IAEA-CN301-195

Investigating Radiation Effects in Materials using State-of-the-Art Particle Accelerators

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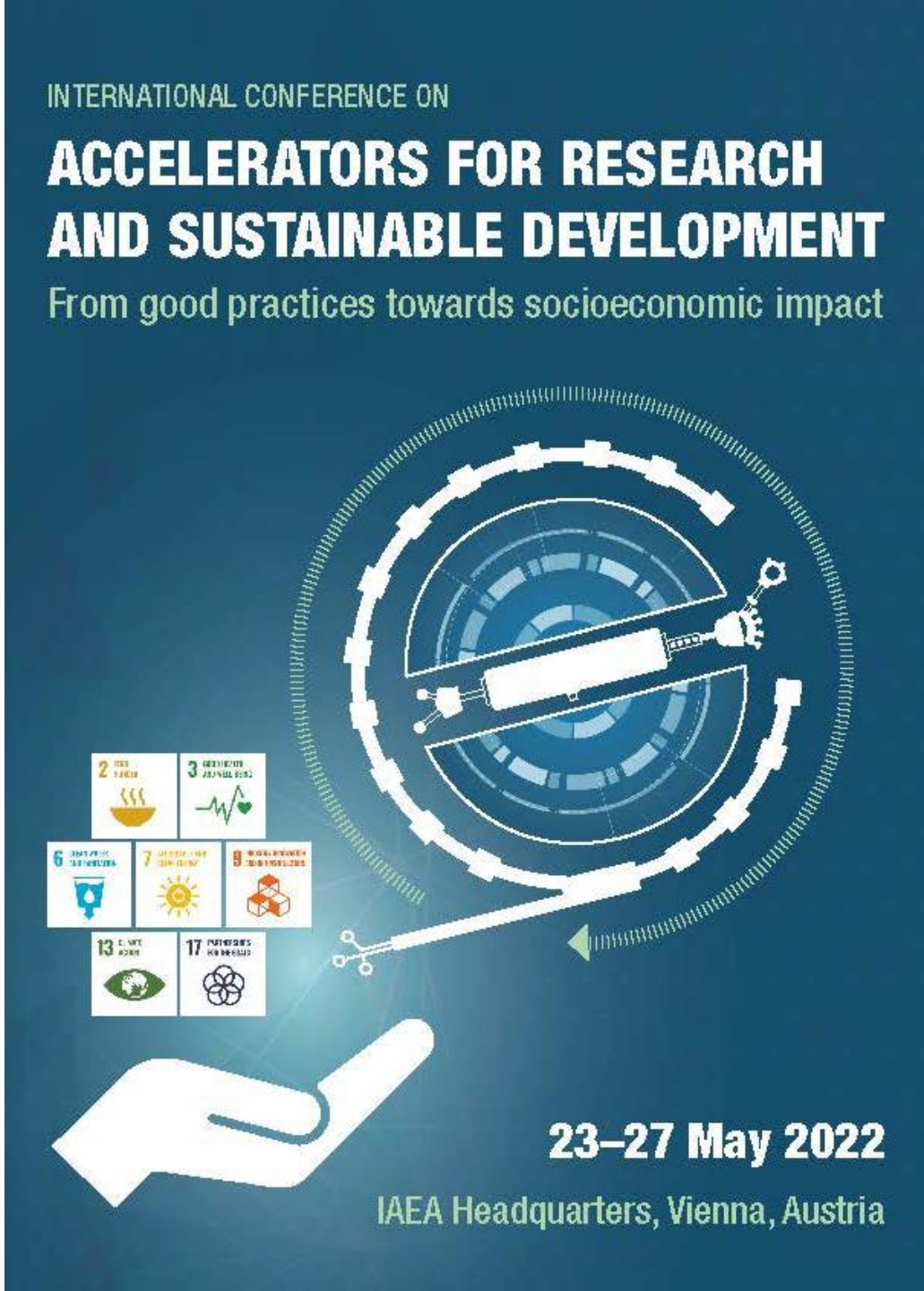


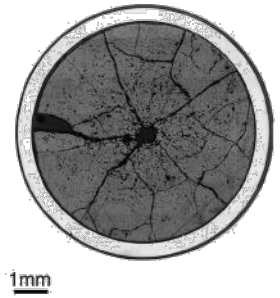
Motivation: Nuclear Materials under Extreme Conditions

https://www.iaea.org/newscenter/news/improving-safety-of-ageing-nuclear-power-plants-in-lockdown

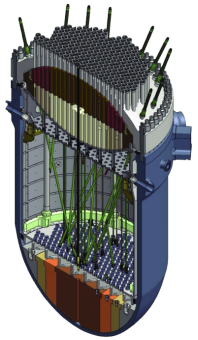
https://www.ans.org/news/article-367/hydraulic-testing-completed-on-akkuyu1-reactor-vessel/

https://cen.acs.org/energy/nuclear-power/Radioactive-waste-stranded-US-shifts/96/web/2018/08

NUCLEAR FUEL



STRUCTURAL MATERIALS

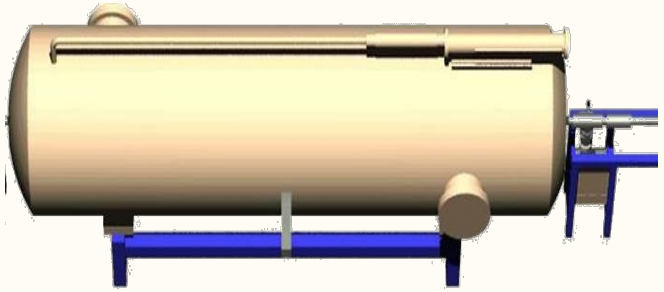


WASTE FORMS

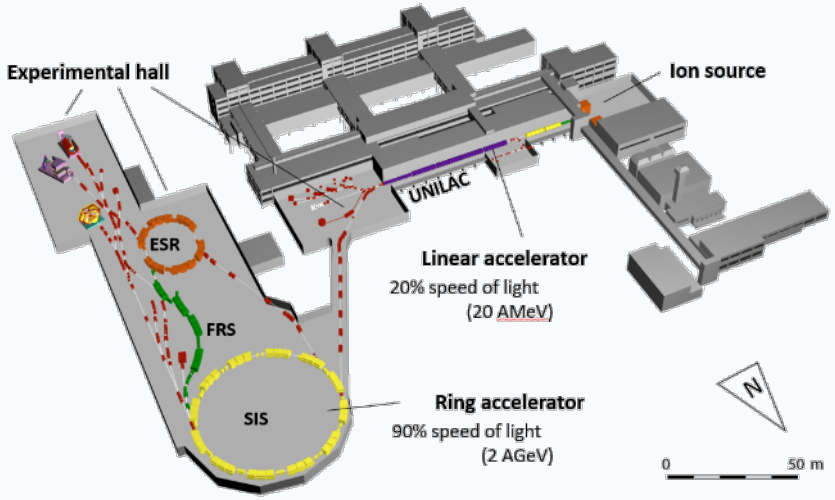
Harsh Environments: intense **irradiation**, high-temperatures with steep gradients, stress, evolving microstructure and chemical composition, etc.

