X-Ray imaging for Earth & Planetary Sciences

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General strategy

WHY ? Nature is heterogeneous

Scale: l.y., psec, km, m, mm μm, **nm**

Nondistructive analysis:

- composition by quantification methods - challenging
- structure/morphology
- local chemistry

Access to:

- initial material (interstellar)
- Solar system re-processed – thermodynamics

Group work on similar samples needs:

- common strategy (methods, standards, databases)
- round-robin tests to evaluate accuracy

UJF: Masters « Terre, Univers et Environnement »
The « sample » community

- ★ crustal samples - feldspars, silicates
- ★ *in situ*: HP (< 300 Mb), HT < 4000°, bio, paleo
- ★ geo-material sciences: physico-chemical properties
- ★ hydrology: hydrated minerals; porosity; permeability
- ★ atmospheric: aerosols, ultra trace elements
- ★ soil/river pollution; radioactive/chemical/biological
- ★ planetary/interstellar return missions

EPS samples are complex...

EPS: sample is everything

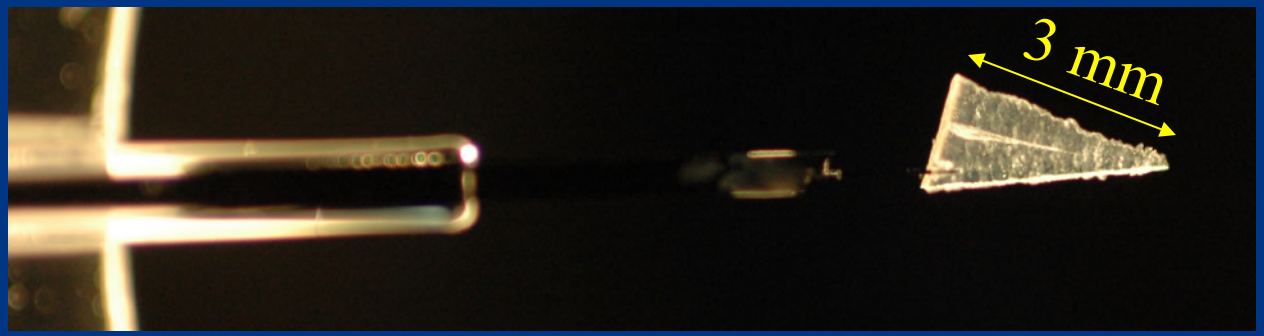
Unique, fragile, challenging samples

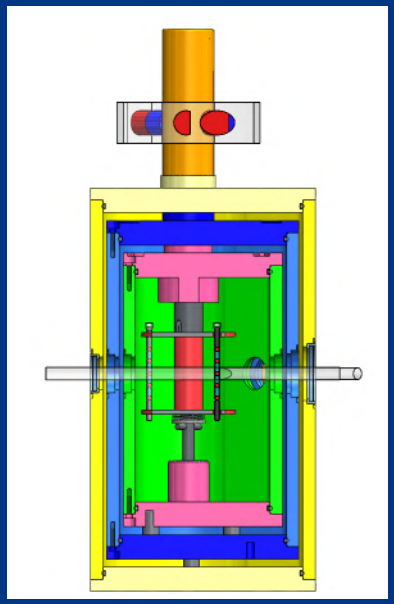
- nondestructive, non-invasive analysis
- sample environment: *in situ*, HP, HT, bio
- hyperspectral analysis: 2-3 simultaneous analyses
- multiscale: sample heterogeneity

In situ

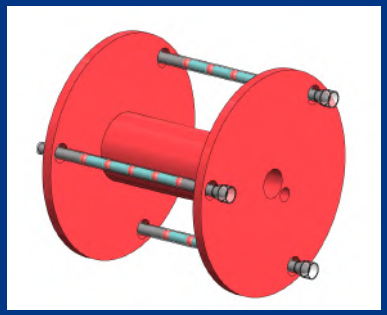
MSR

STARDUST aerogel keystones





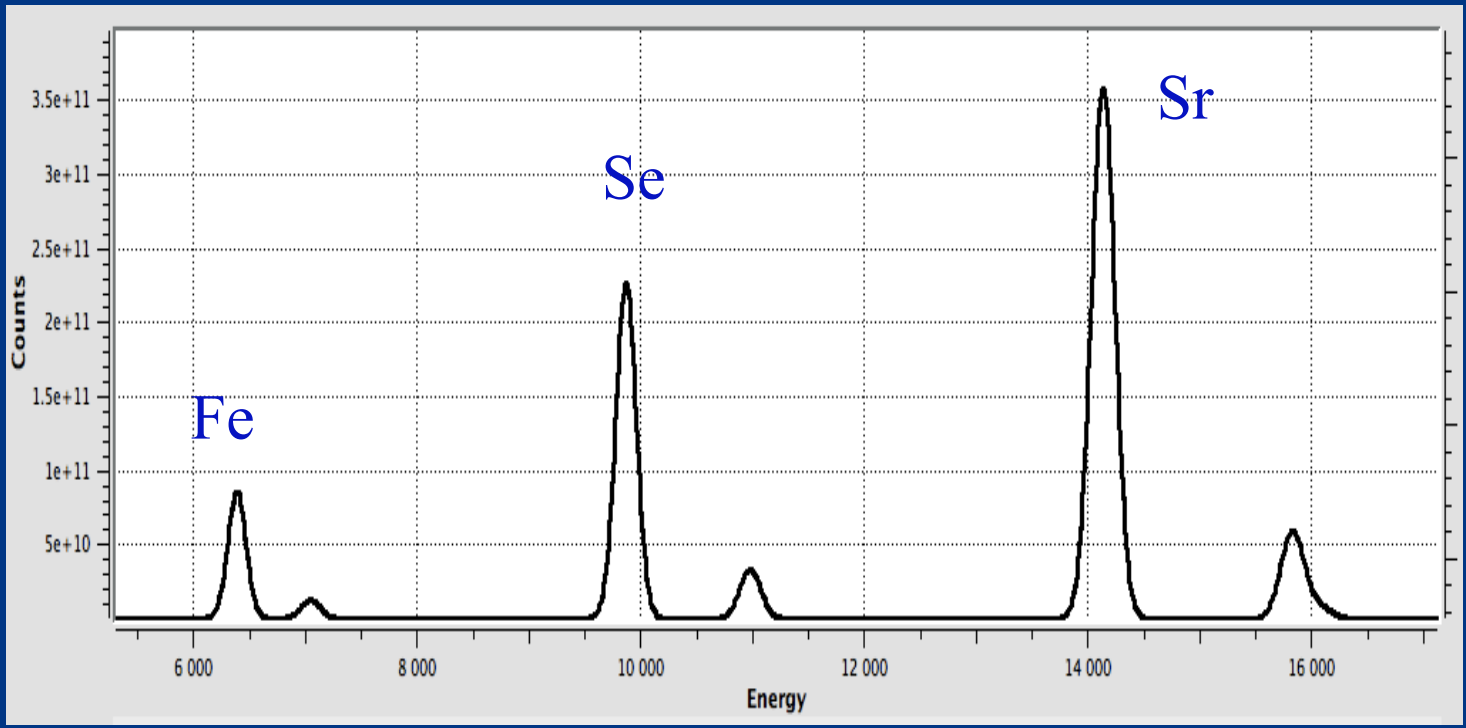
Diamond Anvil Cell



Missions with Sample Return:

Mars > 2022

Example of quantification



E_0=18 keV

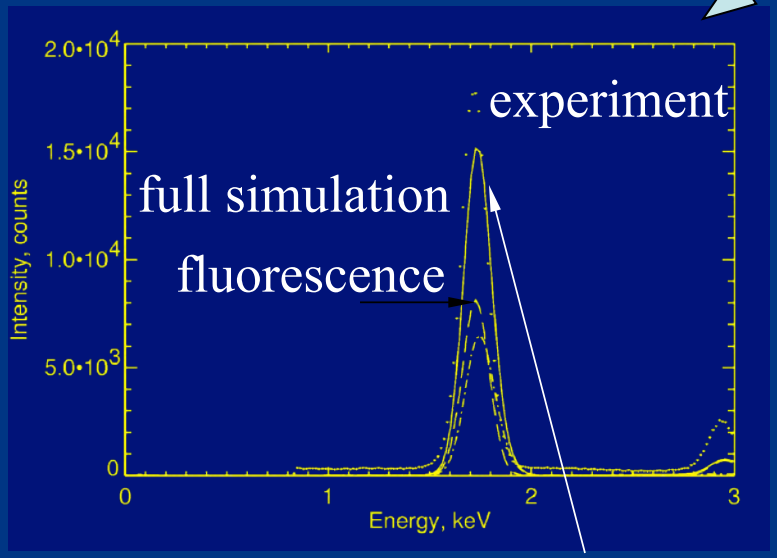
$C_{Fe} = C_{Se} = C_{Sr}$!!!!

