

Single Particle Uranium Analysis by Triple Quadrupole Time of Flight Mass Spectrometry for Nuclear Forensics



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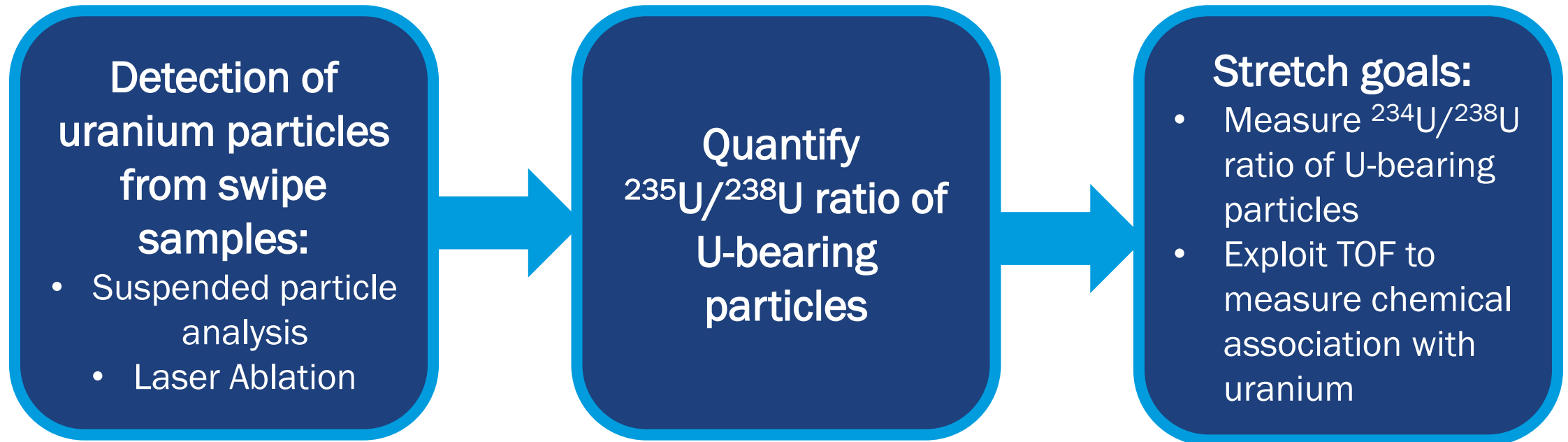
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Goals

Develop novel single particle analysis techniques for nuclear forensics using the Nu Vitesse time of flight inductively coupled plasma mass spectrometer (TOF-ICP-MS).



Compare performance of new single particle analysis techniques to conventional approaches

Background

- The IAEA's NWAL performs particle analysis of swipe samples for the monitoring of nuclear facilities with two focuses: [1][2]
 - Detection of U-bearing particles
 - Confirmation of declared enrichment
- Current analysis performed by LG-SIMS or FT-TIMS [3]
- The number of samples needing analysis is increasing rapidly, challenging the capacity of NWAL. [4]
- TOF provides a possible fast and cost-effective analytical screening method to keep up with the growing number of samples.