Ion-Beam Experiments to Study Irradiation Effects in Materials

Tandem Accelerator (E < 25 MeV)
available in many laboratories

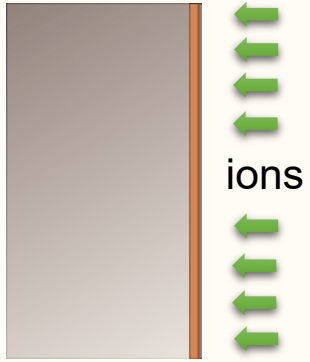


Large Ion Accelerator Facilities (E > 100 MeV)
worldwide limited availability



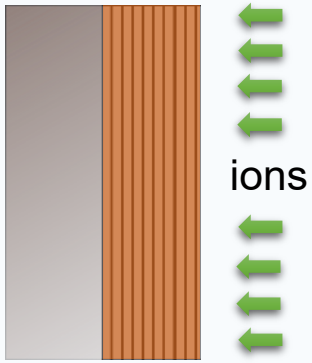
GSI, Darmstadt (Germany)

irradiated layer:
100 nm – 1 µm

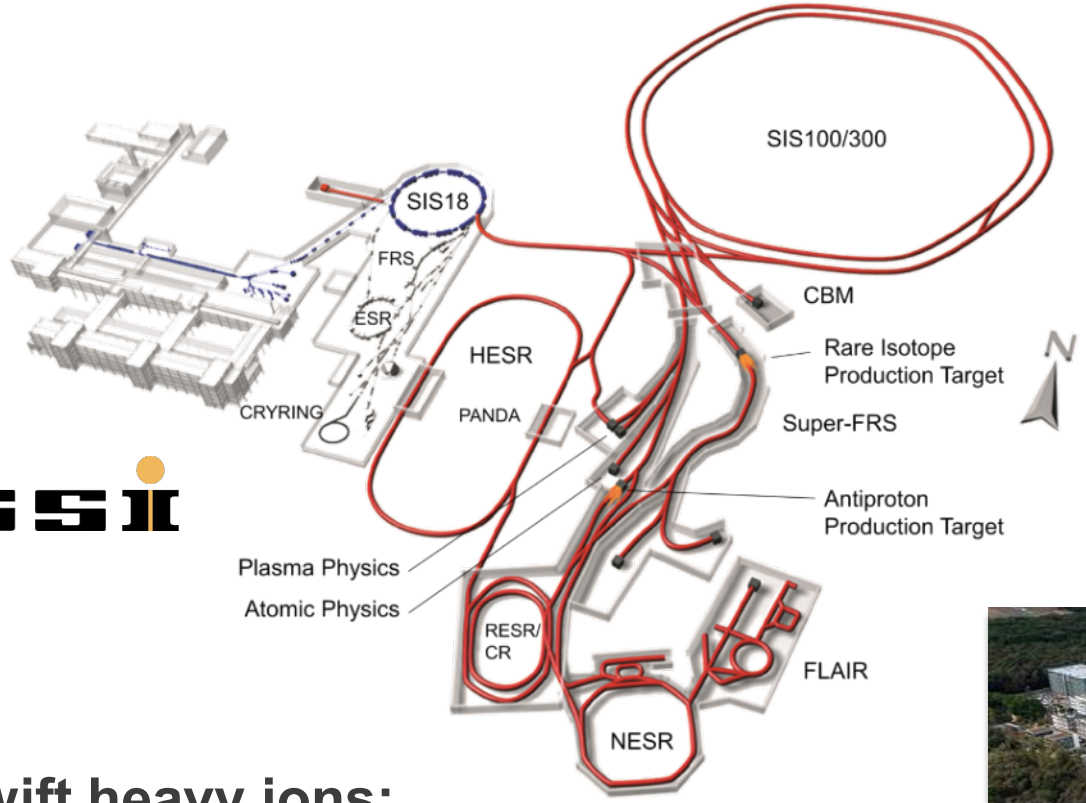


Alpha-recoil damage		Fission-track damage
nuclear	energy loss, d*E*/d*x*	electronic
small	irradiated volume	large
difficult	bulk characterization	easy
high	fluence/dose	medium

irradiated layer:
10 µm – 100 µm



Helmholtz Center for Heavy Ion Research – Darmstadt, Germany

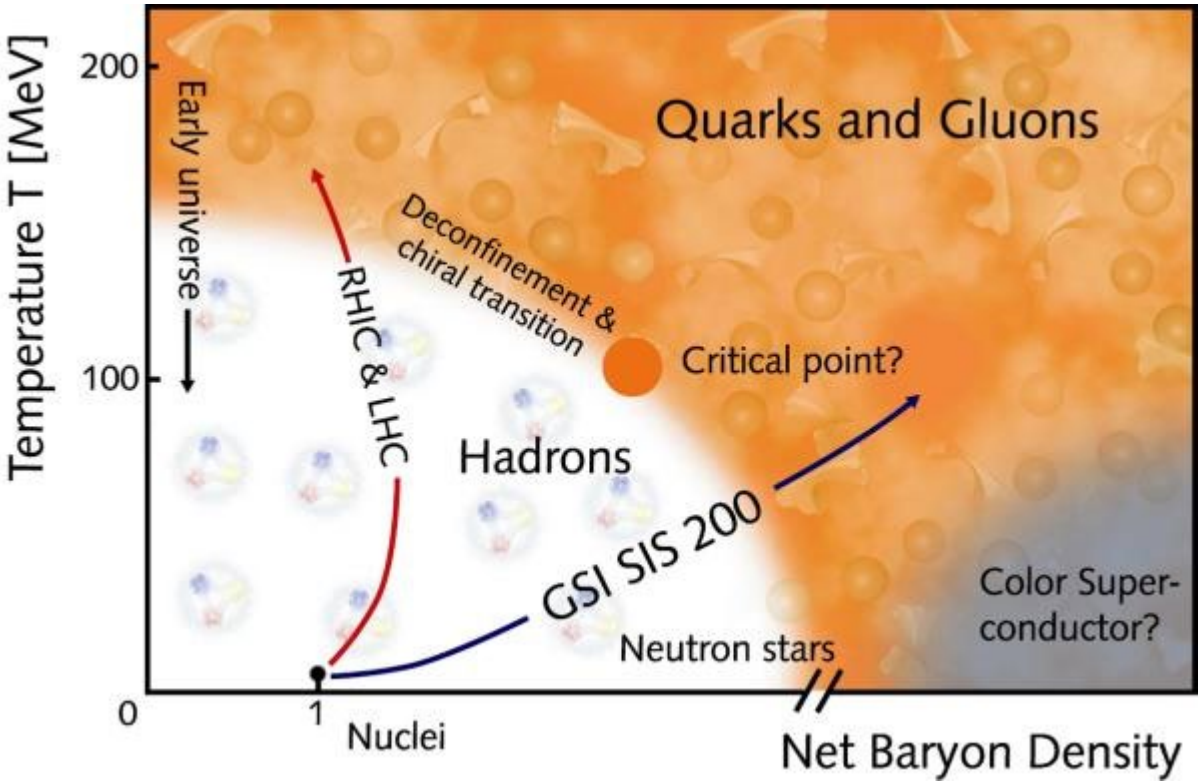


Research with swift heavy ions:

- World's largest heavy ion accelerator facility
- Nuclear physics, atomic physics, Biophysics, plasma physics, materials research