Effect of Z, (E_0-E_K), ρ, A, ε, μ

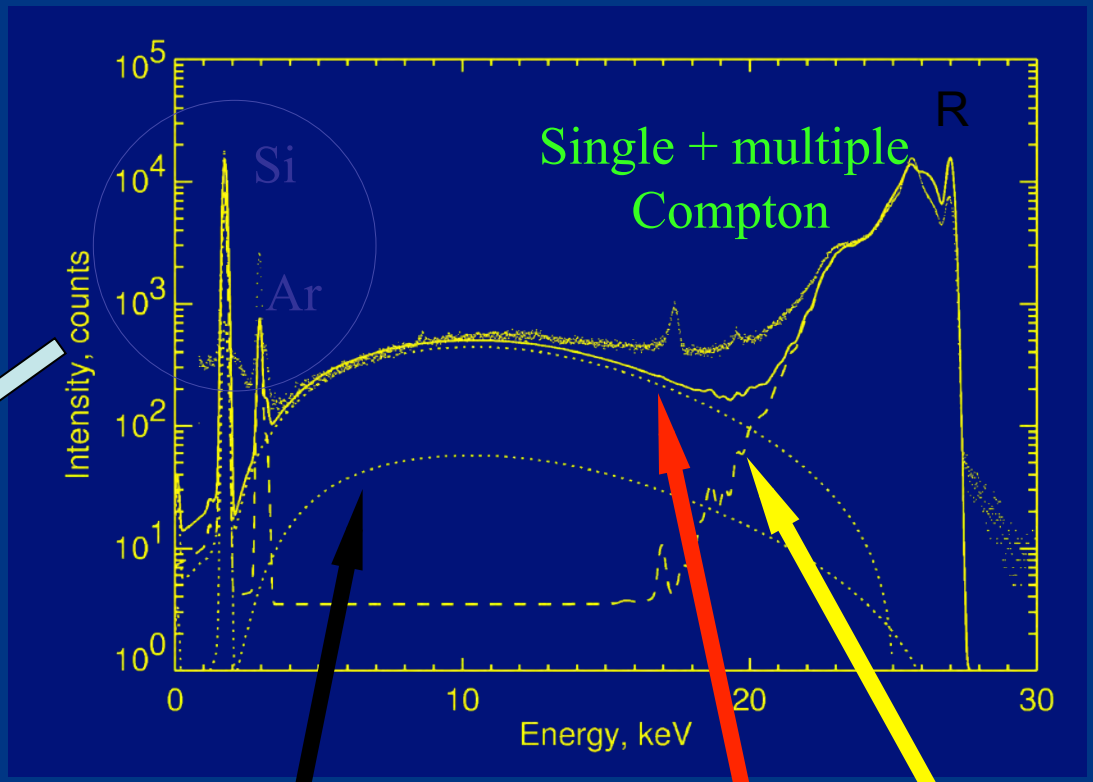
Relative intensities strongly **changed**

Accurate quantification by Monte-Carlo Collaboration L. Vincze, Univ. of Ghent

250 μm thick Si-wafer
Live time: 1000 s
Instrument: ID18F
E_0 = 27 keV
$E_{photoelectron}$ = 25.16 keV



photoelectron impact ionization
CASINO



K photoelectron bremsstrahlung

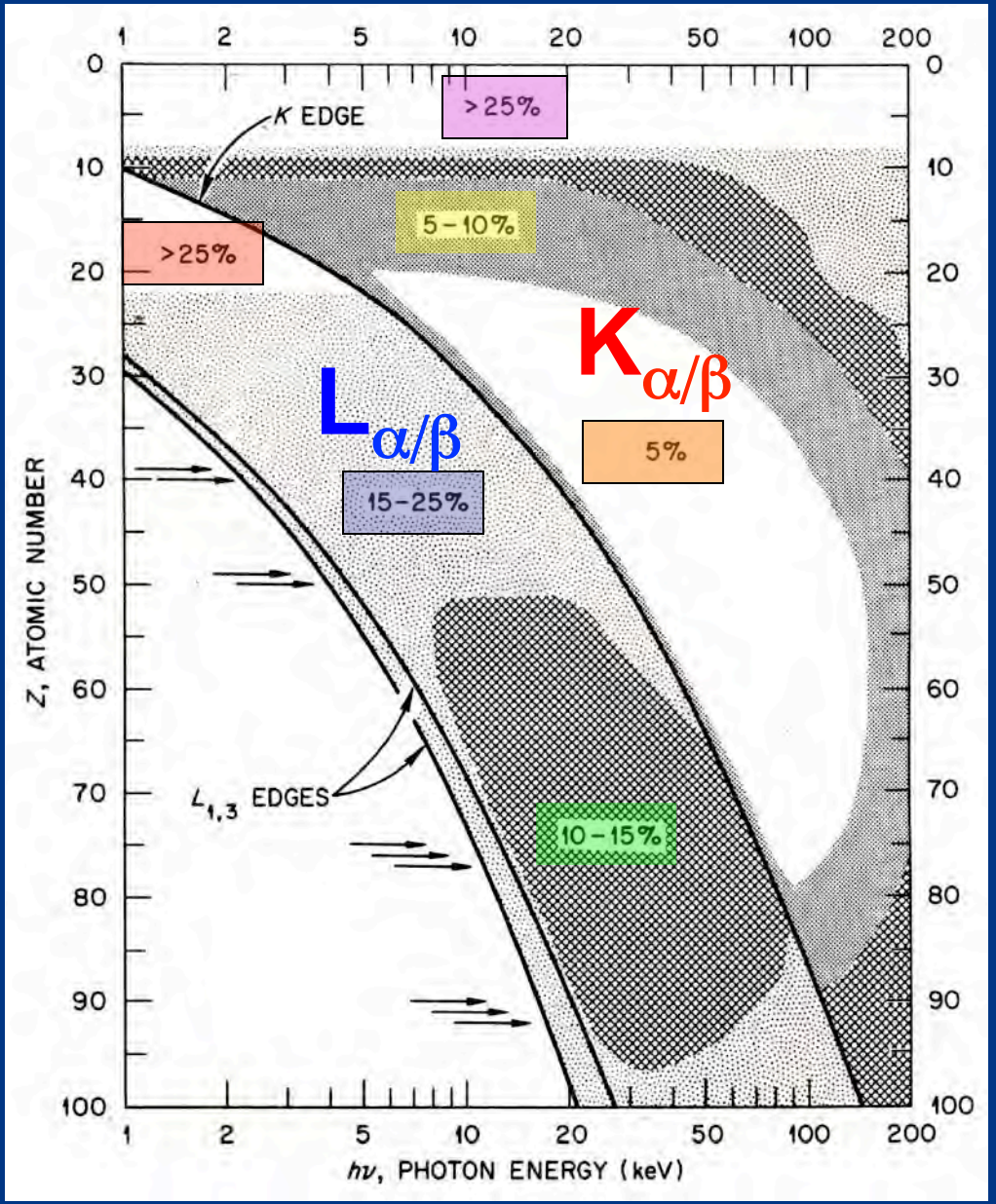
L photoelectron bremsstrahlung

Simulation without photoelectron tracing

Fluorescence cross-sections σ

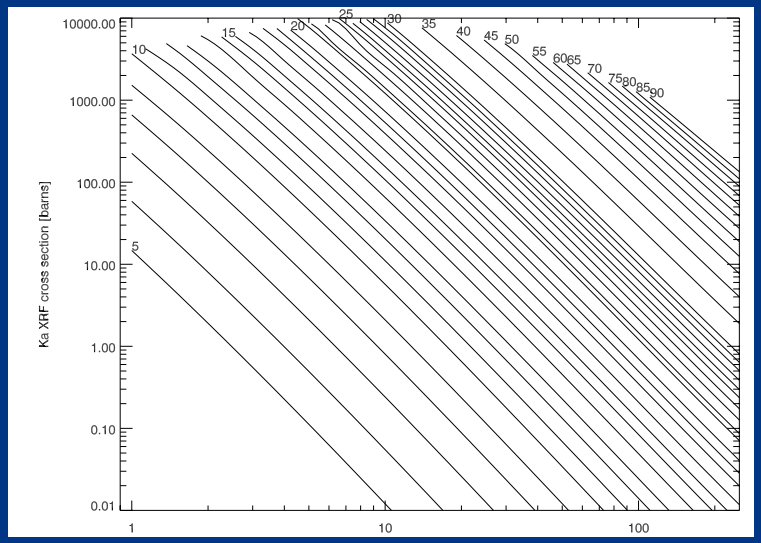
Uncertainties in the tabulated fluorescence cross-sections for the K_α and L_α lines

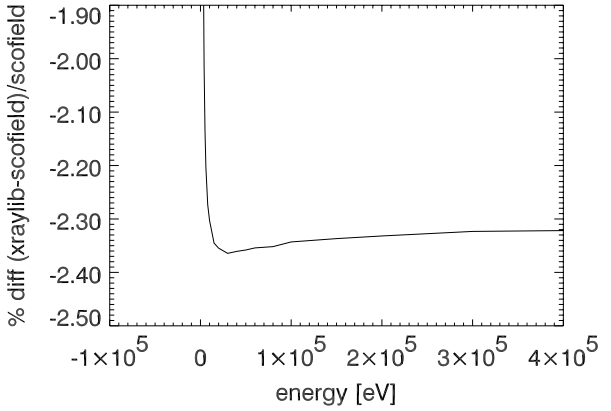
M.O. Krause *et al.*, ORNL-5399

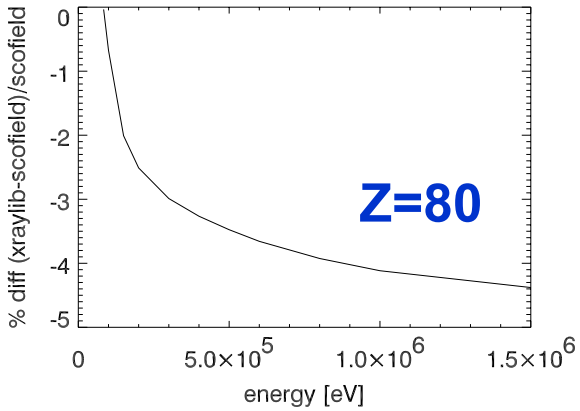


Tools:

X-ray database: XRAYLIB







- ✓ multi-OS/multi-language
- ✓ fluorescence/absorption
- ✓ Compton/Rayleigh (pol / non-pol)
- ✓ form-factor, scattering function
- ✓ transition/edge energies

http://ftp.esrf.fr/pub/scisoft/xraylib/

Brunetti, Sanchez del Rio, Golosio, Simionovici, Somogyi
Spect. Acta B 59, 1725-1731, 2004