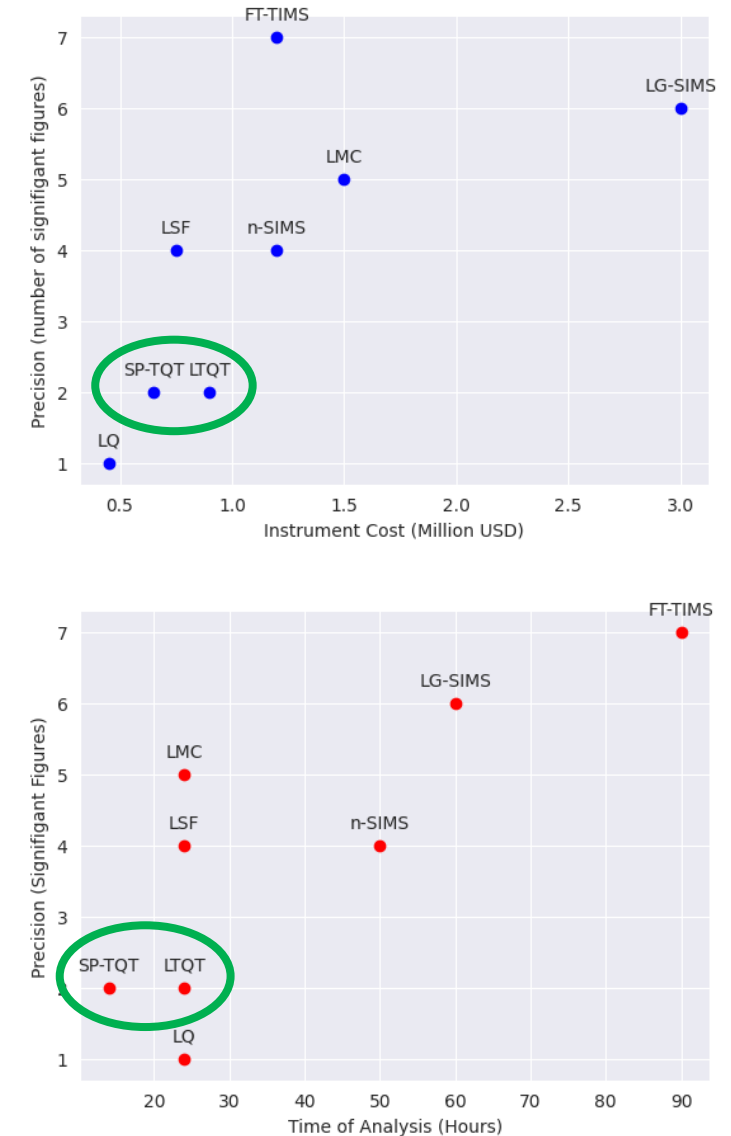
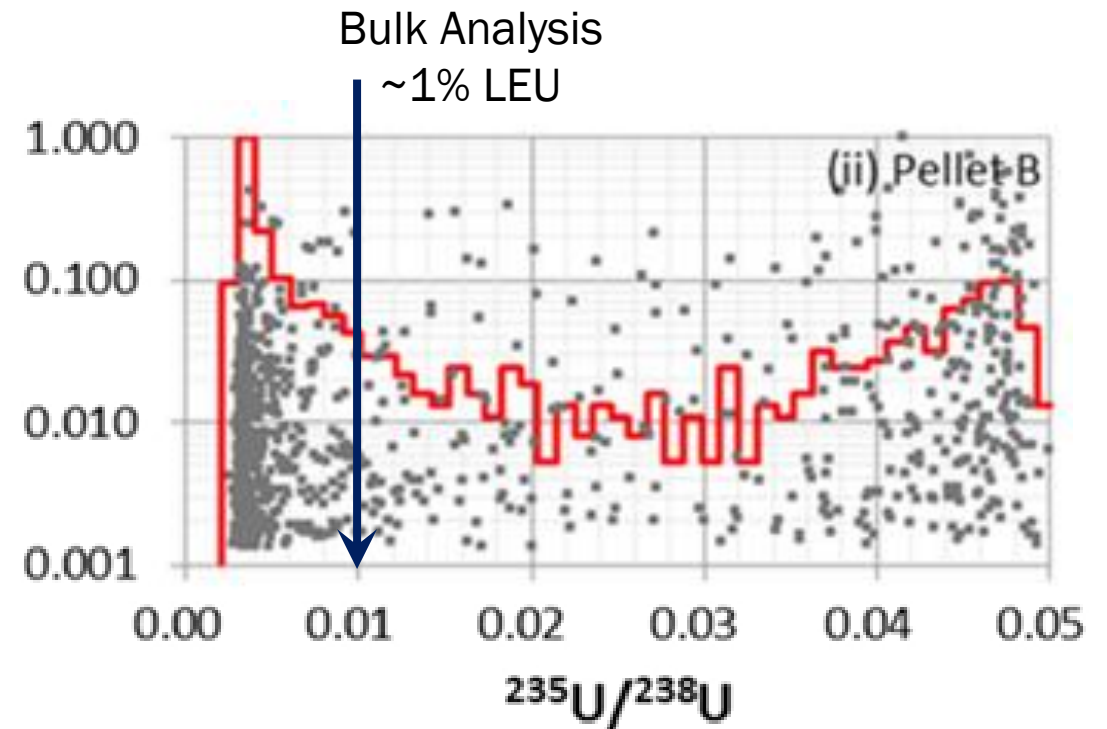


Figure 1: Approximate precision of methods compared to cost and analysis time.