High-Energy Physics with Swift Heavy Ions: Early Stage of the Universe

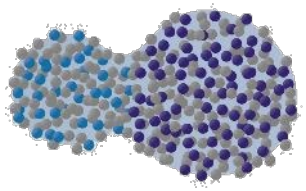
http://www.physics.purdue.edu/henp/



Quark-Gluon Plasma

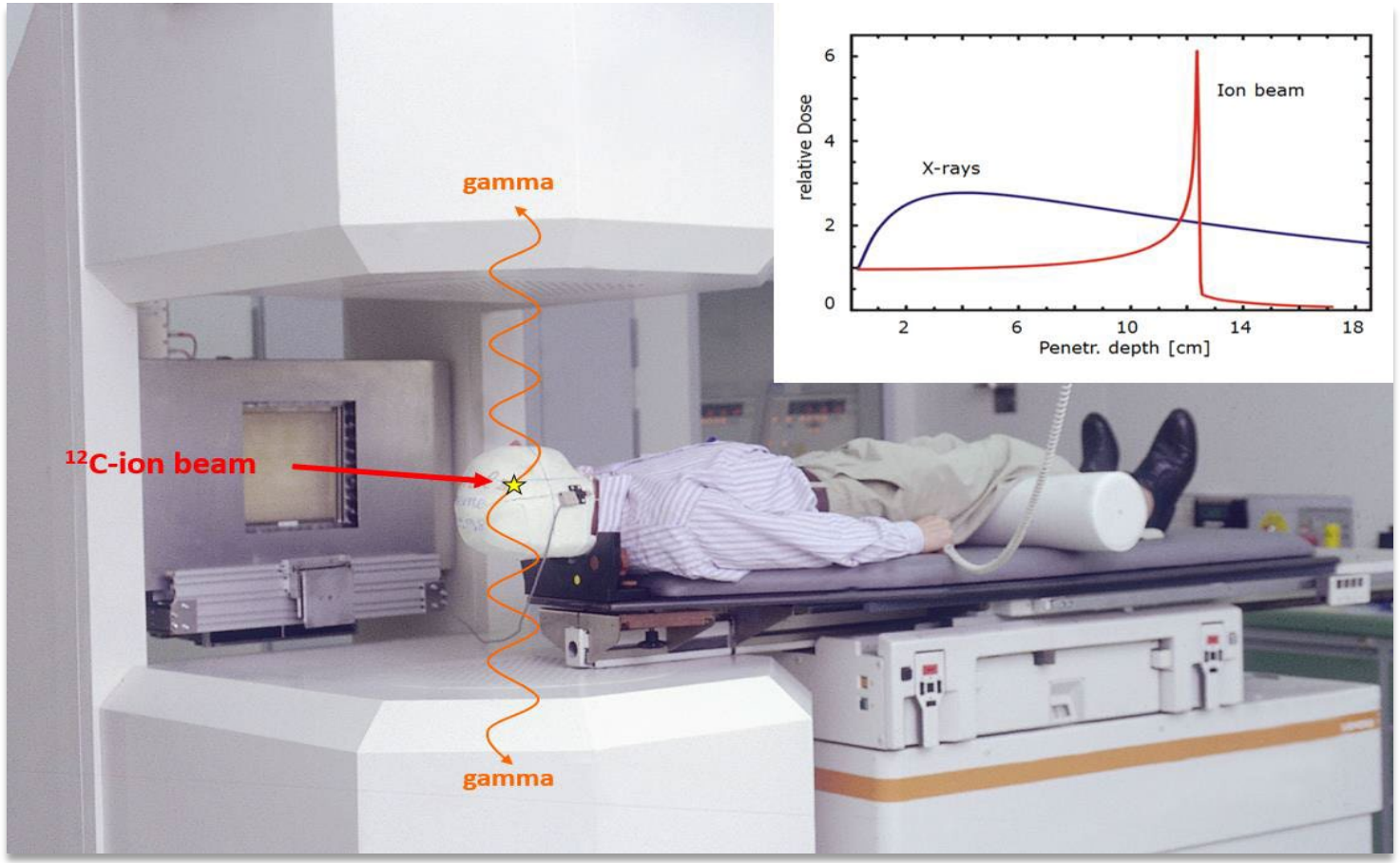
At extremely high densities and temperatures, protons and neutrons 'melt' in their constituents

FOPI-Detector at GSI



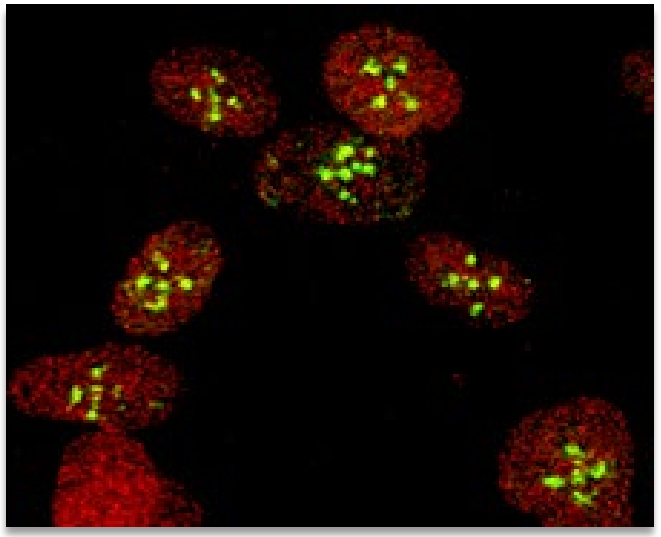
Detecting all particles produced during ion collisions

Swift Heavy Ions in Biophysics: Cancer Therapy



Heidelberg Ion-Beam Therapy Center:

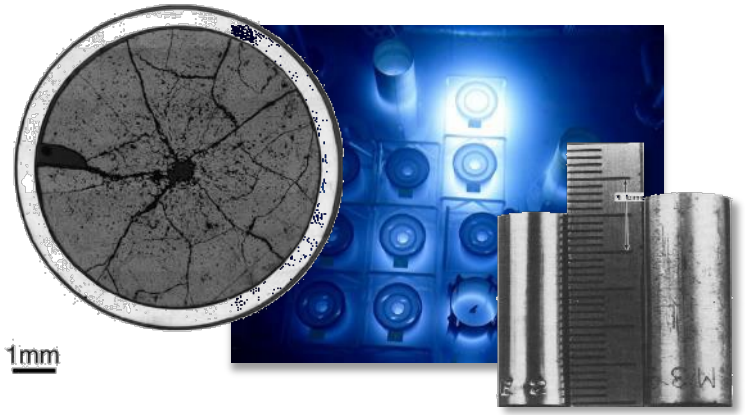
Tumor treatment with energetic carbon ions



Microprobe at GSI:

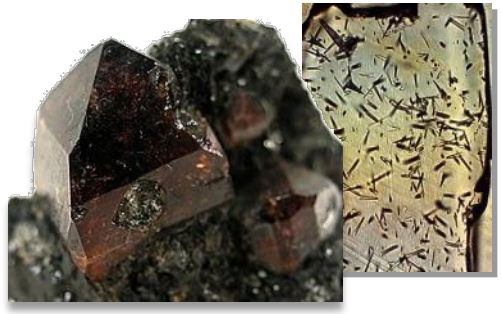
Targeted irradiation of nuclei of living cells

Application of Swift Heavy Ions in Materials Research



NUCLEAR ENGINEERING

Radiation damage

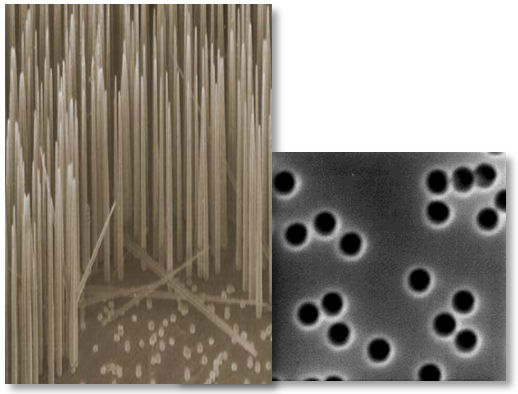


GEOSCIENCE

Fission-track dating

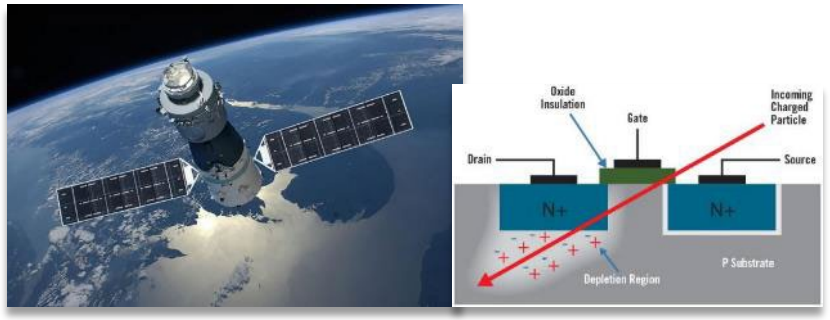
ACCELERATOR TECHNOLOGY

Degradation of components



ION-BEAM APPLICATIONS

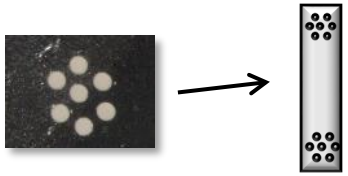
Nano-structuring



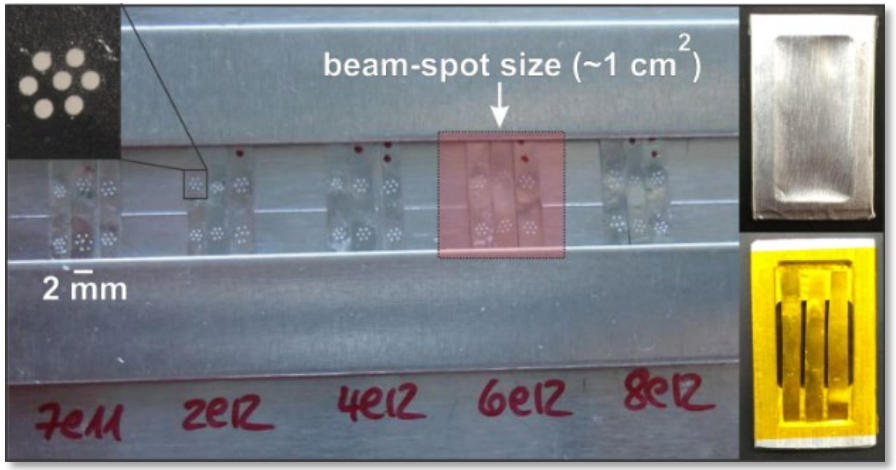
SPACE TECHNOLOGY

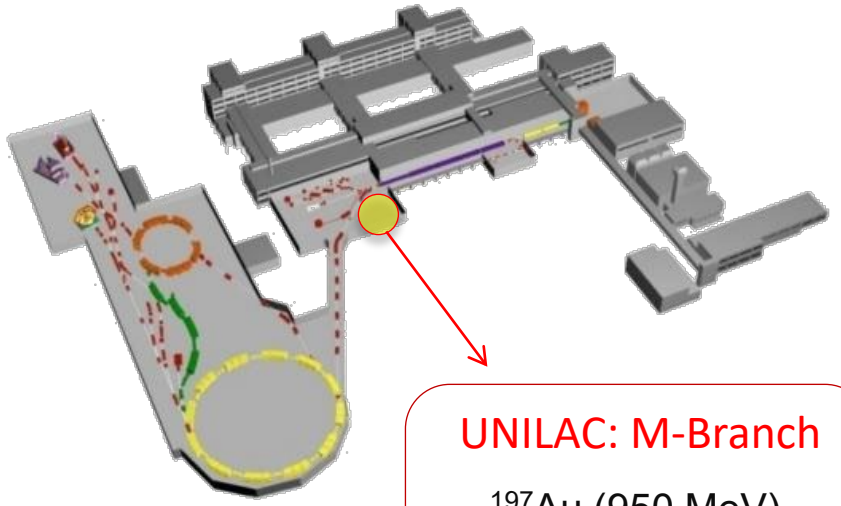
Single-event upsets

Irradiation Experiments for Synchrotron X-Ray Characterization

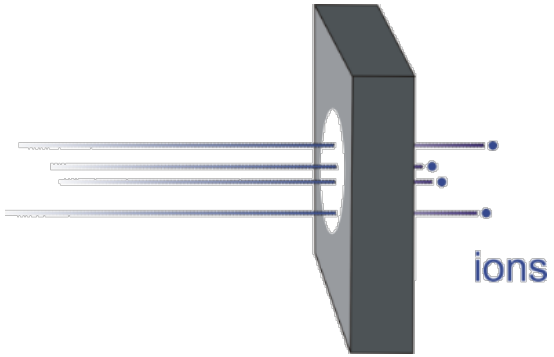


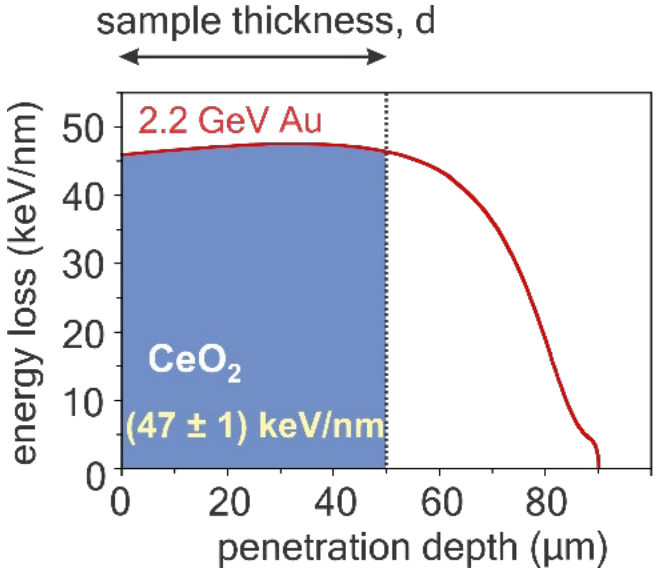
Sample chamber
diameter: 100 µm
thickness: 50 µm
thickness: 12.5 µm





UNILAC: M-Branch
^{197}Au (950 MeV)
10^{11} – 10^{14} ions/cm^2





M. Lang, *et al.*, *Journal of Materials Research* (2015).

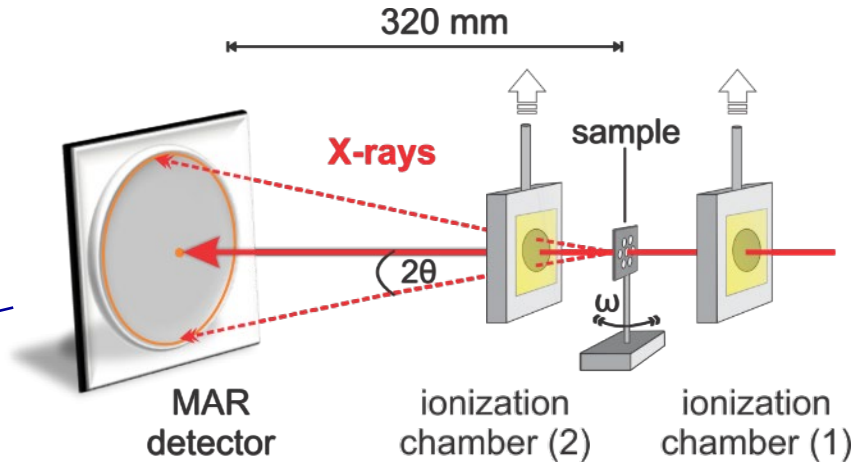
Small Volume Irradiations

- ⇨ High energy and ion range
- ⇨ Small beam spot
- ⇨ Fluence series

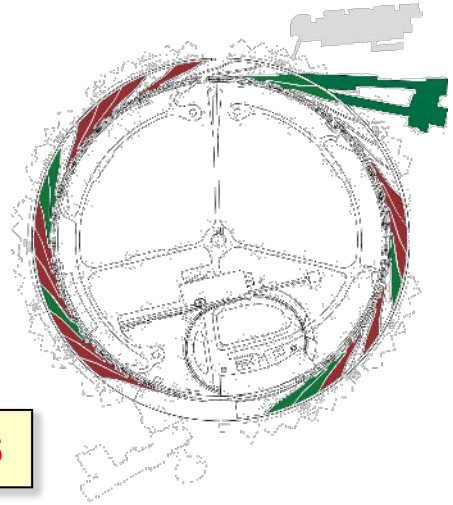
Irradiation Experiments for Synchrotron X-Ray Characterization

Advanced Photon Source

- ⇨ synchrotron X-rays (25 keV)
- ⇨ focused, intense beam

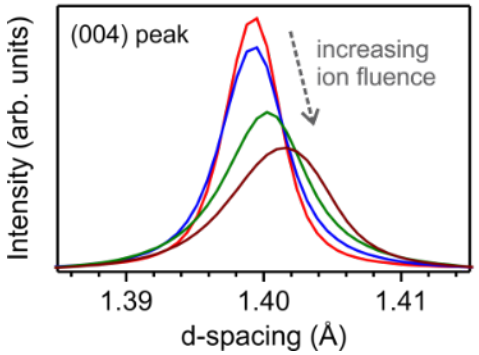


M. Lang, *et al.*, *Journal of Materials Research* (2015).



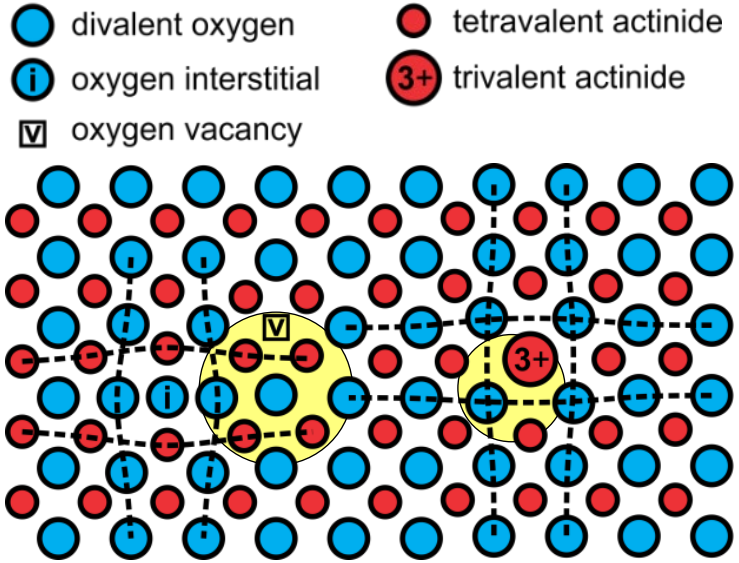
XRD

X-ray diffraction



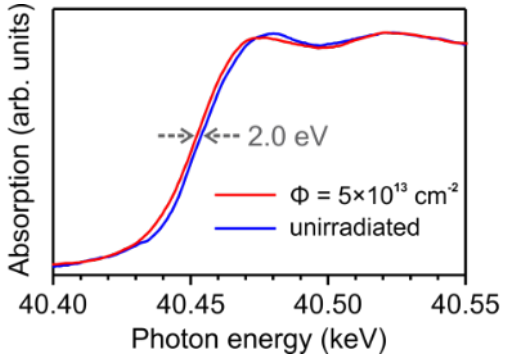
Structural properties

- ⇨ unit-cell parameter
- ⇨ microstrain



XAS

X-ray absorption spectroscopy

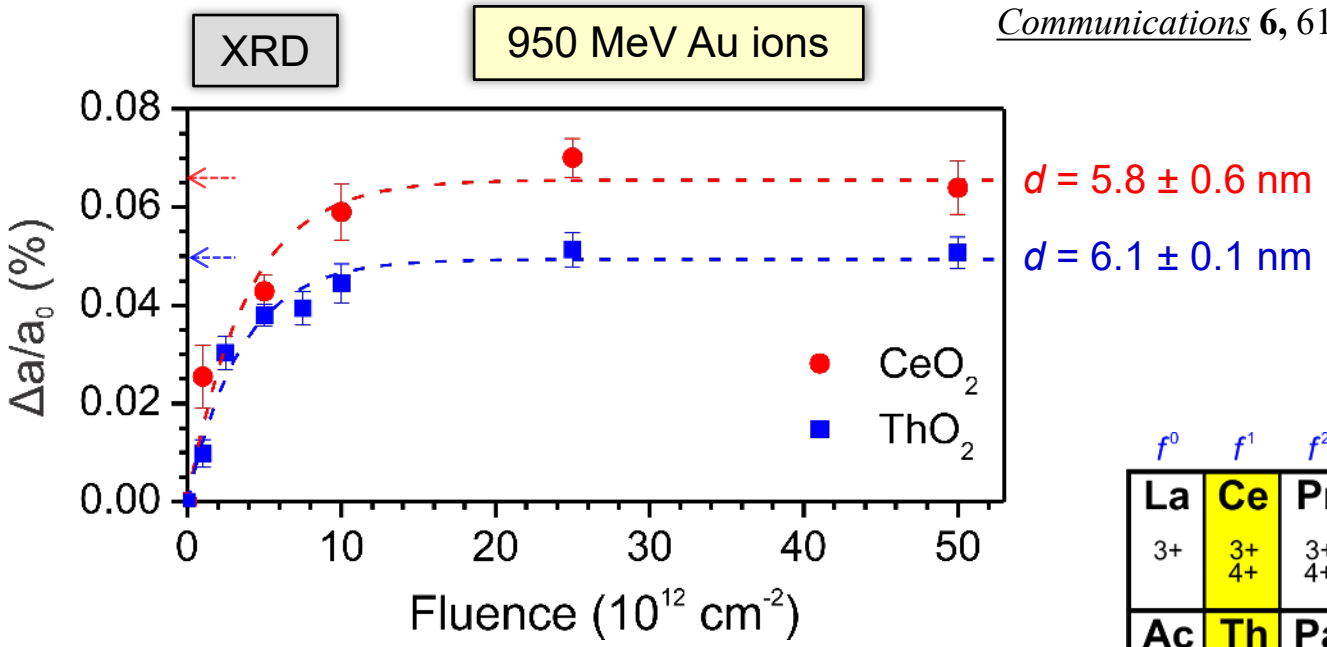


Electronic properties

- ⇨ oxidation state
- ⇨ local bonding

Example: Radiation Effects in Nuclear Fuel Type Materials: AO_2

C.L. Tracy, *et al.*, *Nature Communications* **6,** 6133 (2015).



$$\frac{\Delta a}{a_0} = \frac{a_{sat} - a_0}{a_0}\left(1 - e^{-\sigma\phi}\right)$$

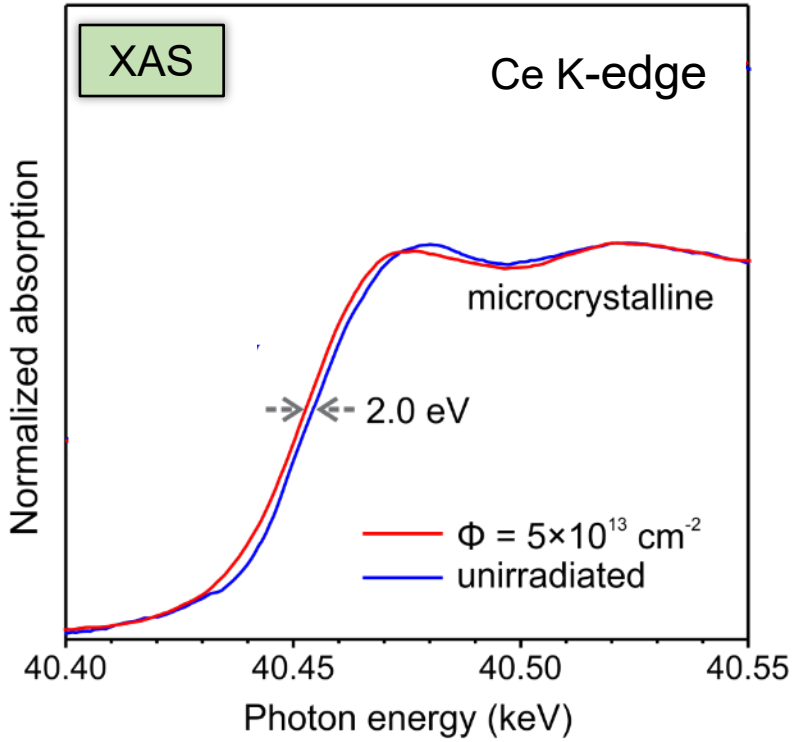
f^0	f^1	f^2	f^3
La 3+	Ce 3+ 4+	Pr 3+ 4+	Nd 3+
Ac 3+	Th 4+	Pa 4+ 5+	U 4+ 5+ 6+

H. Ohno, *et al.*, NIMB **266,** 3013 (2008).

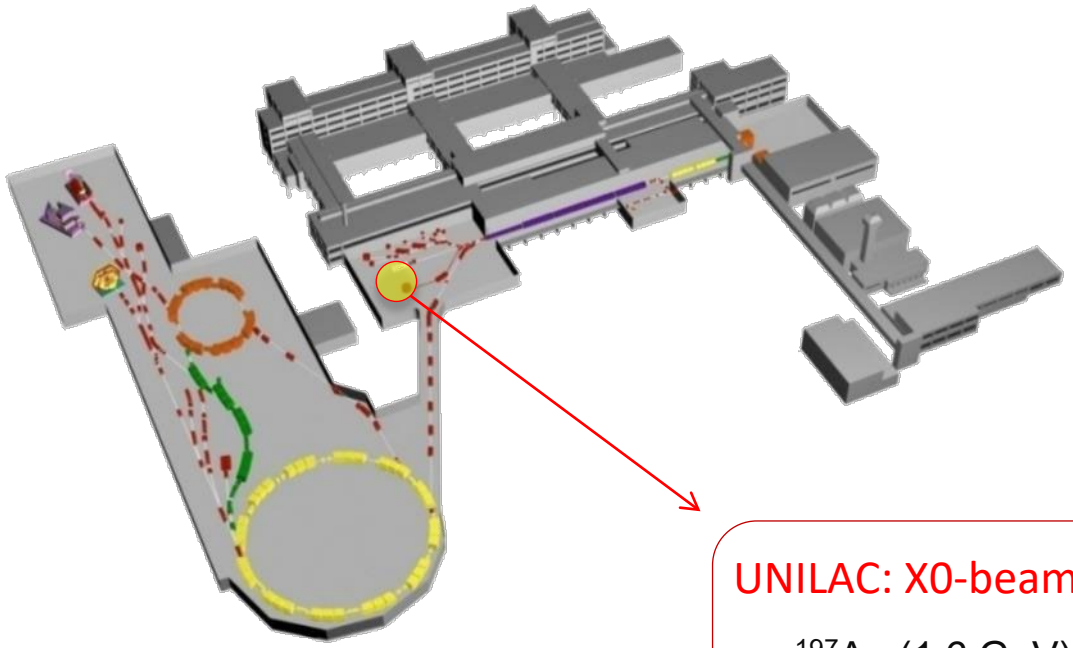
A. Iwase, *et al.*, NIMB **267,** 969 (2009).

Redox response of actinide materials to ion irradiation

⇨ partial reduction of Ce^{4+} to Ce^{3+} determines radiation response



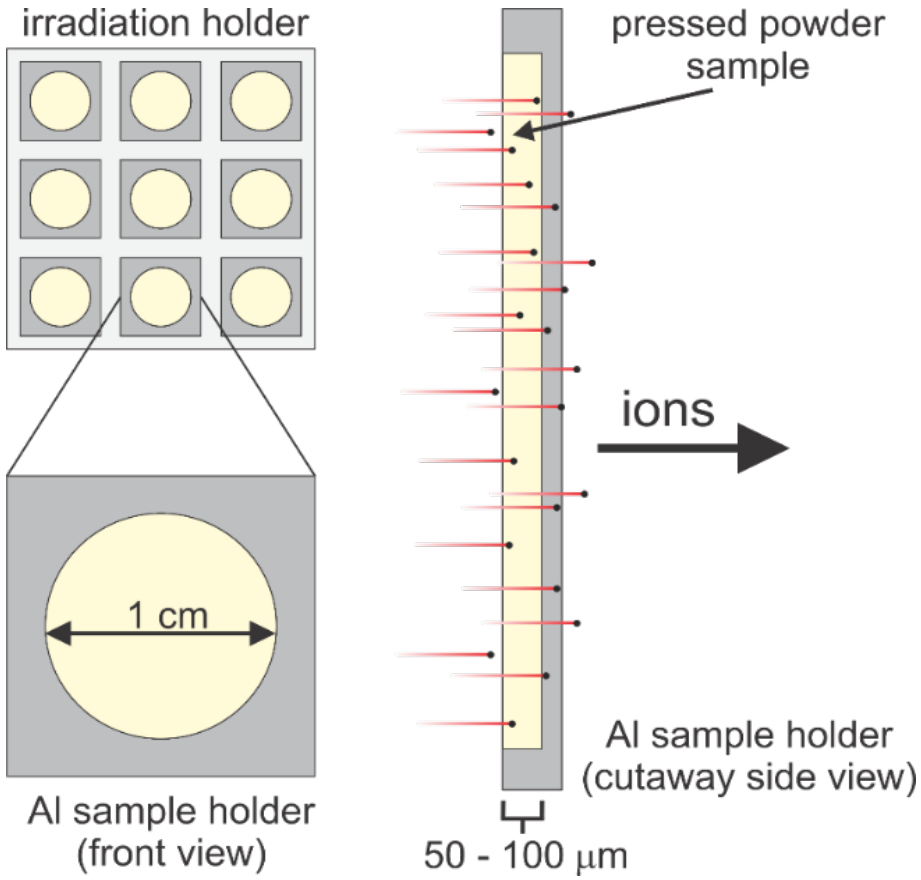
Irradiation Experiments for Spallation Neutron Characterization

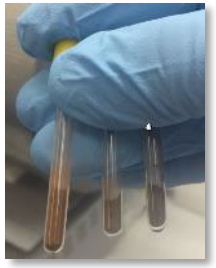


UNILAC: X0-beamline

^{197}Au (1.6 GeV)

10^{13} ions/cm^2





~100 mg

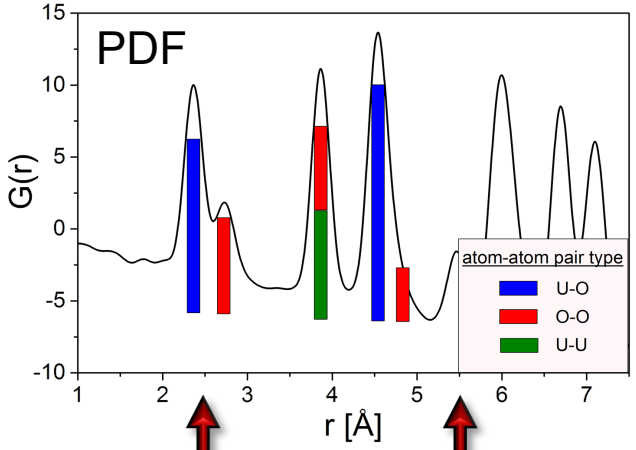
Large Volume Irradiations

- ⇨ High energy and ion range
- ⇨ Large beam spot
- ⇨ High fluences

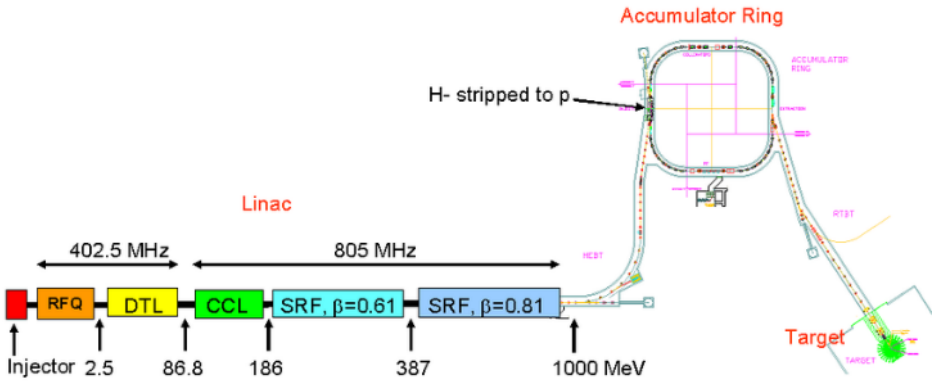
Irradiation Experiments for Spallation Neutron Characterization

The Nanoscale-Ordered Materials Diffractometer (NOMAD)

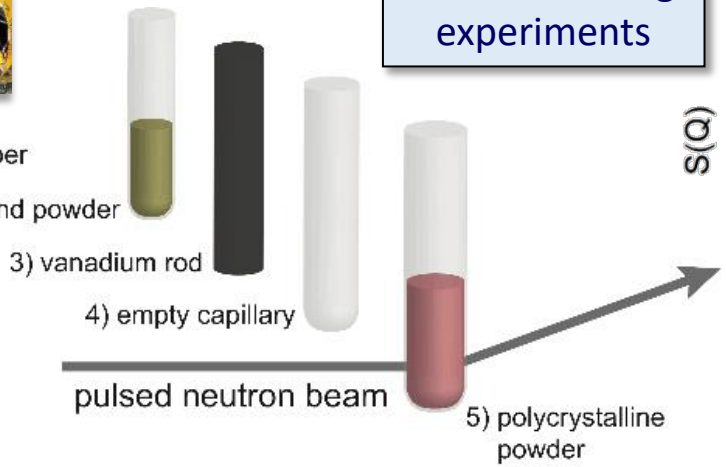
- ⇨ neutron wavelength: 0.1 – 3 Å
- ⇨ flux on sample: 10^8 $cm^{-2}\cdot sec^{-1}$
- ⇨ large detector coverage
- ⇨ extending to very high Q-values
- ⇨ high-res pair distribution function (PDF)
- ⇨ sample mass: **100 mg**

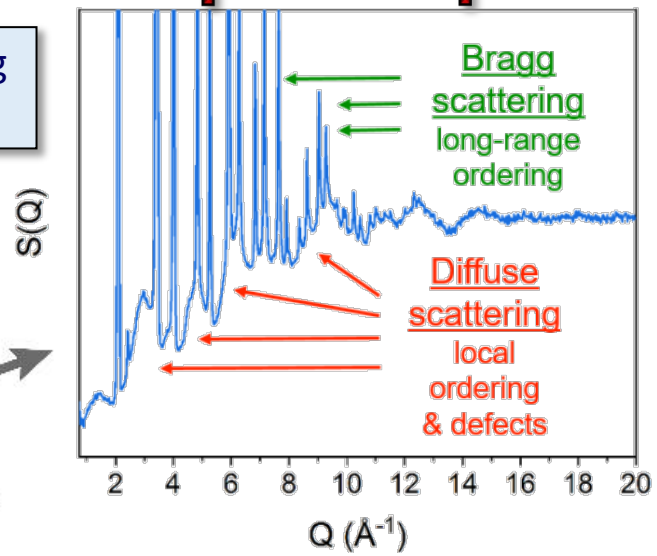


$$G(r) = \frac{2}{\pi}\int_{Q_{min}}^{Q_{max}} Q[S(Q)-1]\sin(Qr)dQ$$



Total scattering experiments

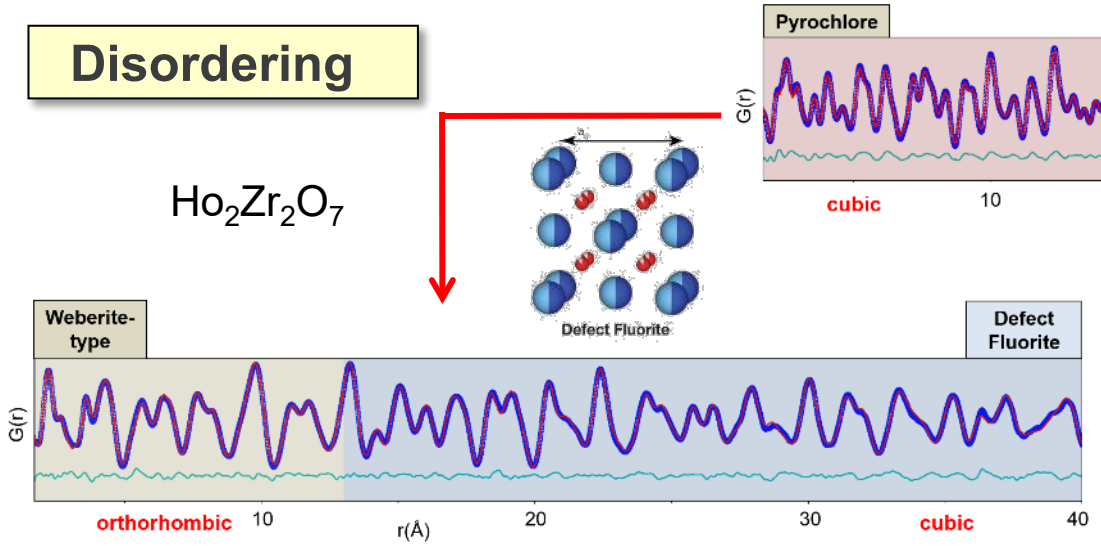




Example: Radiation Effects in Nuclear Waste Form Materials: $A_2B_2O_7$

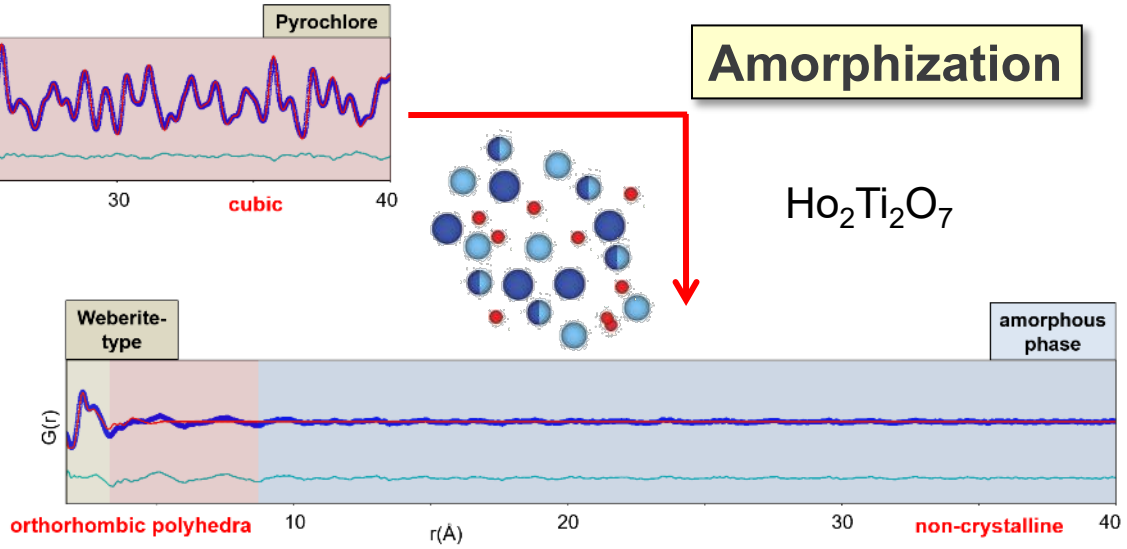
Disordering

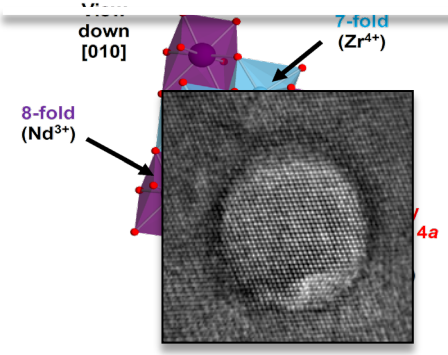
$Ho_2Zr_2O_7$

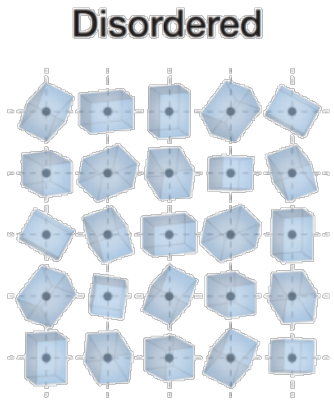


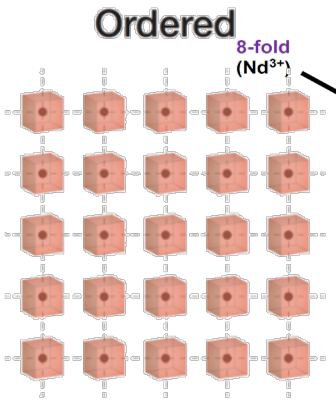
Amorphization

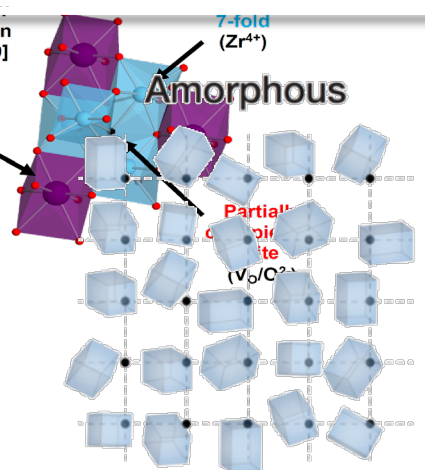
$Ho_2Ti_2O_7$

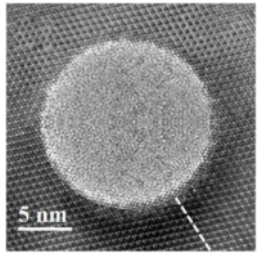












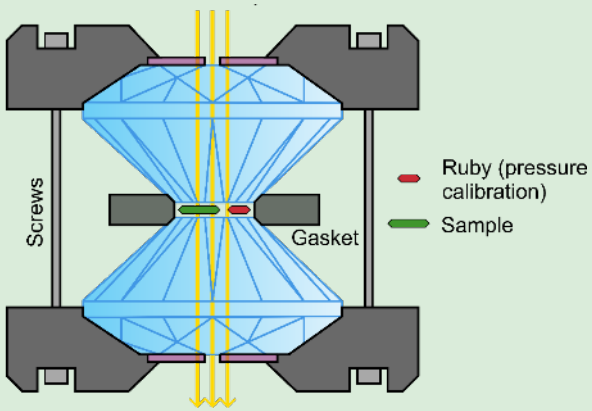
Resistant Pyrochlores
- short-range: weberite
- long-range: defect fluorite

J. Shamblin, *et al.*, *Nature Materials*, 15, 507-511 (2016).

E.C. O'Quinn, *et al.*, *Science Advances*, 6, eabc2758 (2020).