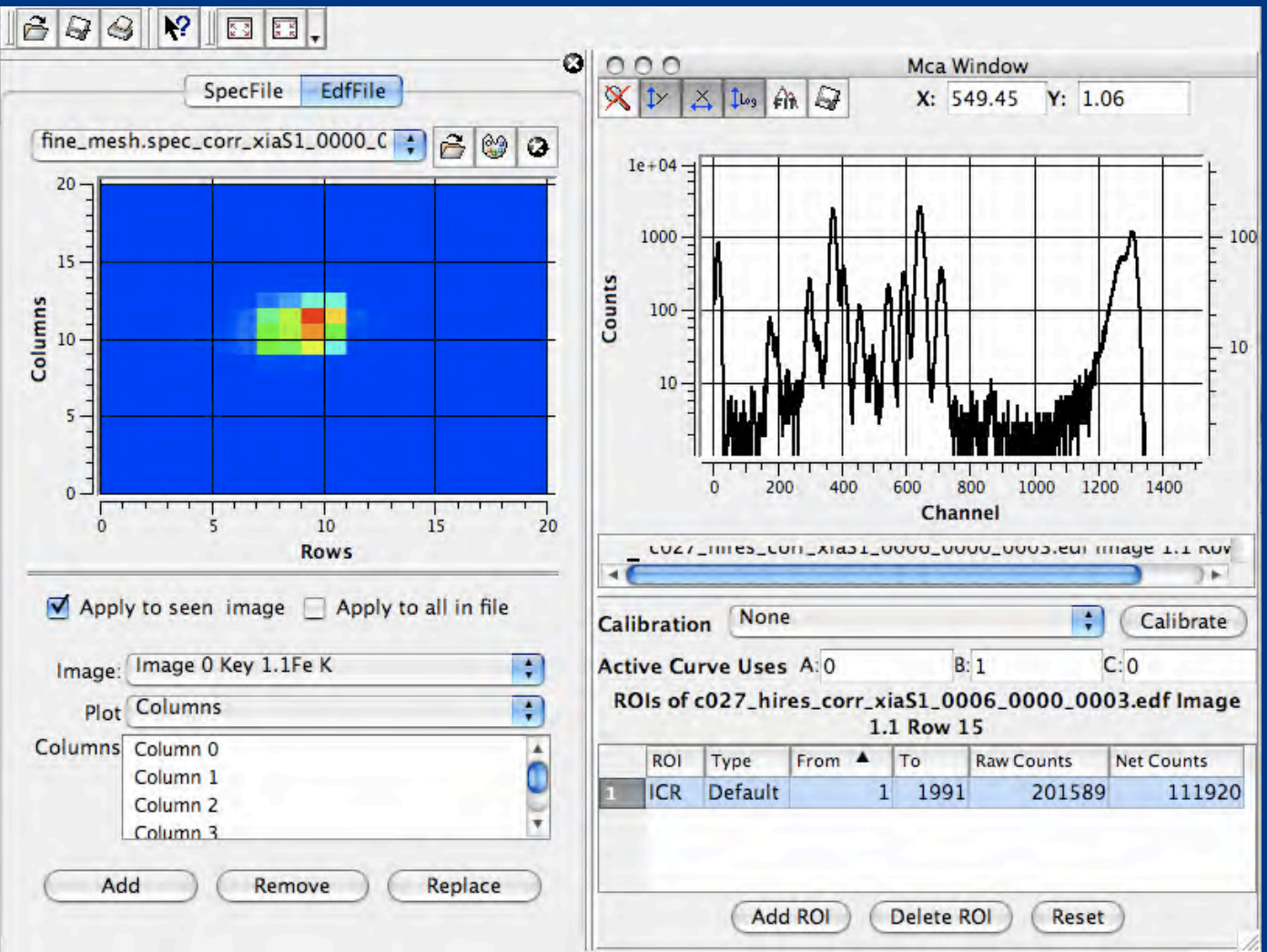
Tools:

PyMCA , hyperspectral XRF+XRD,
Spectrum fitting & quantification

ESRF BLISS group:

Armando Sole

http://pymca.sourceforge.net/download.html



XRF+XRD

Soon

XRF+Raman

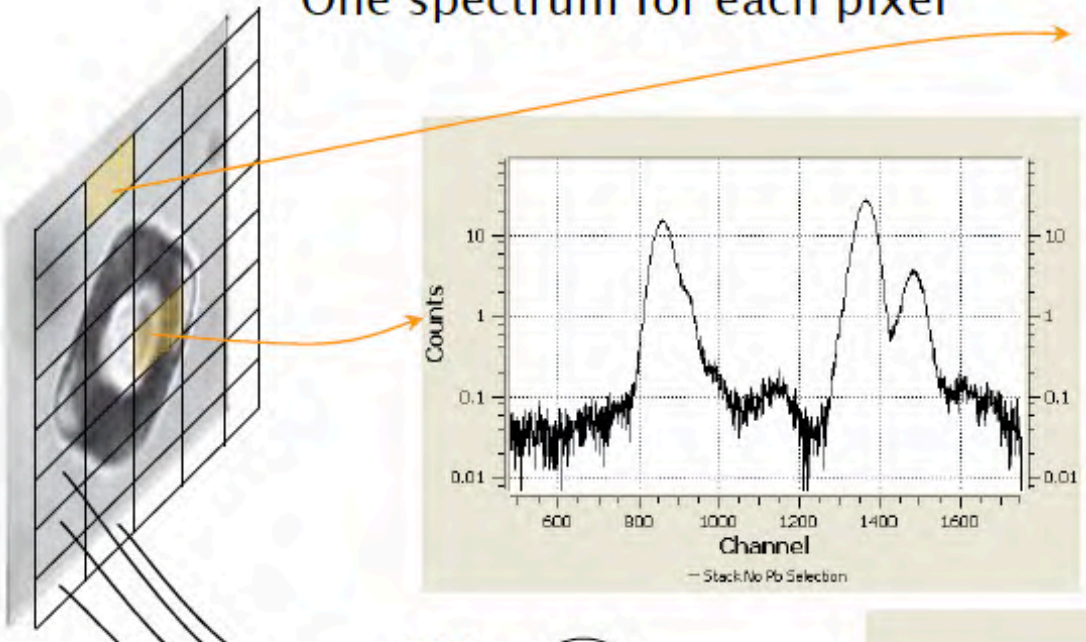
XRF+IR

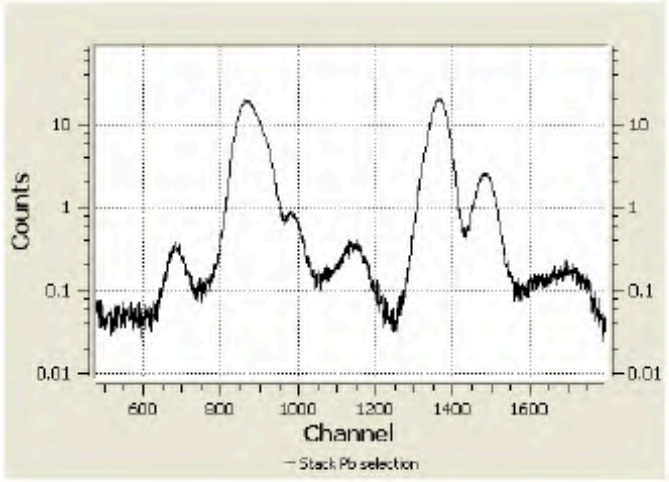
XRF+Xanes

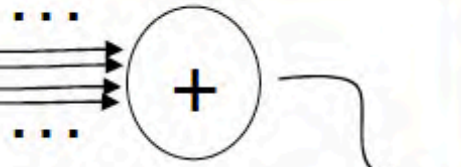
F-CT

2D Imaging

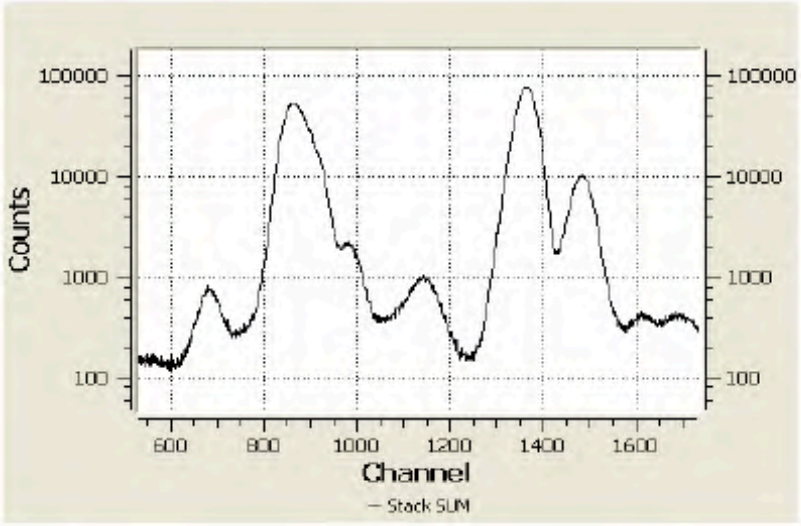
One spectrum for each pixel





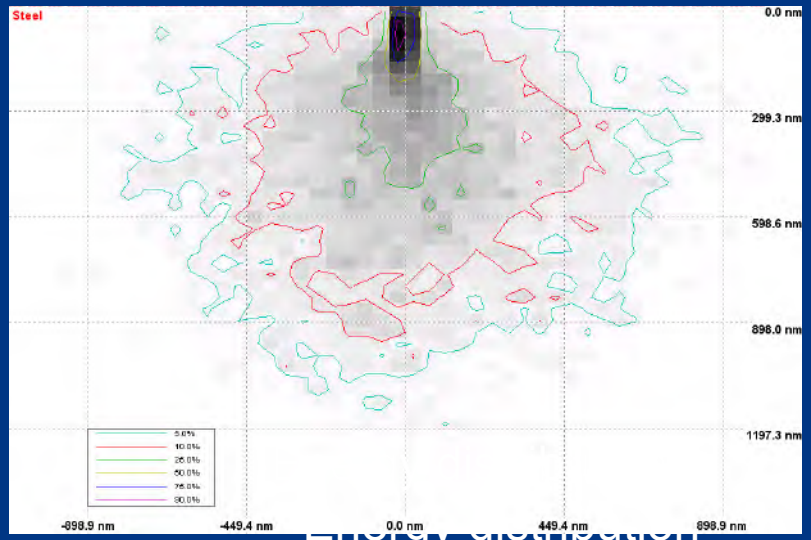


A sum spectrum for the whole image

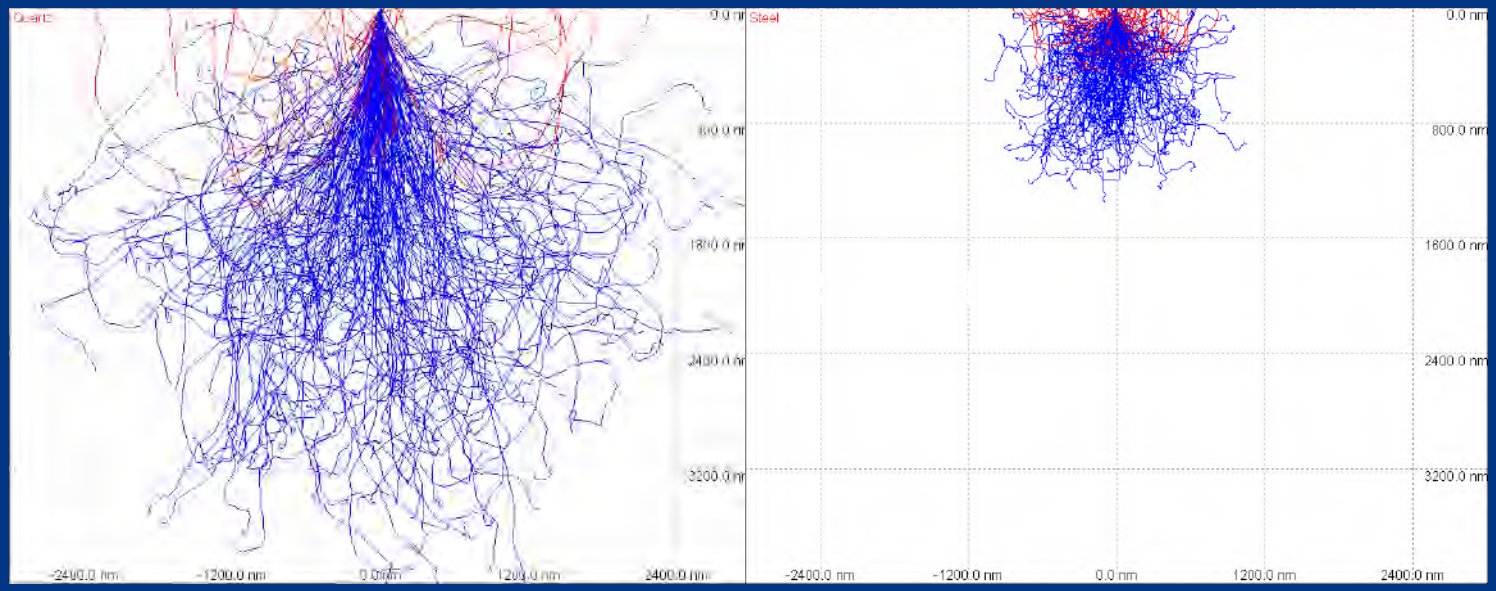


e--beam mapping

"skin" mapping
E = 20 keV

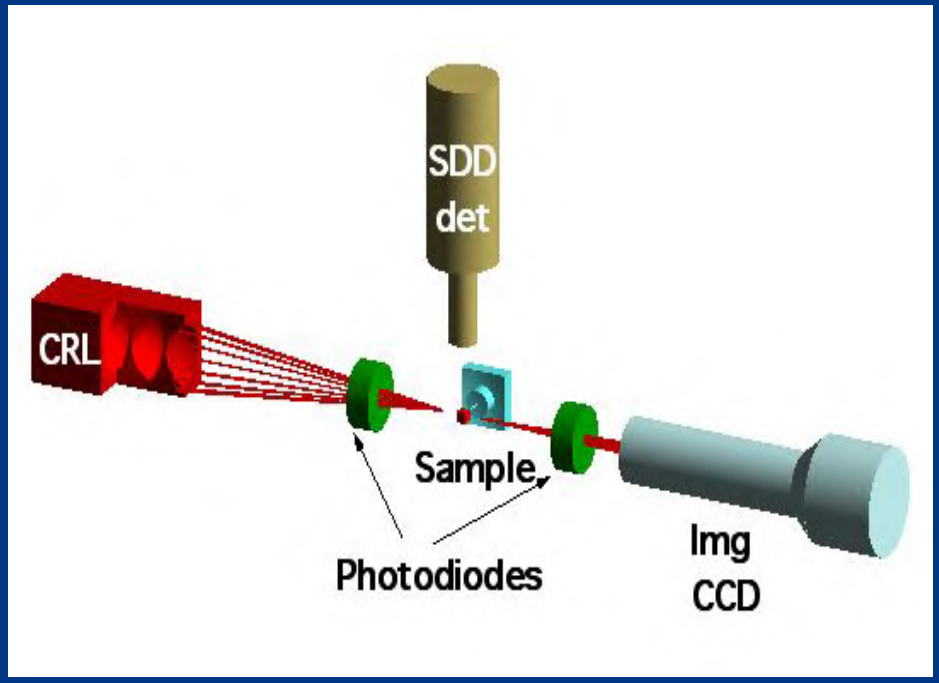


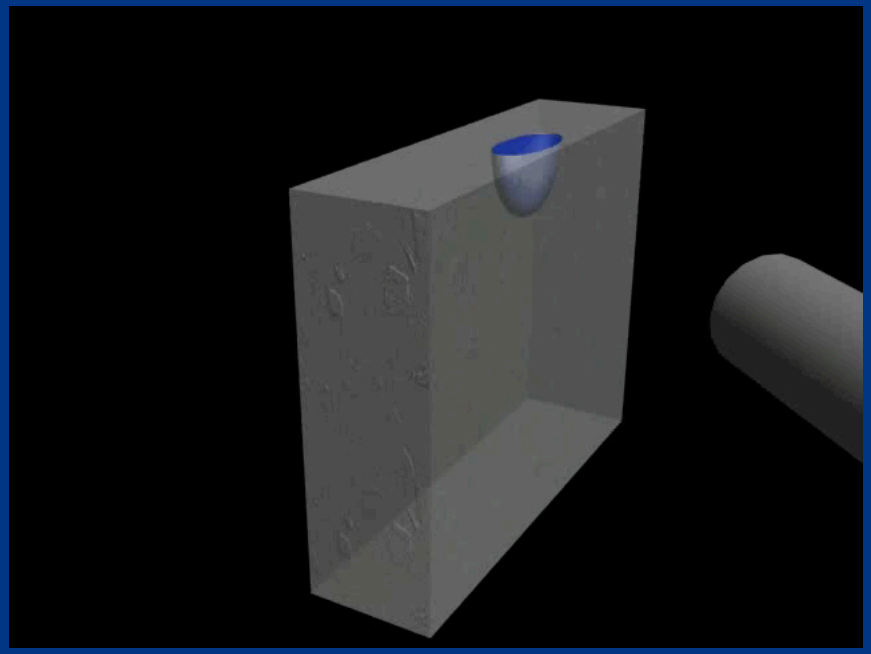
Energy distribution



✓ **resolution ~ μm**
✓ **local concentrations > 1‰**

How to map thicker samples
Solution : fluorescence tomography

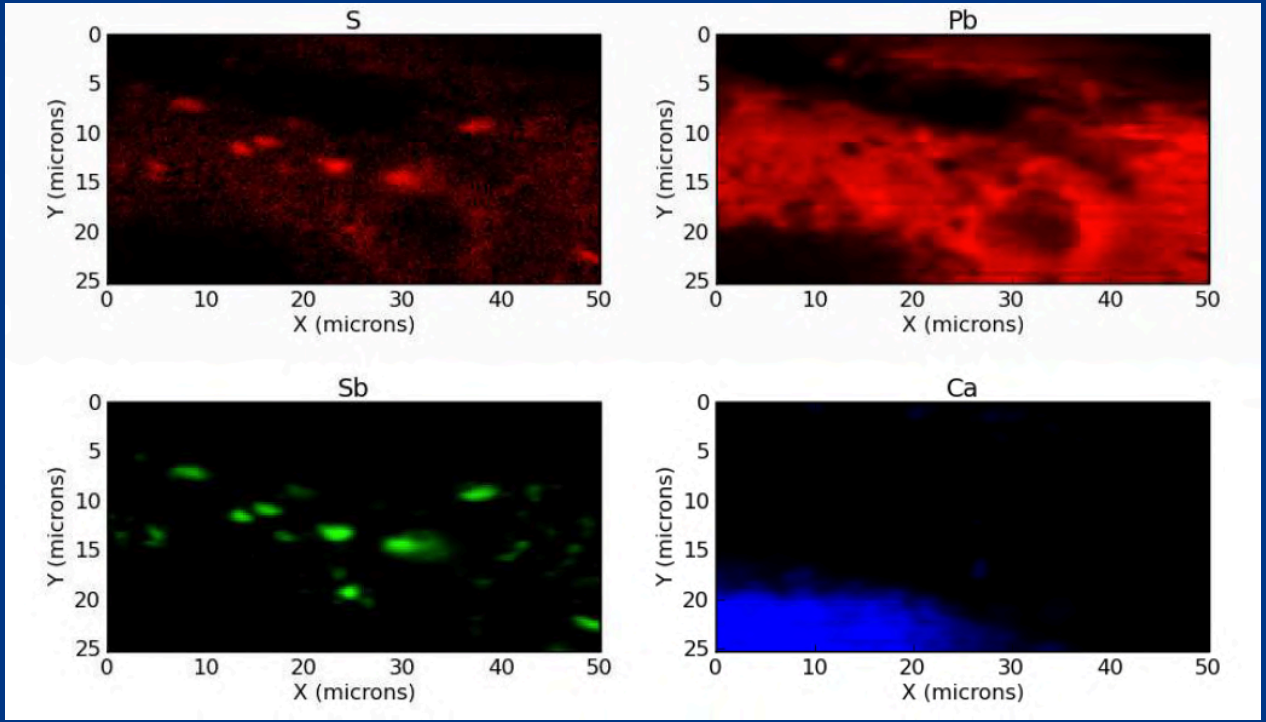


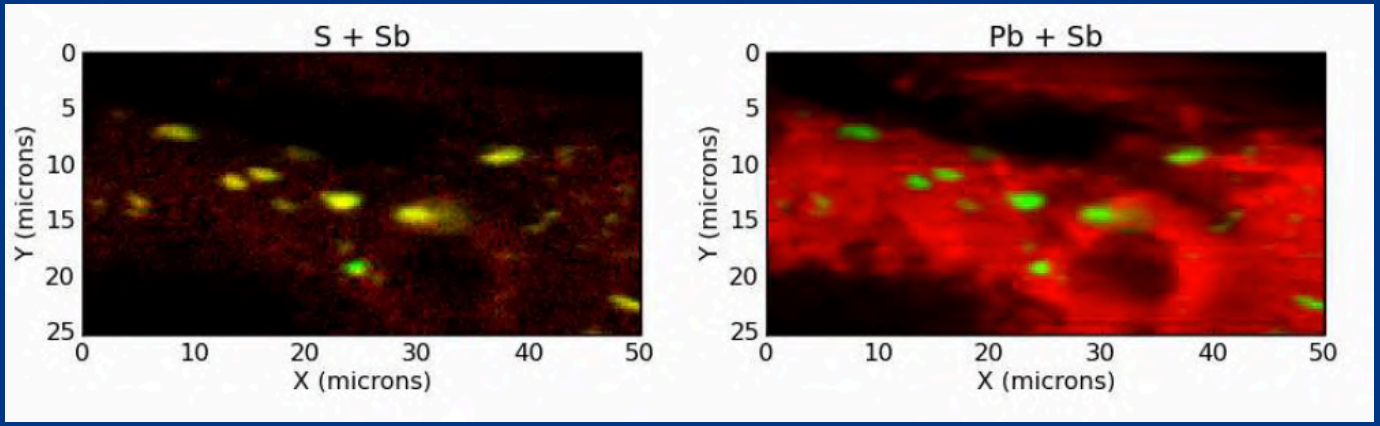


✓ polar scan z et θ
✓ long acquisition
✓ reconstruction : inverse problem

Simionovici et al.,
IEEE Trans. Nucl. Sci., 47, 2736-2740 (2000)
SPIE 3772, 304 (1999)
SPIE 5535, 232 (2004)

Elemental correlations

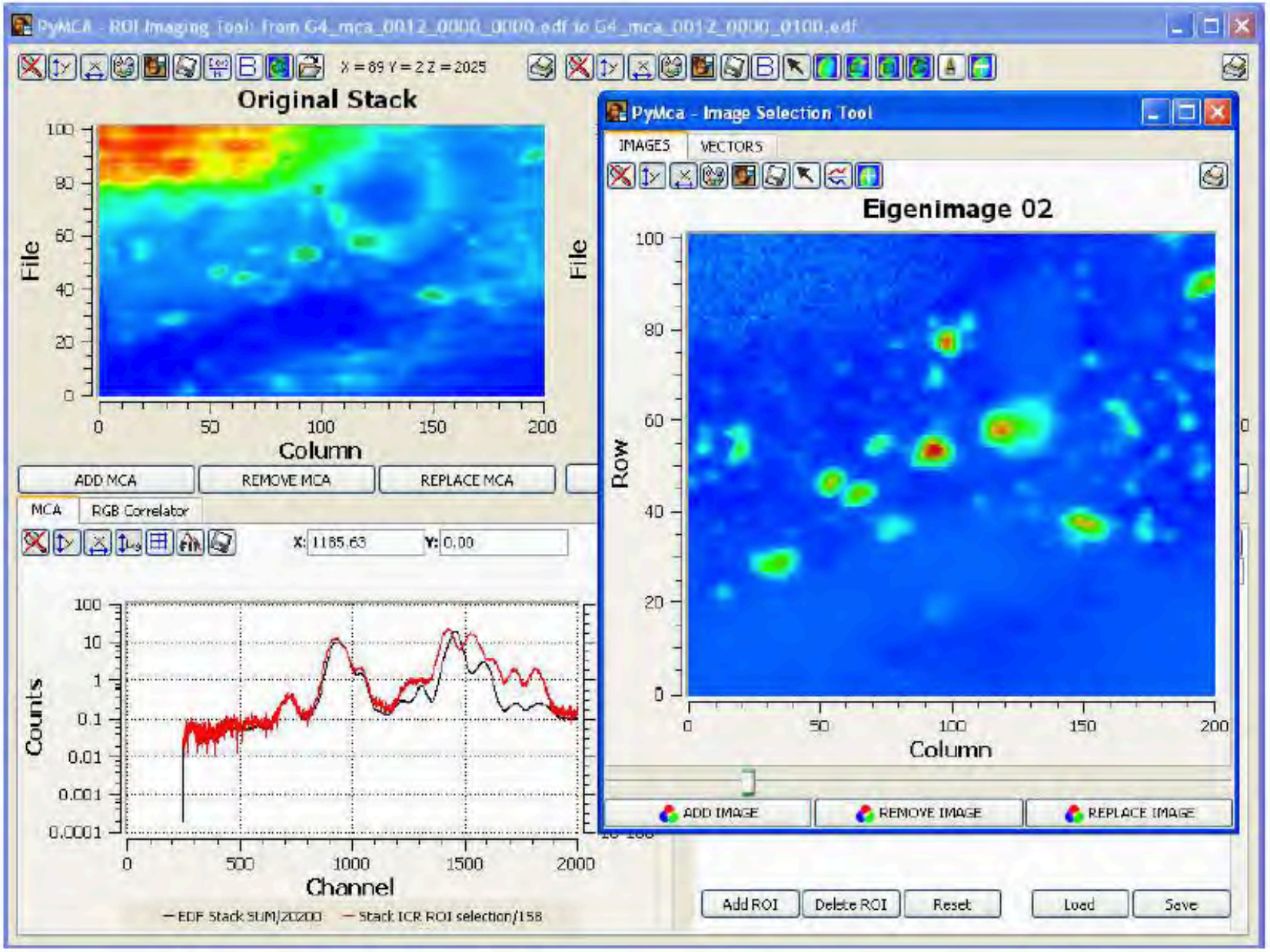




Correlated S & Sb

Uncorrelated Pb & Sb

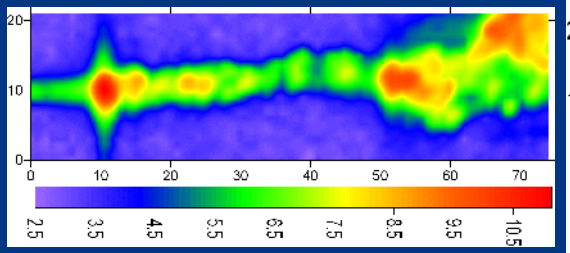
Multivariate analysis: PCA

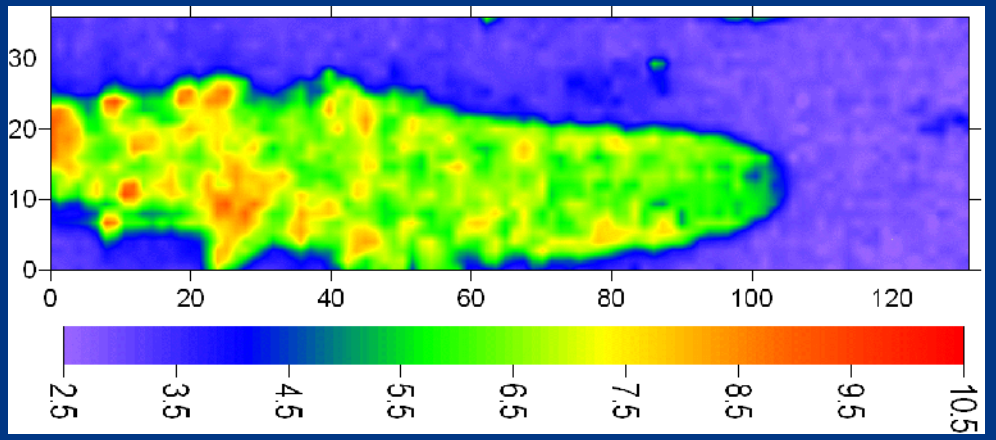


NASA Stardust cometary keystones

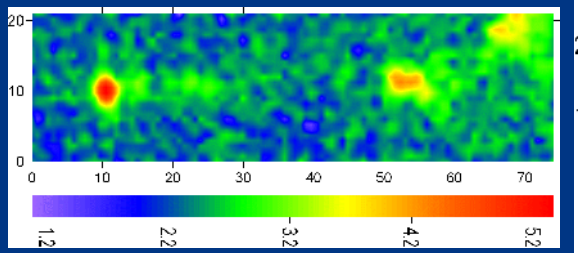
Fragmentation = f (Z, ρ_Z, ρ_{aer})

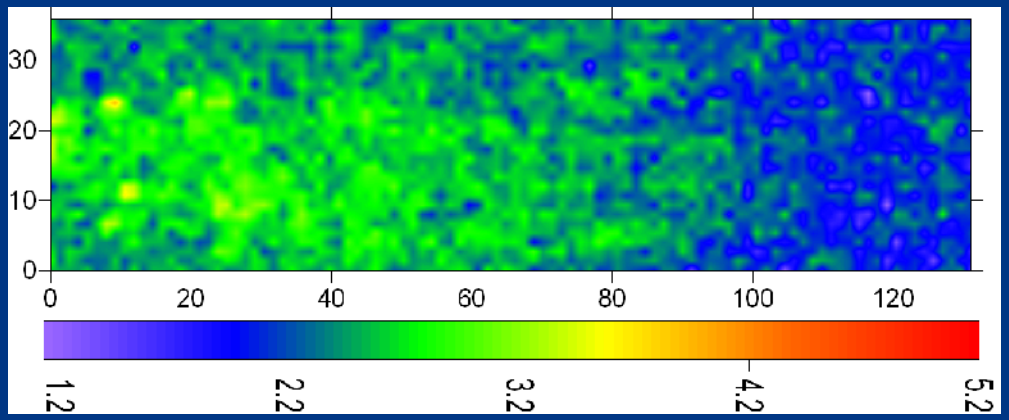
Fe



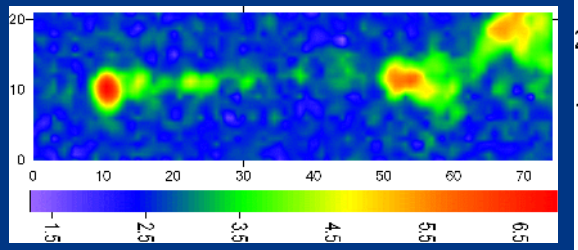


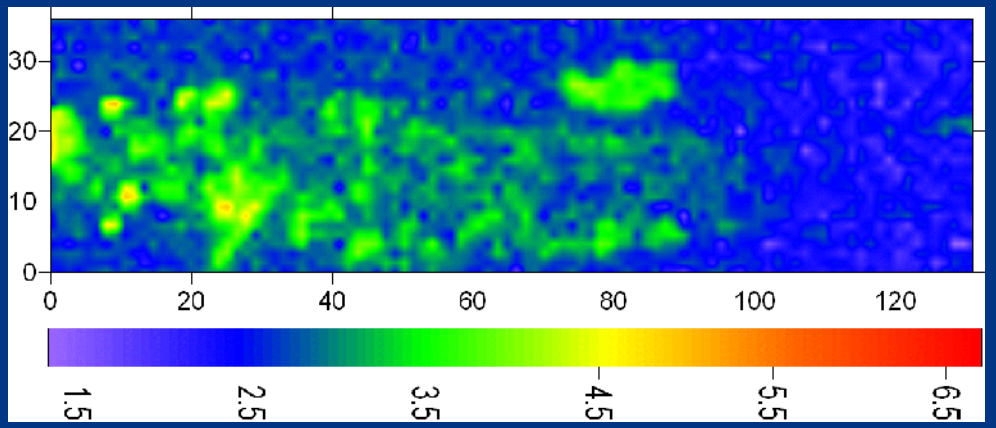
S



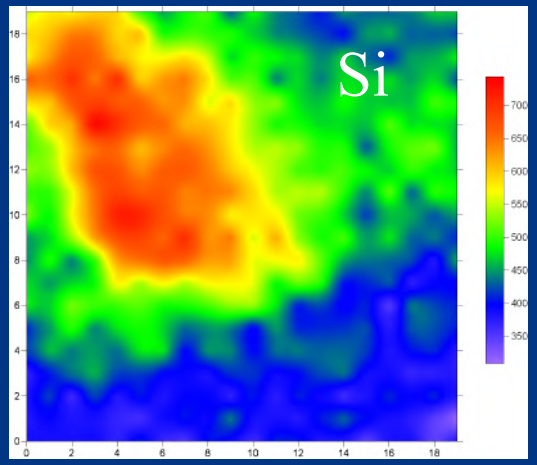


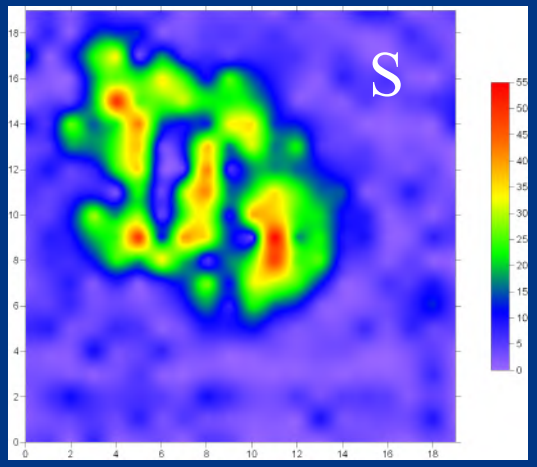
Cu





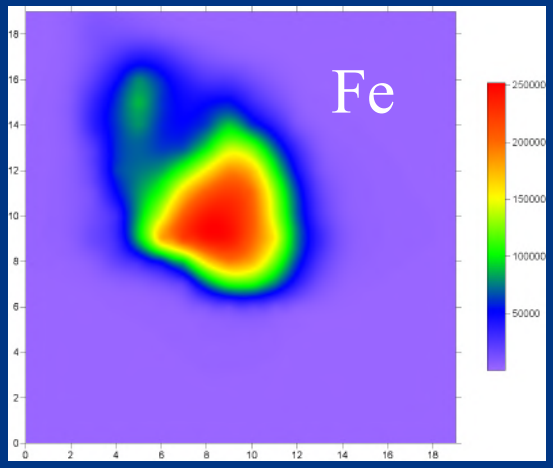
Heterogeneity of Terminal Particle

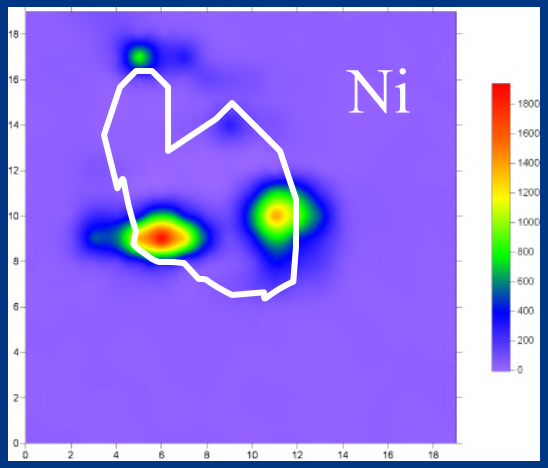


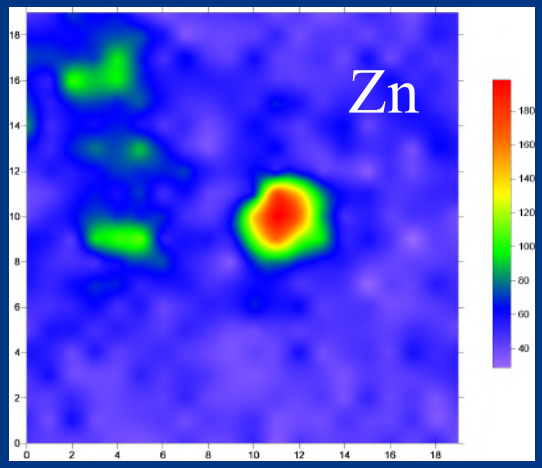


Resolution

$1 \times 1\ \mu m^2$



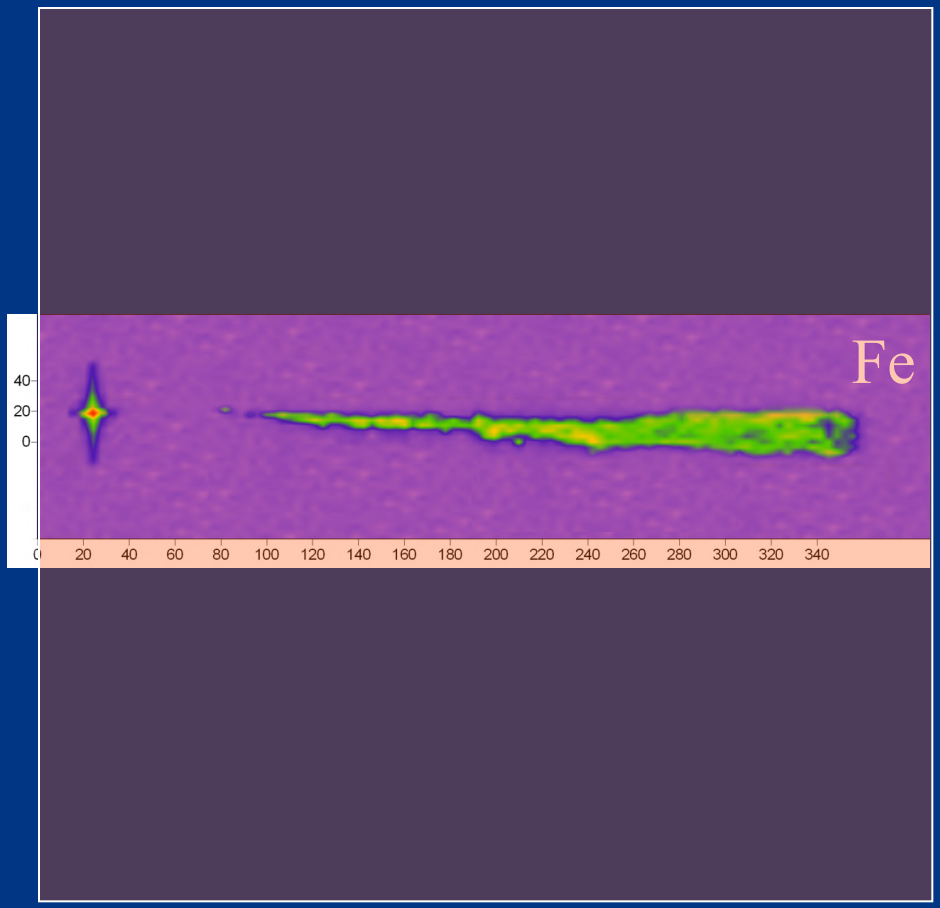


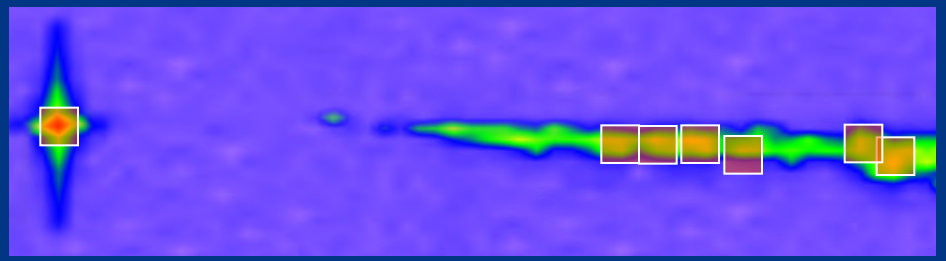


Fe/Ni ≈ 21.5 !!!

Fe/Ni ≈ 2 - 200

Spatial sampling: resolution effect





Low res single point

1 pix = L_{sample}

Fast but large error

Hotspot mapping

Few pixels:

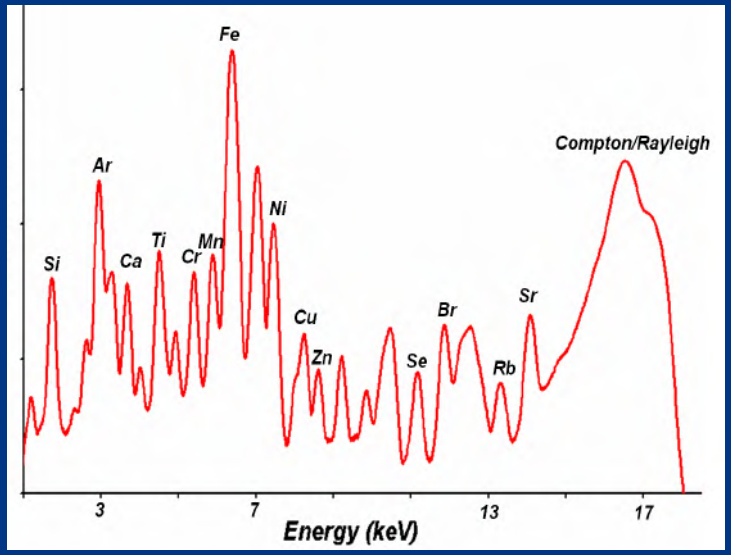
- miss signal

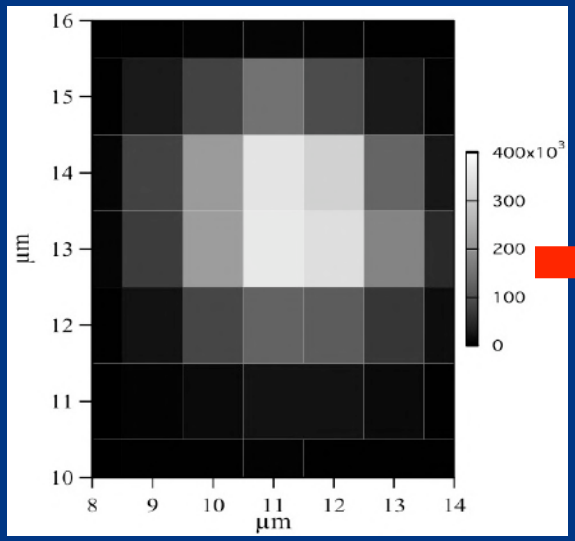
Fe - thresholding element

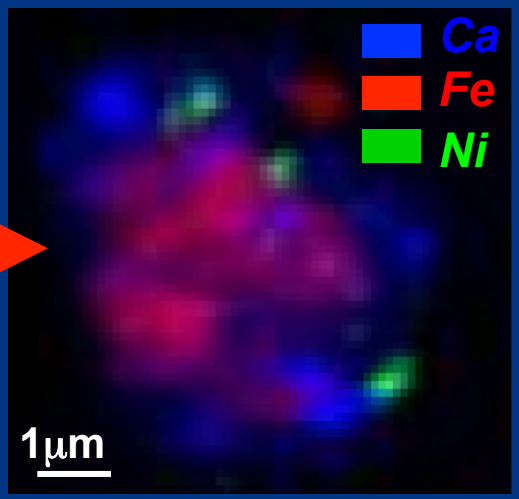
Spatial sampling: resolution effect

Resolution

- ✓ beamspot mm/μm/nm
- ✓ scanning step - oversampling ≤ 30%
- ✓ mis-interpretations/mis-quantifications







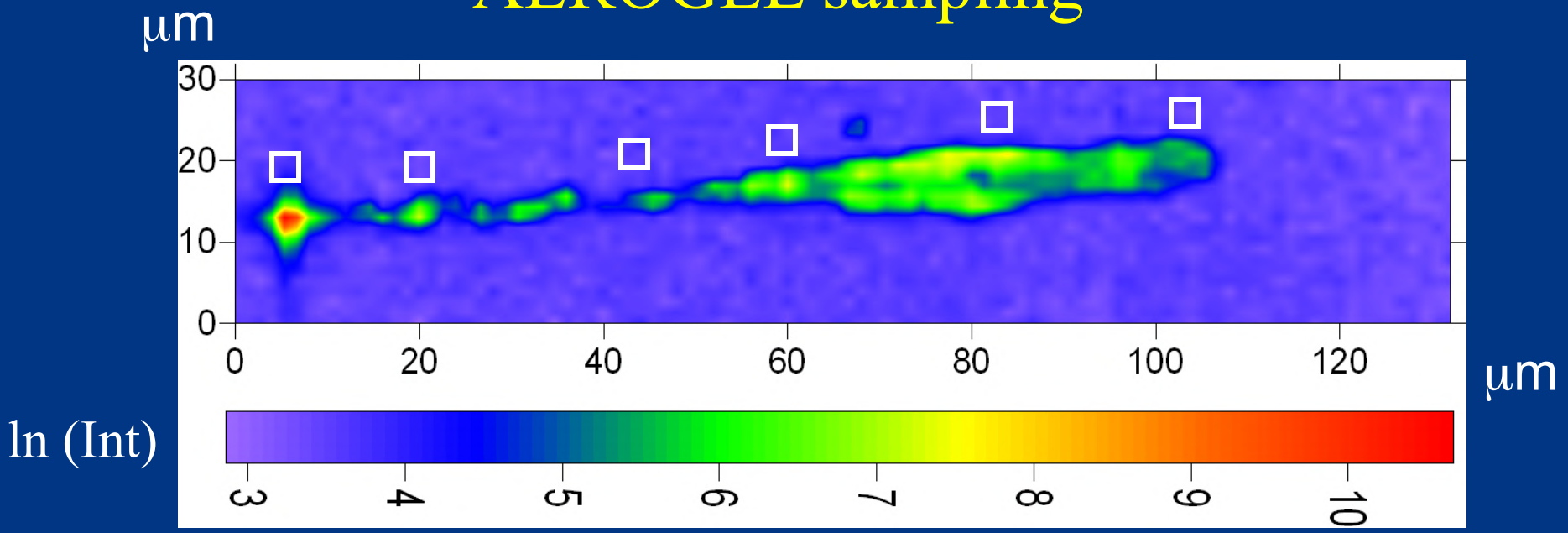
1 x 1 μm²

200 x 150 nm²

Bleuet, Simionovici et al., App. Phys. Lett. **92**, 213111-1–3, 2008

Aerogel composition

AEROGEL sampling



aerogel

"dirty", heterogeneous, insulating medium

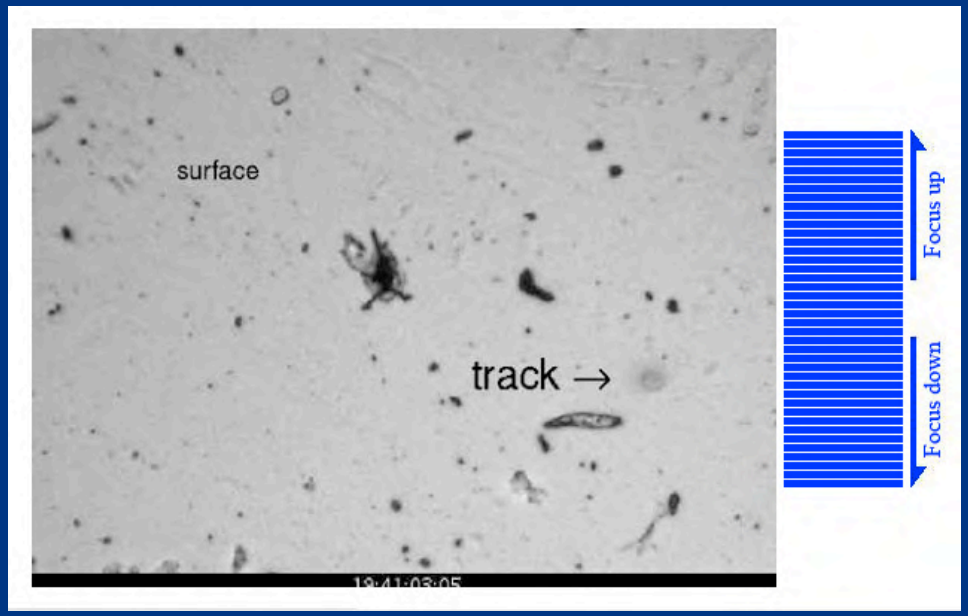
Aerogel sampling

- correlated by Si fluorescence
- longitudinal/radial density profiles - **gradient** (30 %)
- **no** density variation for track vs aerogel

ISPE: Preliminary Analysis of Interstellar Grains

STARDUST @ HOME

- search for **Interstellar** grains
- nano-imaging (XRF, XRD, XCT)
+ XAS ?



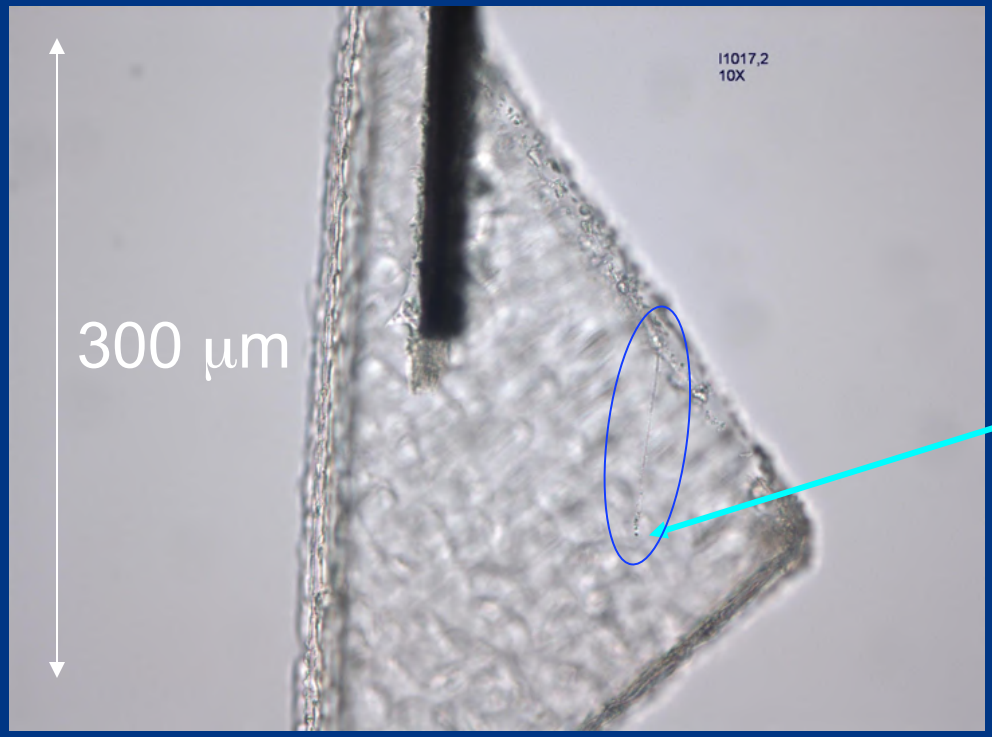
- **search grains < 1 μm**
- **113000 "hunters"**
- **redundant reliability control**

virtual microscope

ISPE Treasure Hunt in process

orders of magnitude more challenging than the cometary samples.
1000 times smaller in mass, 1% of the statistics

- **"Look, but don't touch" NEW policy?**
- **intact trajectory ?**
- IS/IP impactors
- total mass distribution
- xtal ? ExTerr bio-organisms ???

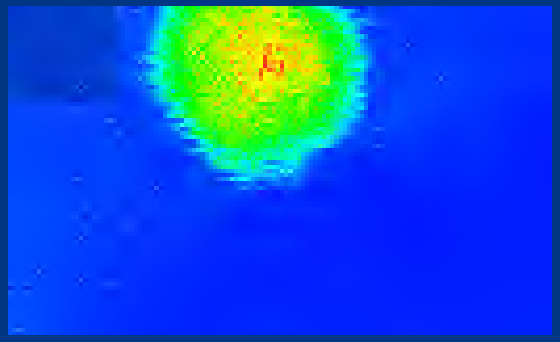


2 μm diameter track
Identified by >300
Stardust@home
"dusters"

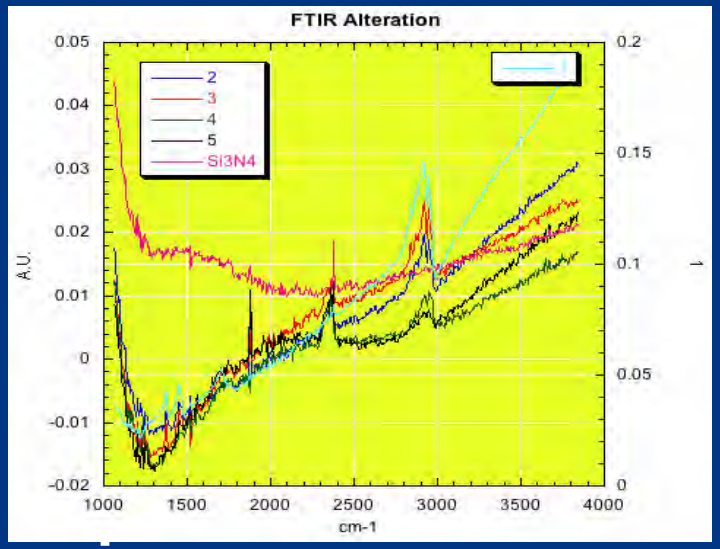
ISPE: Beginning of a long story

Work on FISC

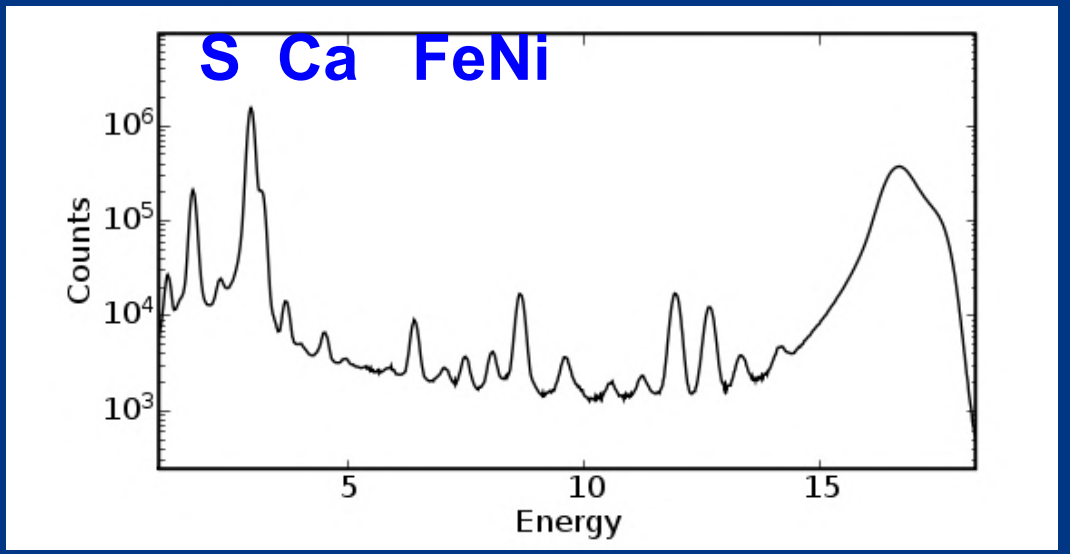
Non – (little) destructive (CH_2/CH_3 contamination check by SR-FTIR)



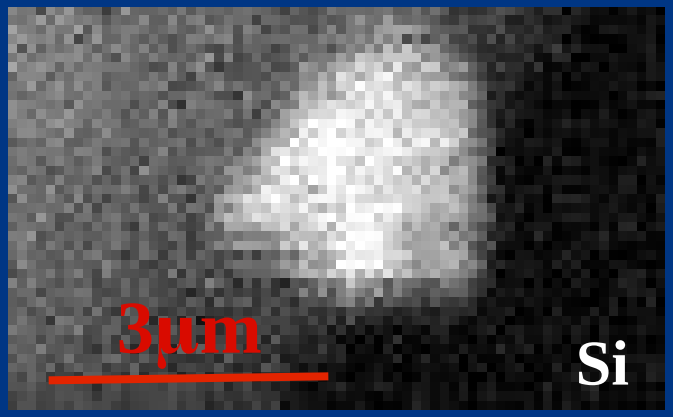
1 µm

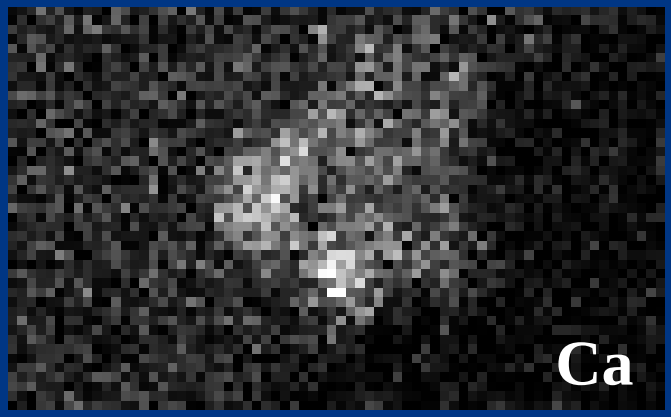


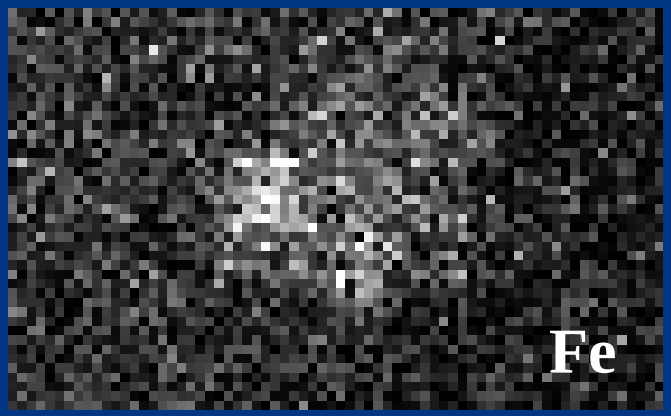
NO NEW ELEMENTS !!!

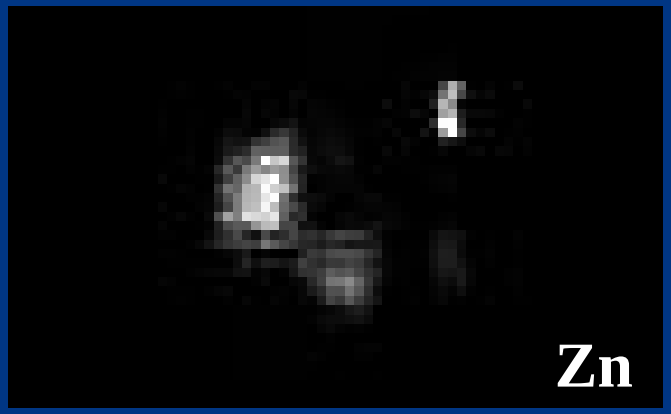


1027,1,9 (First interstellar candidate)









Si/Fe = 3661 ± 239

Ca/Fe = 8.6 ± 1.0

Zn/Fe = 6.5 ± 0.4

Synchrotron: Wish list

To do

- ✓ beam stability ≤ 1/4 beamspot = f(*t*, *E*)
- ✓ XRF, XCT, XRD = OK but no n-Xanes yet
- ✓ larger E range (7 ≤ E ≤ 52 keV)
- ✓ beamspot ≤ 50 nm

To look into

- ✓ beam damage assessment (FTIR, AFM, repeats)
- ✓ mapping point & click contours
- ✓ n-goniometers (sphere of confusion = beamspot)
- ✓ hyperspectral treatment: XRF+XRD+Raman/IR (PyMCA)