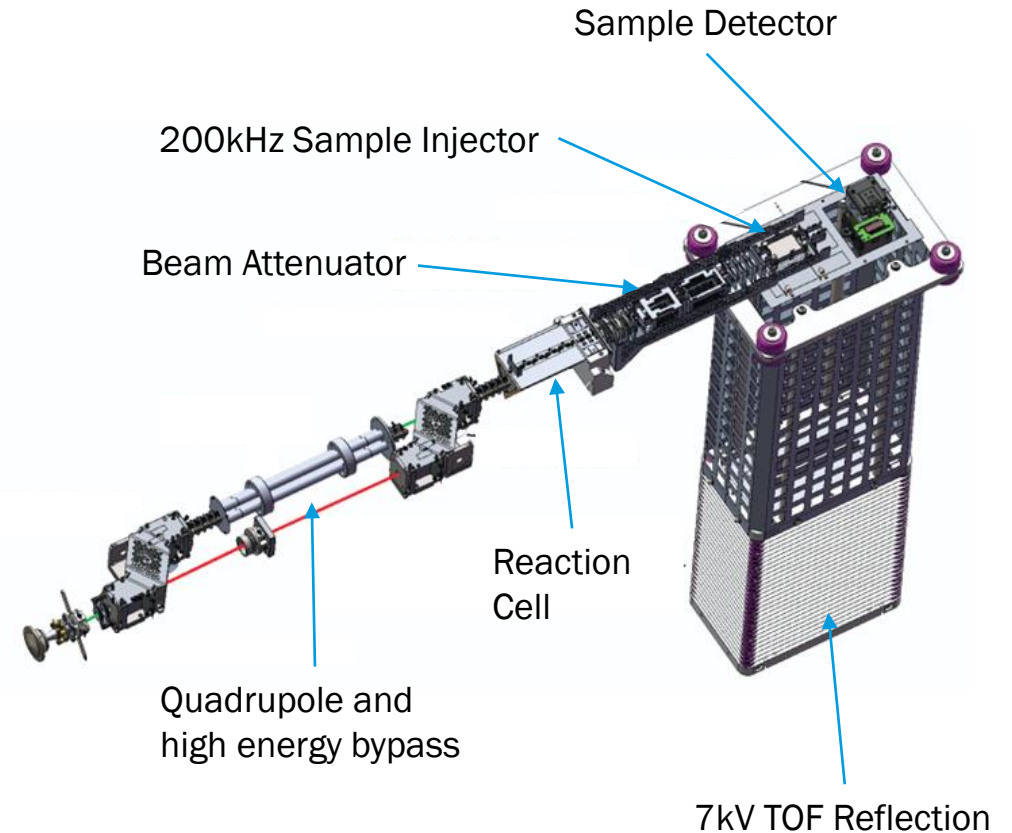
Single Particle Analysis VS Bulk Dissolution: Detecting a Needle in a Haystack

- Bulk dissolution homogenizes individual particles
- Homogenized particles give an average signal of the bulk make up
- Single particle analysis retains this information [4]
- Heterogeneity can be used to infer sample history [4]