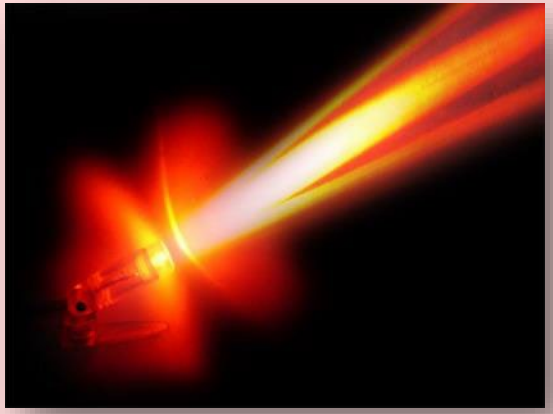
Susceptible Pyrochlores
- short-range: weberite
- long-range: amorphous

Coupling Pressure Cells with Swift Heavy Ion Irradiation: Multiple Extremes

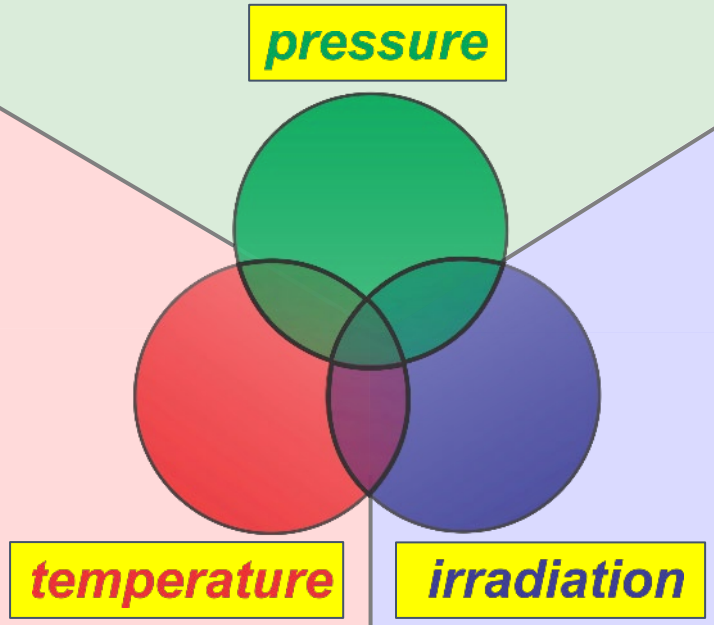


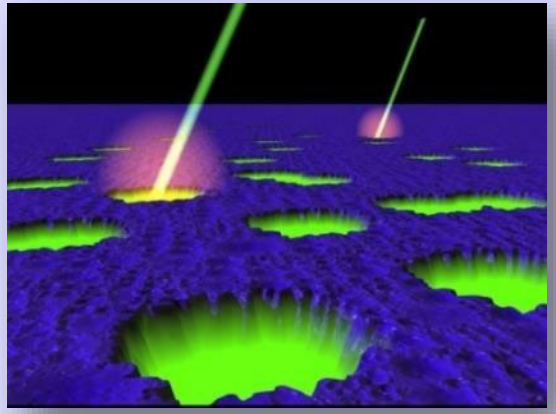
High-Pressure Cell
***P* ~ 1 Mbar**

Intense Laser Pulses
40 J/cm²



from: *www.sunbeamtech.com*



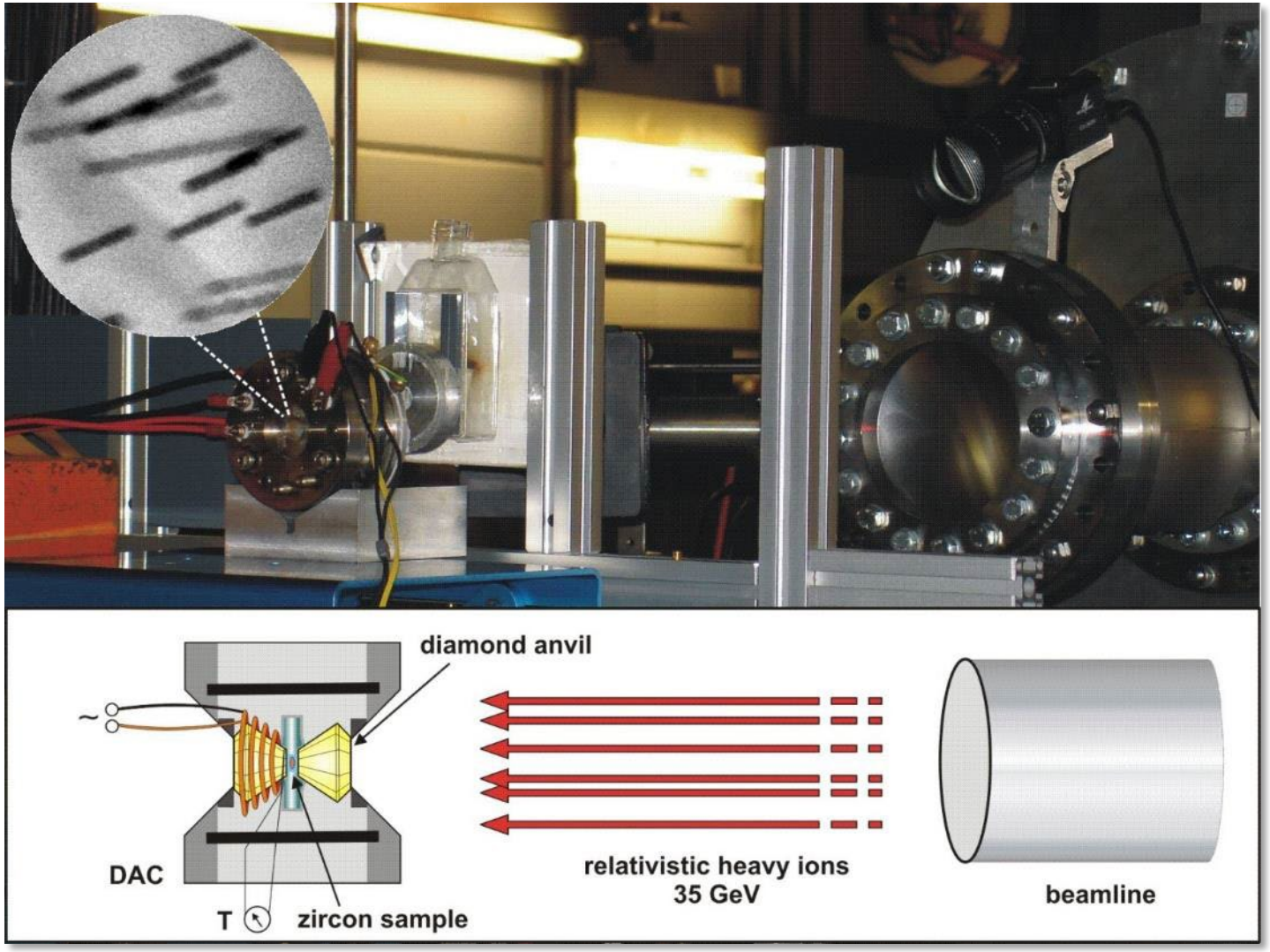


from: *www.simsworkshop.org*

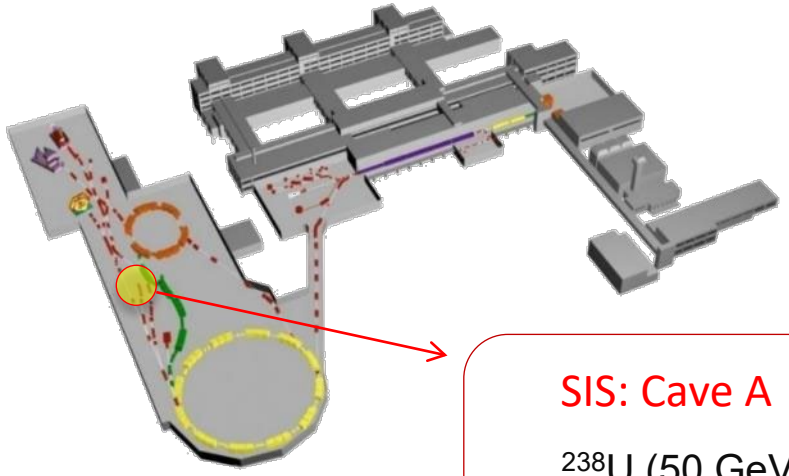
Ion Irradiation
ρ_E ~ eV/atom

International Conference on Accelerators for Research and Sustainable Development
#Accelerators2022
23–27 May 2022
IAEA, Vienna, Austria

Coupling Pressure Cells with Swift Heavy Ion Irradiation: Multiple Extremes



M. Lang, *et al.*, *Nature Materials* (2009) M. