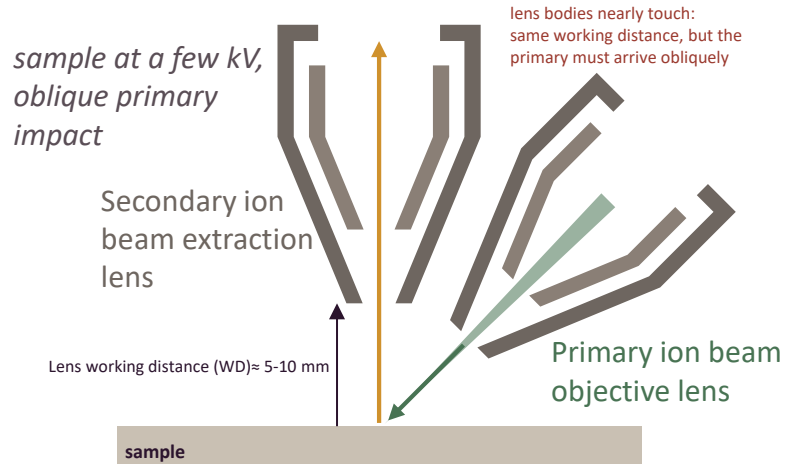
net charge per ion: -1 , balanced by ≈ 1 escaping electron
→ spot settles a few volts negative and stays there

- O^- in and M^+ out drive the surface slightly negative which ejects secondary e^- into the surrounding area. Secondary e^- are only a few eV, so they stay close and re-land, but analysis region becomes stable.

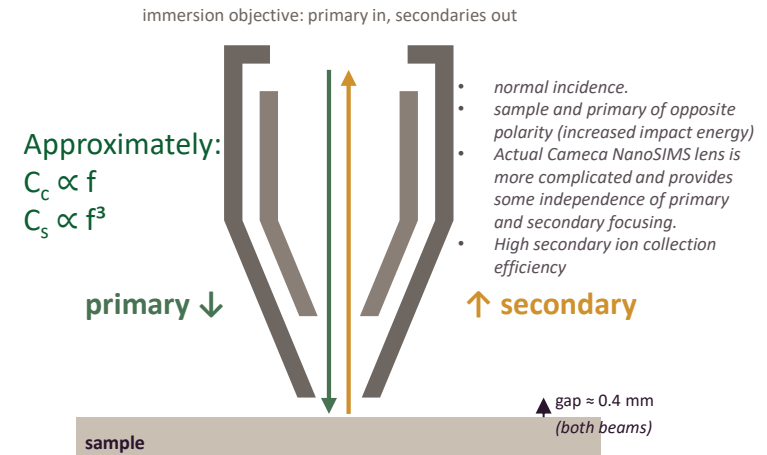
A negative primary brings its own neutralisation.
No low energy electron flood gun needed.
Stable images and spectra.

O⁻ Primary Ions Can Share an Objective lens with Positive Secondary Ions

Conventional SIMS: two separate objective lenses



Coaxial (NanoSIMS): one objective for both beams



Primary probe size

aberrations grow with working distance:

C_s, C_c scale with WD $\rightarrow d_{\min} \uparrow$

WD = 5–10 mm vs 0.4 mm

Extraction field at the sample

$E = V / \text{gap}$: 5 kV / 5 mm $\approx$ 1 V/ μ m

5kV / 0.4 mm $\approx$ 12.5 V/ μ m

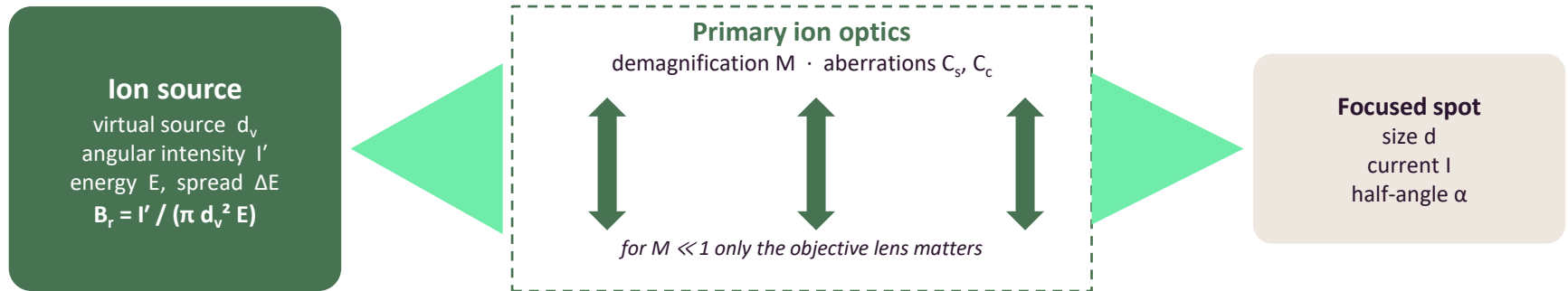
stronger field $\rightarrow$ increased secondary ion collection efficiency

Beam Polarity

The two beams must have opposite sign,

O⁻ required for positive secondaries

How a Primary Ion Column Works



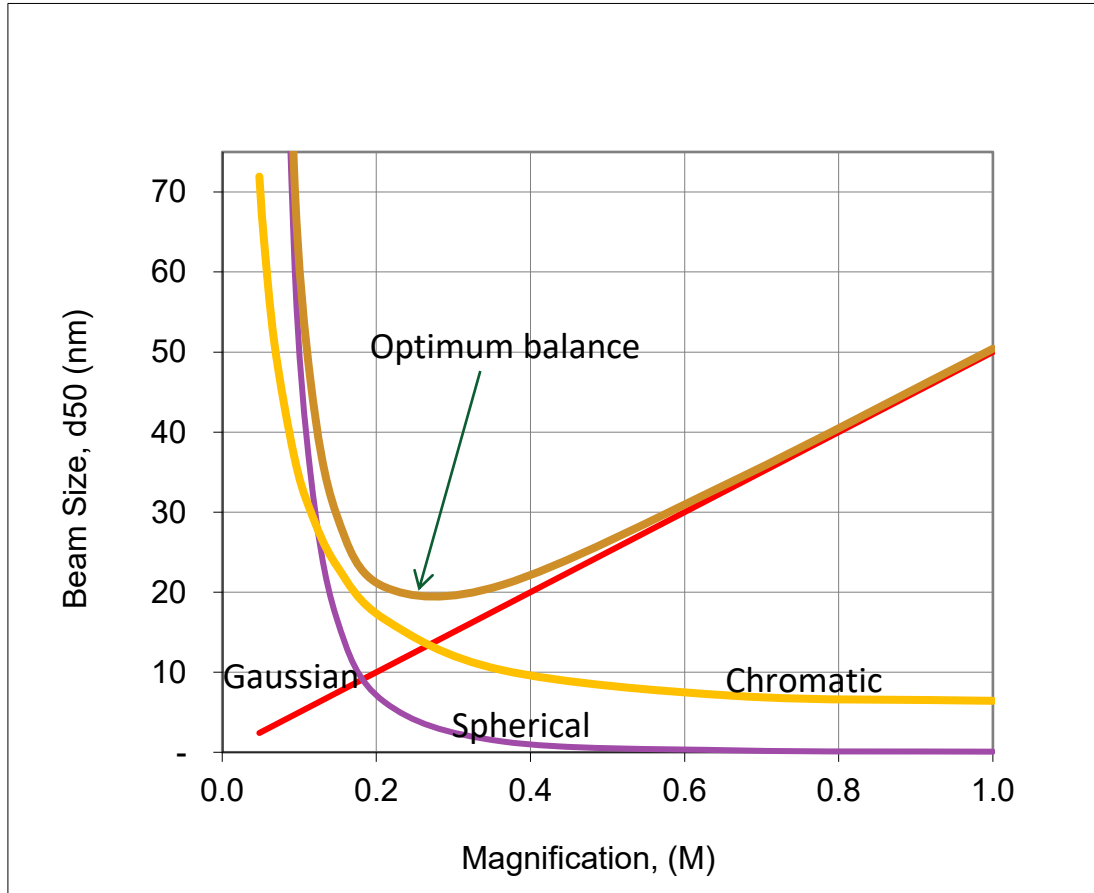
Three blurs, added in quadrature

$\alpha = \sqrt{I / (\pi \cdot I' \cdot M^2)}$	<i>Half-Angle needed for current, I</i>
$d_g = M \cdot d_v$	<i>Gaussian image</i>
$d_c = (\Delta E / E) \cdot C_c \cdot \alpha$	<i>Chromatic blur</i>
$d_s = \frac{1}{2} \cdot C_s \cdot \alpha^3$	<i>Spherical blur</i>
$d = \sqrt{d_g^2 + d_c^2 + d_s^2}$	<i>Total spot</i>

- The **source** fixes B_r and ΔE — nothing downstream can improve them
- The **column** fixes C_s, C_c (design) and M (lens excitation — the one knob you turn)
- Ask for more current $\rightarrow$ larger $\alpha \rightarrow$ aberrations grow; demagnify harder $\rightarrow$ smaller image but larger α again
- So for every current there is an **optimum M**

"Classical (quadrature) estimate; Barth & Kruit, Optik 101 (1996) give a more accurate FW50 addition rule."

Minimum Spot: Balancing the Three Blurs



Sweep M at a fixed current

- Gaussian image $\propto M$, chromatic $\propto 1/M$, spherical $\propto 1/M^3$
- The quadrature sum has a minimum $\rightarrow$ that M is the optimum lens excitation
- Repeat for each current $\rightarrow$ the spot-size-vs-current curve shown later for Hyperion II

What the source buys you

At low I (chromatic-limited):

$$d_{\min} \propto (C_c \Delta E/E)^{1/2} \cdot (I/B_r)^{1/4}$$

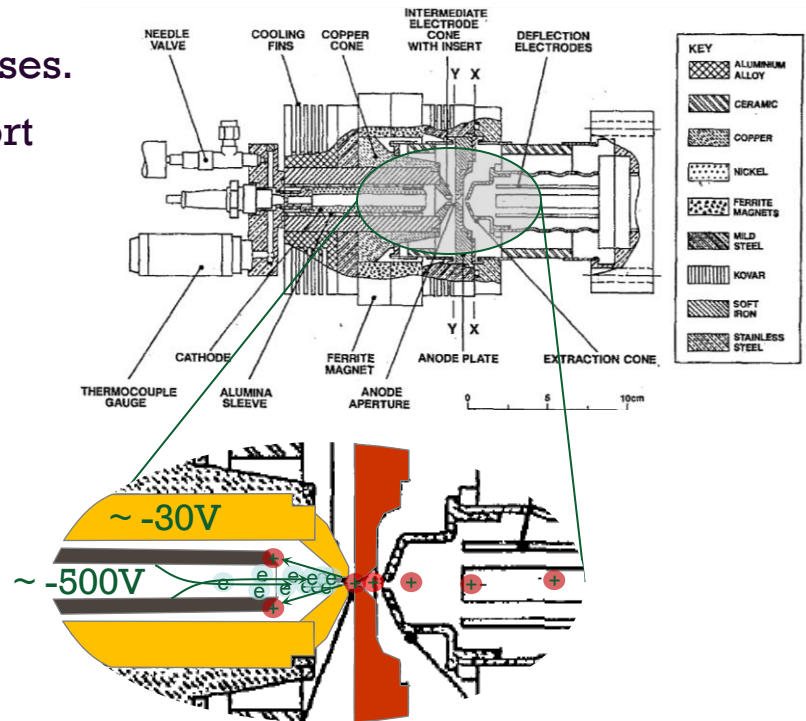
- $d_{\min}$ scales with the inverse 4th root of the brightness, such that 10x gain in brightness results a factor of 1.8 reduction on spot size.
- Reducing the energy spread by a factor of 2 and you get a factor of 1.4 reduction in $d_{\min}$.

~1965-2015 - Only Duoplasmatrons for O_2^+ and O^- Beams

Duoplasmatron

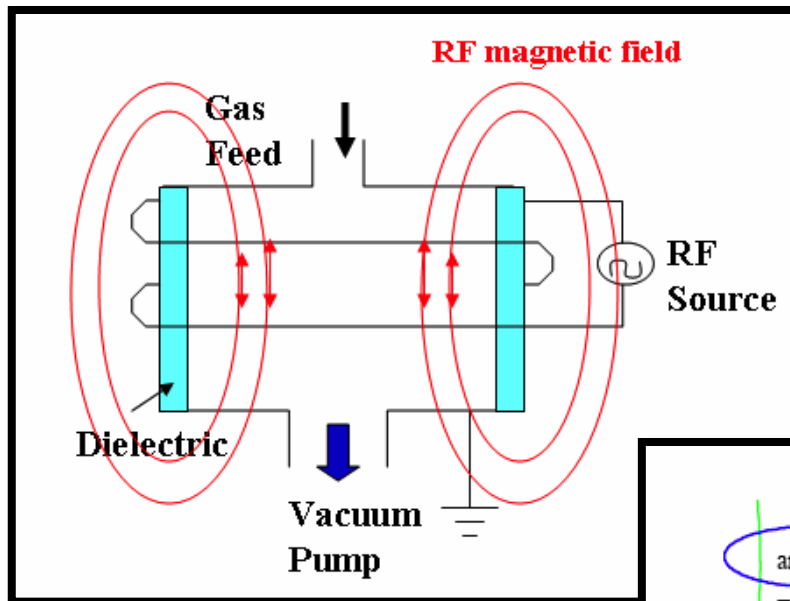
- Highest brightness for O_2^+ , O^- and inert gases.
- For high brightness, you suffer with the short and unpredictable lifetimes.

Duoplasmatron	
Brightness ($Am^{-2}sr^{-1}V^{-1}$)	~ 500 (O_2^+) and ~ 5 (O^-)
Energy Spread (eV)	10-15eV
Lifetime	1 week – 6 months
Beam Stability	$\pm 1\%$ over 30 minutes
Gases	oxygen, hydrogen, inert



Long, Coathe and Drummond – Cambridge Univ – 1995

Cold Plasma Creation with Inductive Power Coupling



Time varying current in an external coil creates a time varying $\underline{B}$ field, which induces an azimuthal $\underline{E}$ field.

$$\nabla \times \underline{E} = -\frac{\partial \underline{B}}{\partial t}$$

Induction $\underline{E}$ field accelerates electrons to ionize resident gas.

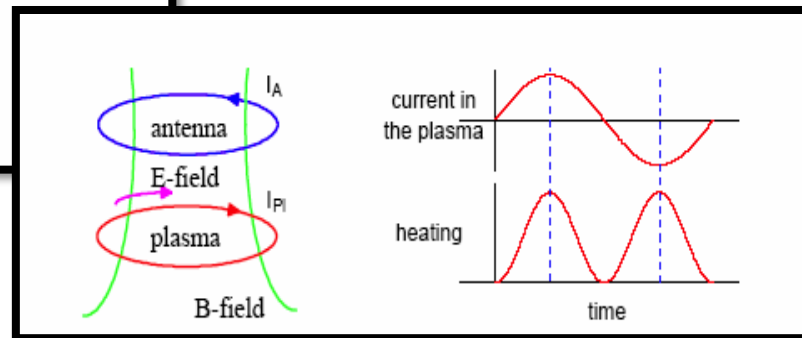
electron plasma frequency

$$\omega_{pe} \triangleq \left(\frac{n_0 e^2}{\epsilon_0 m_e} \right)^{1/2} \sim 9 \text{GHz}$$

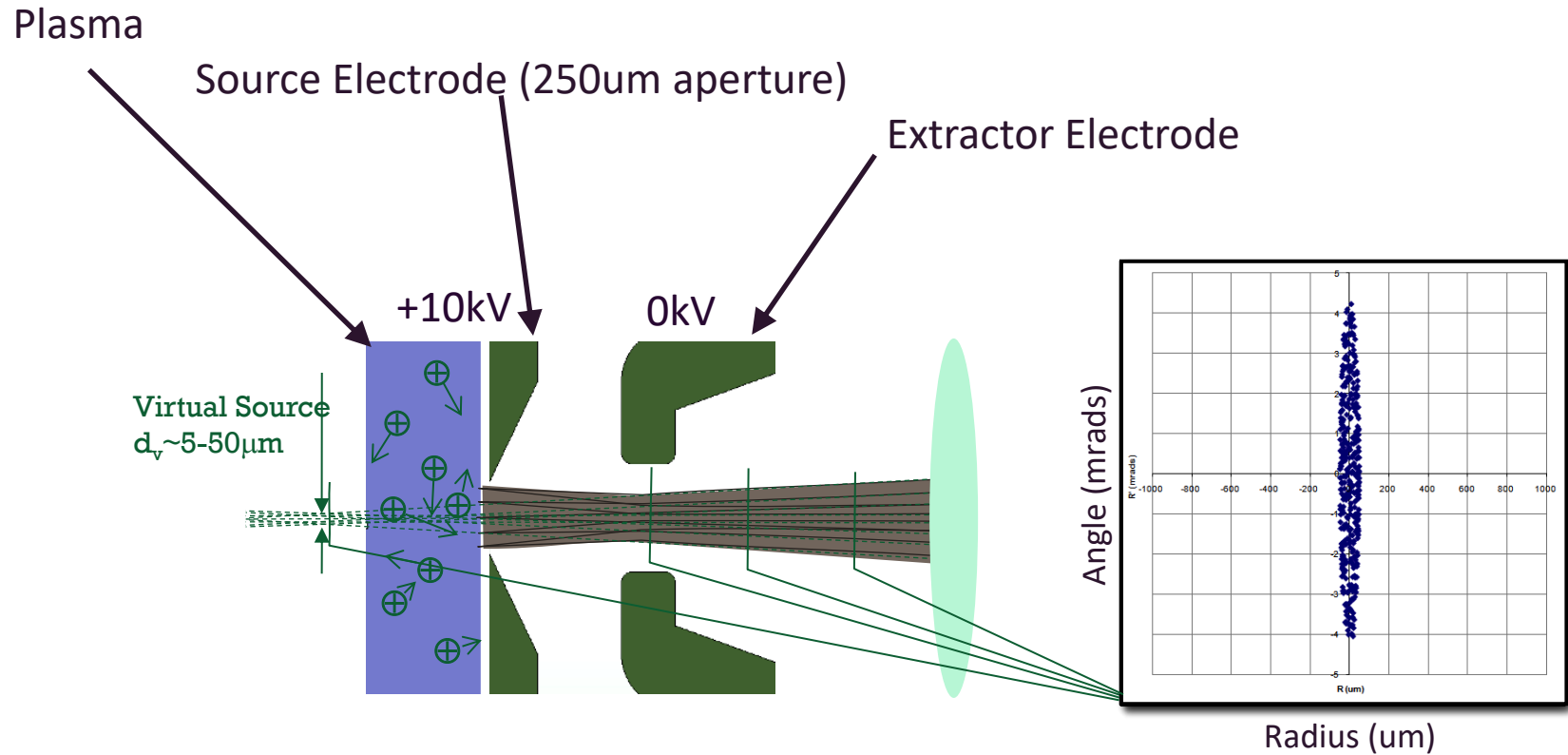
ion plasma frequency

$$\omega_{pi} \triangleq \left(\frac{n_0 e^2}{\epsilon_0 m_i} \right)^{1/2} \sim 18 \text{MHz}$$

(when Xe plasma density, $n_0 = 1 \times 10^{12} \text{cm}^{-3}$)

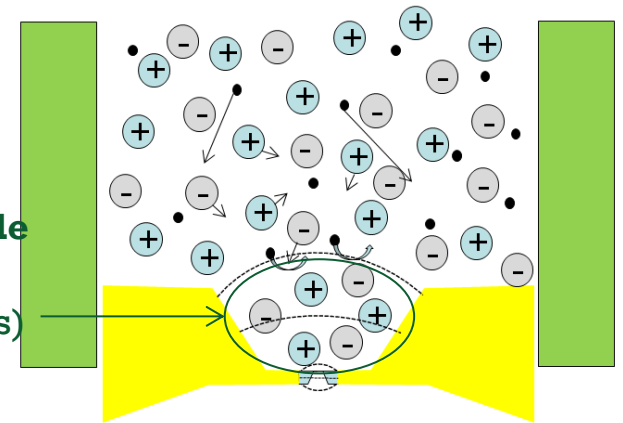
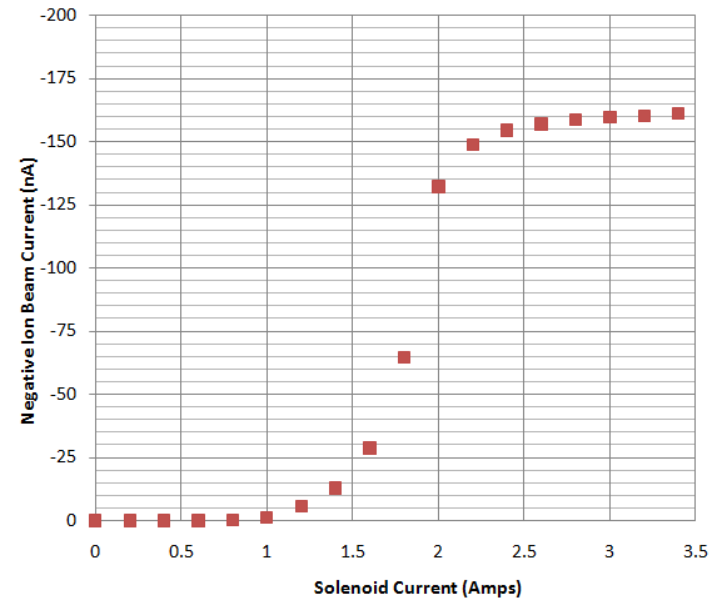
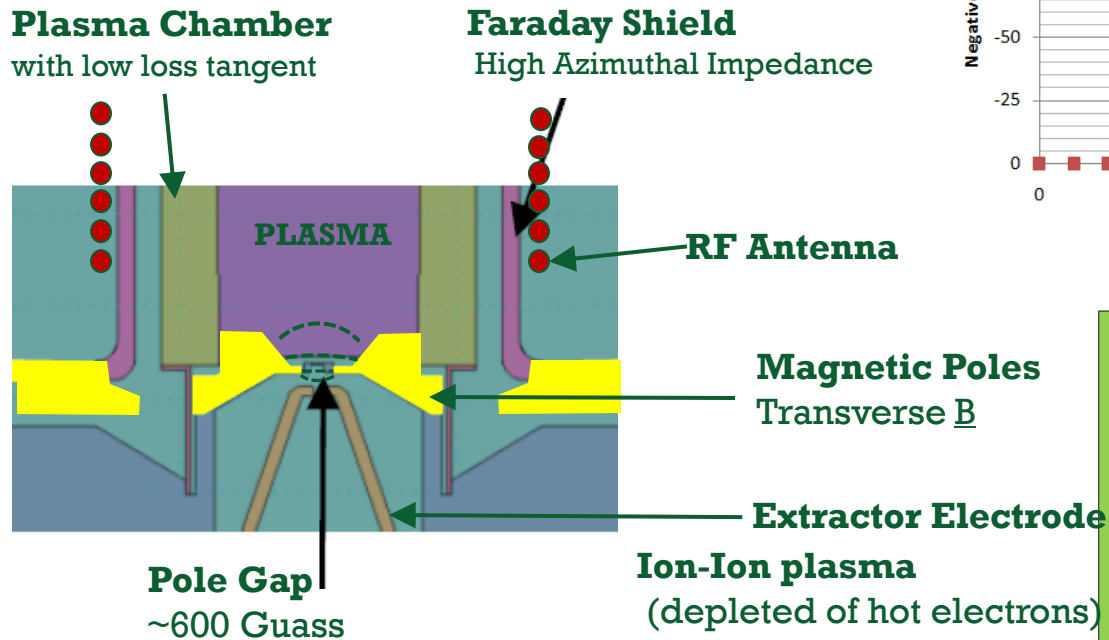


Ion Sources for Focused Ion Beams



Extracting Negative Ions (High Res. SIMS Imaging)

O⁻ for SIMS imaging of geological, semiconductor and biological materials

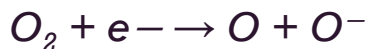


$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$

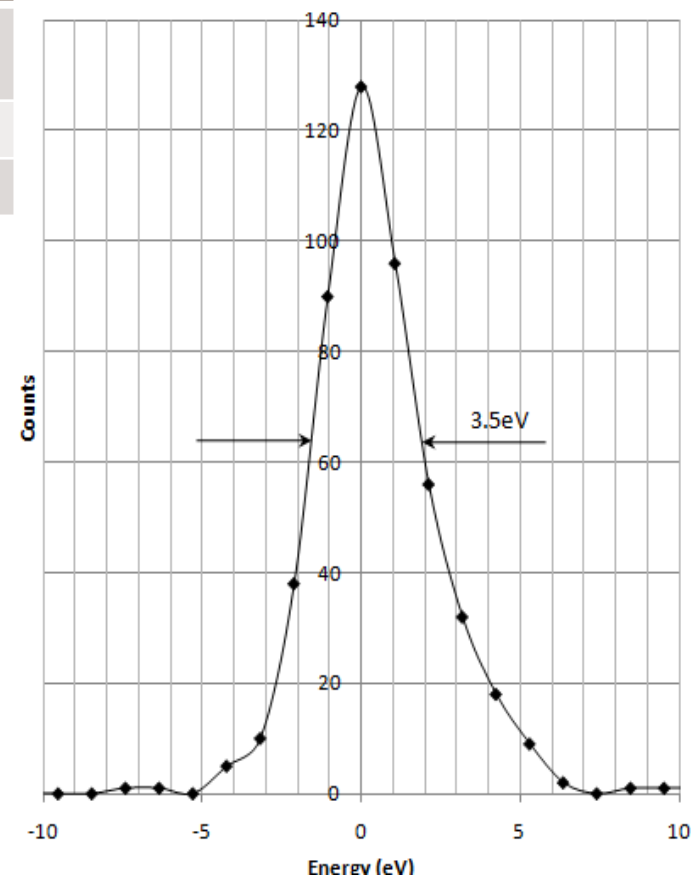
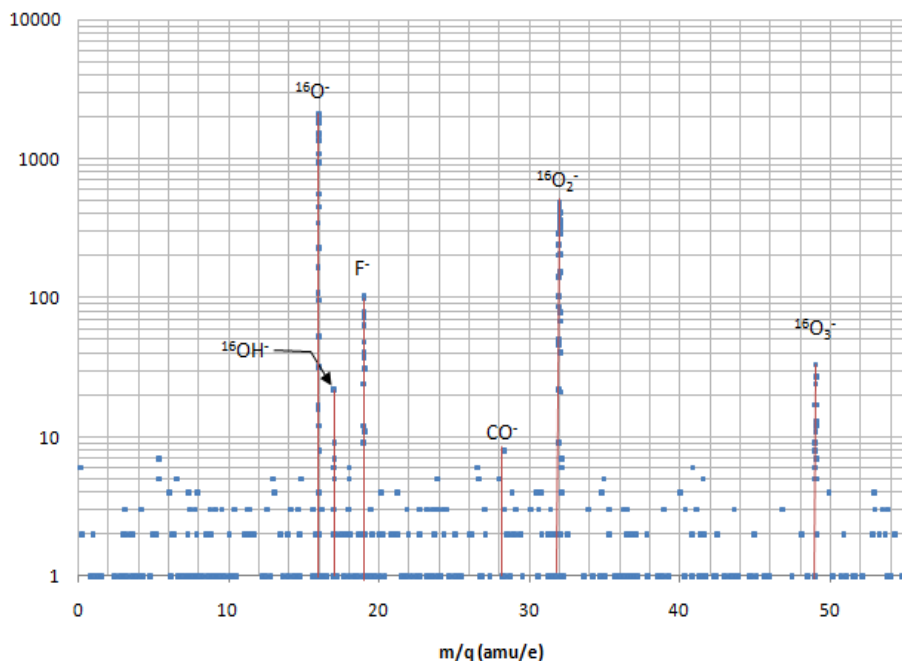
O⁻ Ion Source Parameters

Oxygen Plasma

O⁻ formed primarily by dissociative electron attachment.



	Duoplasmatron	ICP (Hyperion)
Energy Normalized Brightness (Am ⁻² sr ⁻¹ V ⁻¹)	5	100
Axial Energy Spread (eV)	10-15	3.5
Lifetime	1 week – 6 months	>2 years



Hyperion II - *Parameters*



- **0 to $\pm 30\text{kV}$**
- **Typically operated with Xe, O₂, He or N₂**
- **Energy Normalized Brightness**
 - + ions Xe: 1×10^4 , O₂: 4×10^3 , He: $6.5 \times 10^3 \text{ Am}^{-2}\text{sr}^{-1}\text{V}^{-1}$
 - - ions O: $1 \times 10^2 \text{ Am}^{-2}\text{sr}^{-1}\text{V}^{-1}$
- **Virtual source (dv)**
 - 5 μm or 12 μm (+ ions)
 - 50-70 μm (- ions)
- **Axial Energy Spread**
 - 5eV (+ ions)
 - 3.5eV (- ions)
- **Lifetime > 2 years**
- **Stability <1% drift/ hour, <5% drift/24 hours**



SIMS Imaging Performance at LLNL

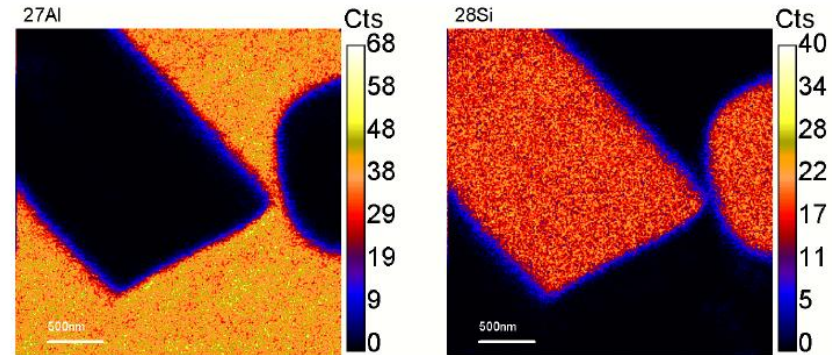


Hyperion II + LLNL
NanoSIMS

*Supported by DoE and Glenn Seaborg Inst.
(P. Weber, M. Kristo and I. Hutcheon)*

First Hyperion™ II installed on NanoSIMS50 (LLNL)

- Aluminum/Silicon sample
- Providing 40-50nm with 1pA O⁻ on Cameca NanoSIMS50



Data courtesy of Peter Weber (LLNL)

High Imaging Resolution Trace Element SIMS

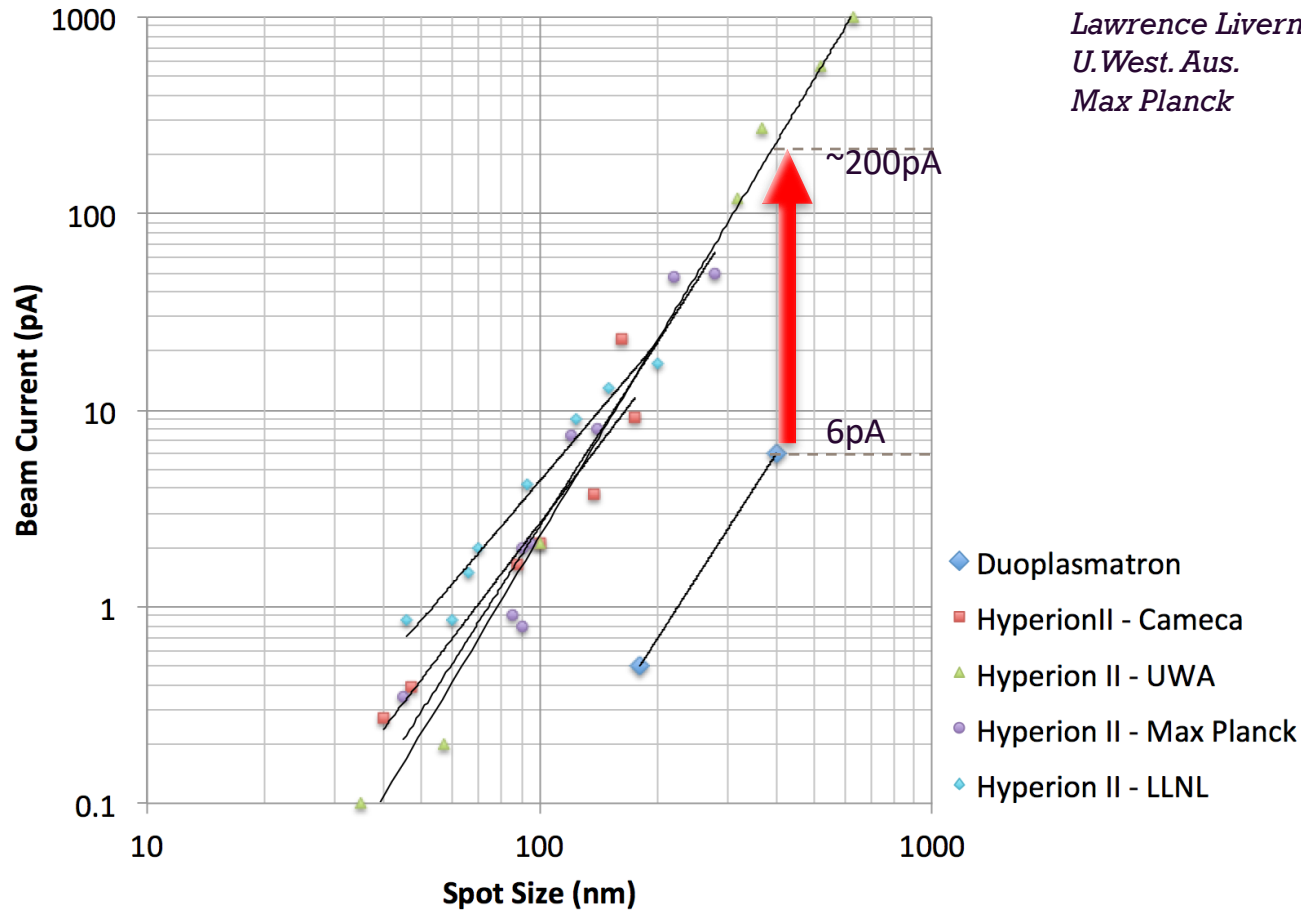
- Direct replacement for duoplasmatron on Cameca NanoSIMS.
- Image resolution, with O⁻ reduced from 200nm to 50nm.
- Lifetime increased from 1 week to >2 years.

Hyperion II on Cameca NanoSIMS50

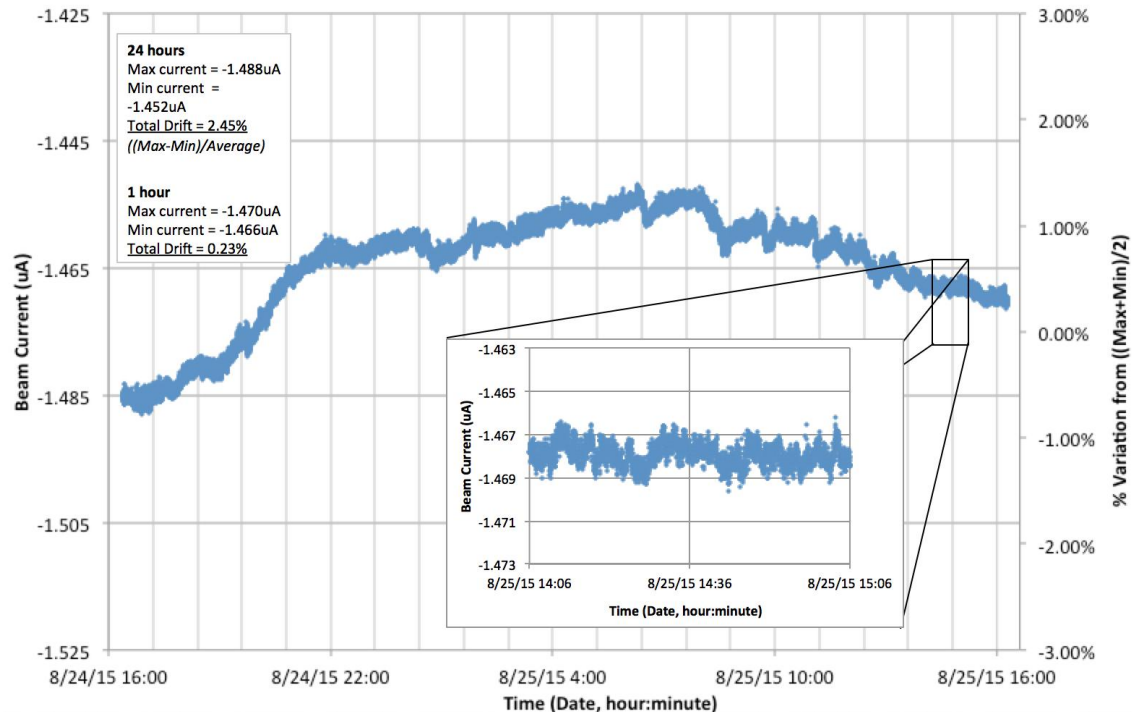
Data Courtesy of:

Cameca
Lawrence Livermore
U. West. Aus.
Max Planck

(F. Horreard)
(P. Weber)
(M. Kilburn)
(P. Hoppe)



Beam Current Stability



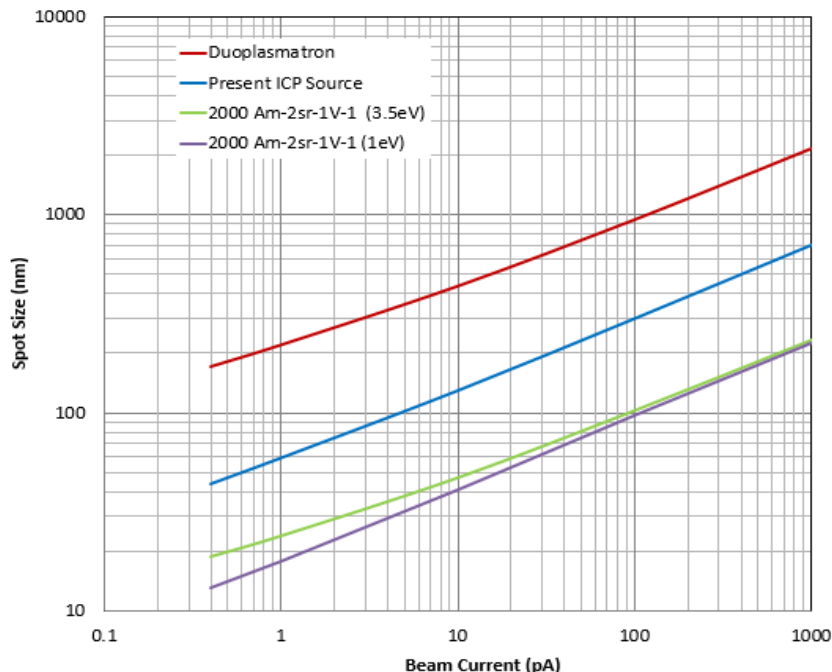
No sputtering in plasma source

With steady gas flow and temperature regulation, beam current is very stable.

Residual drift is primarily due to lab. temperature drift ($\sim 0.5\%/^{\circ}\text{C}$)

Possible Useful Developments

- High Brightness
 - Higher throughput
 - Higher spatial resolution imaging



- Molecular oxygen ions (O_2^- and O_3^-)
 - Higher Depth Resolution
 - Increased Useful Ion Yield
- O_3^- provides ~2x gain in useful ion yield for U isotope analysis*

	O^-	O_2^-	O_3^-
Typical	~80%	~19%	<1%
NIST	67%	20%	13%

* 'Improved Uranium Particle Analysis by SIMS using O_3^- Primary Ions' E. Groopman et al., NIST, J. Anal. At. Spectrom., 2022, 37(10)

THANKYOU!