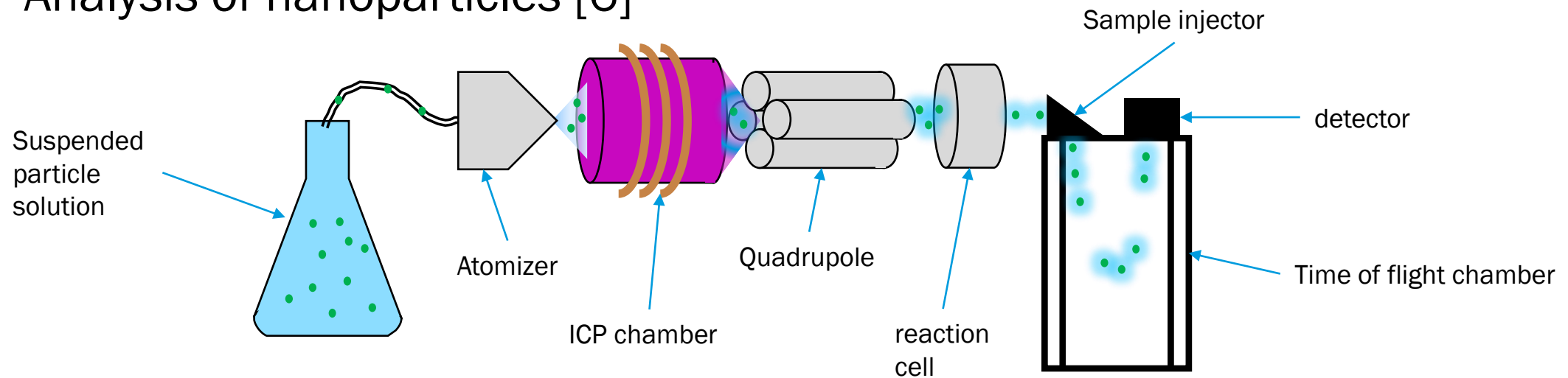
Nu Vitesse Inductively-Coupled-Plasma Time-of-Flight Mass-Spectrometer

- Triple Quadrupole addition (QQQ)
 - Prevents isobaric interferences
- Pairs with a laser ablation system
- Mass Range: 6-280 AMU
- Full range spectral acquisition in $30 \mu\text{s}$
- Dynamic Range: 10^6
 - 10^9 with ion beam attenuator
- ^{238}U sensitivity bulk solution:
 - $>50,000$ cps/ppb
 - $>160,000$ cps/ppb with beam attenuator



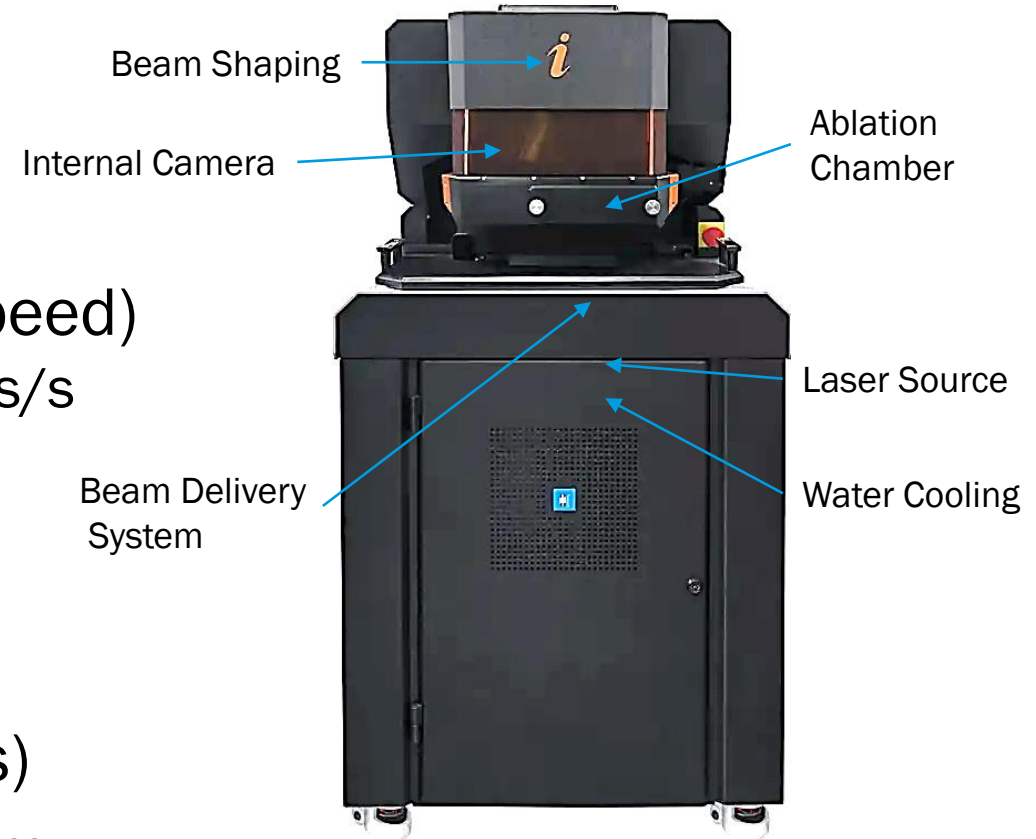
Suspended Particles in Solution

- Fast acquisition speed of TOF system allows differentiation of suspended particles from solution [6]
 - Continuously inject sample and rapidly take spectra
 - Signal from suspended particles superable from background solvent
- Corelate spectra in time for chemical analysis differentiate form of uranium
- Analysis of nanoparticles [6]



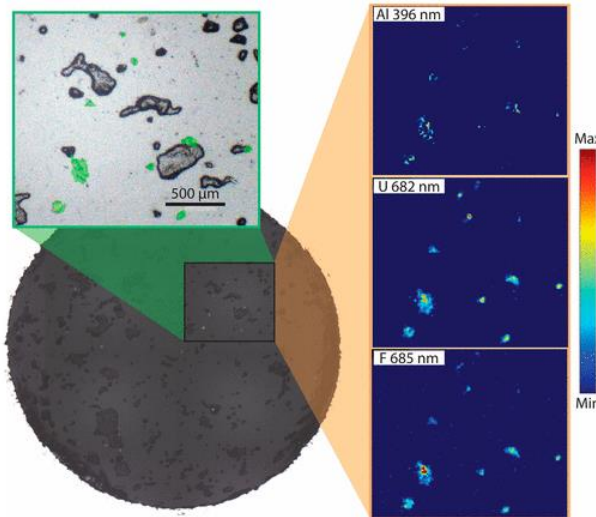
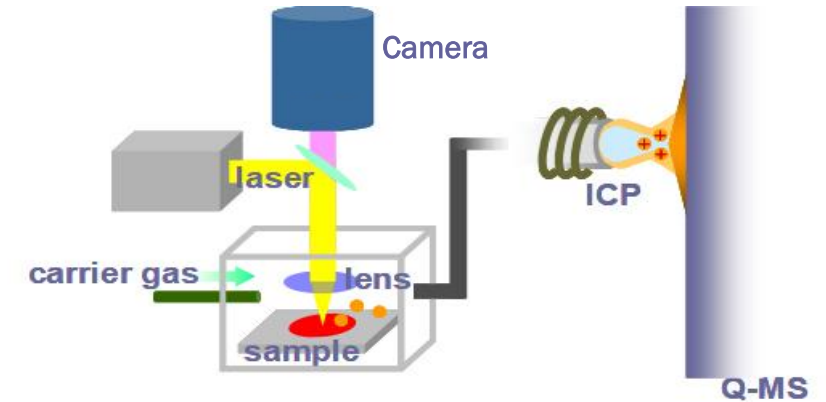
ESL Image Geo 193 Laser Ablation System

- 193nm ArF excimer laser
- $>15 \text{ J/cm}^2$ fluence
- 1-220 μm variable spot size
- Fast washout (limiting factor analysis speed)
 - $<1\text{ms}$ single pulse response for 1000 pixels/s scan rate
- Sealed precision stage to prevent contamination
- Internal camera for imaging target area ($<0.55\mu\text{m}$ resolution, 20x objective lens)
- ^{238}U sensitivity with laser ablation system:
 - $>250 \text{ cps/ppm}$



Laser Ablation from Swipe Sample

- Laser ablation allows ablation of particles on a swipe sample or solid samples
- Directly ablate selected particles
- Spatial isotopic imaging



- Dry sample prep is fast and allows other imaging techniques such as SEM
- Modular system allows combination with other instruments such as laser induced breakdown spectrometer

TOF corelative chemical analysis

- Able to differentiate chemical form e.g. UO_2F_2 , UF_4 , yellowcake, nat. U minerals
- Methods of performing corelative chemical analysis:
 - Spatial correlation of sample with laser ($\sim 1\mu\text{m}$)
 - Time correlation with the TOF
- Knowing U form gives information about process history, intended use, source



Current Status

Step	Status
Instrument acquisition	Completed
Lab renovations	June 2026
Instrument installation	July 2026
Method development	Ongoing

Conclusions

- Has potential to support nuclear forensic applications for single particle analysis by increasing the speed of analysis and reducing cost
- Comes at the expense of sensitivity
- Nanoparticle analysis of suspension provides a great size advantage to
- Pursuing development of two particle analysis techniques:
 - Direct suspension measurement
 - Laser ablation

Acknowledgements

This work was developed in collaboration with colleagues of the Consortium for Nuclear Forensics who are funded by Department of Energy, National Nuclear Security Administration award number DE-NA0004142.

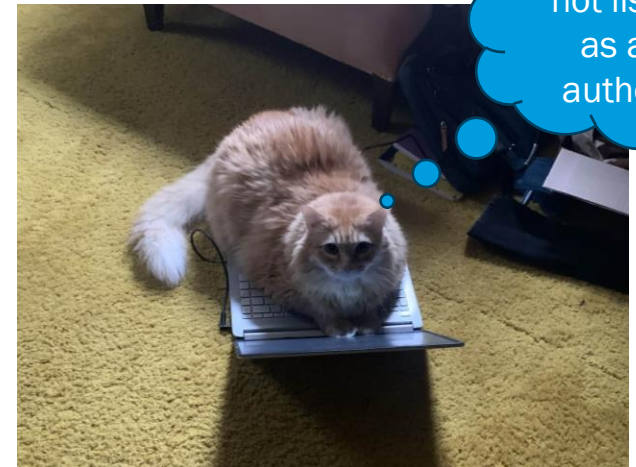


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Why am I
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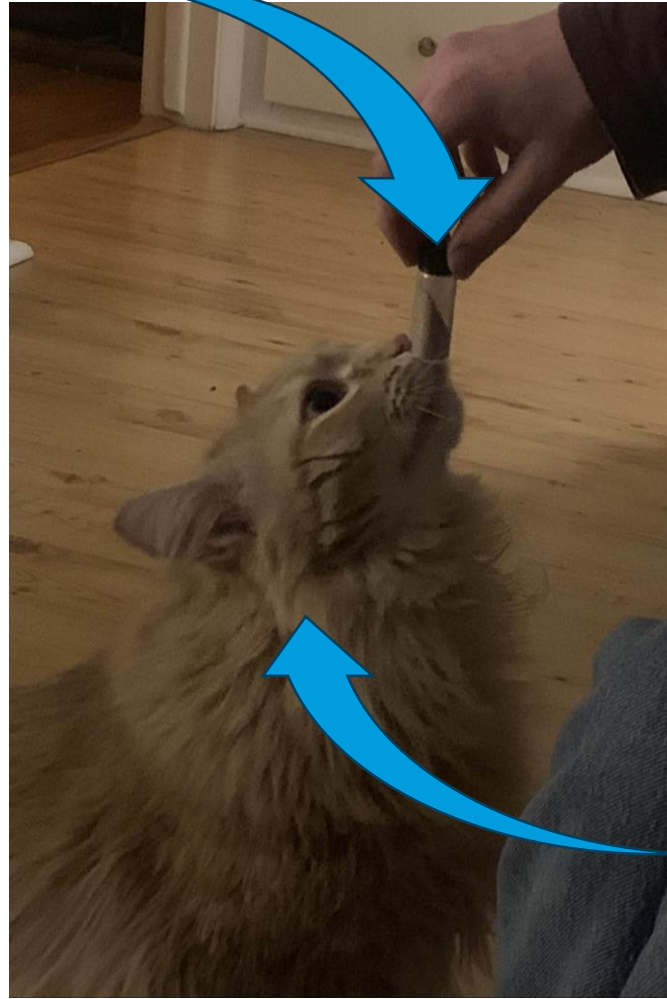
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Questions?

Sample?



Mass spectrometer?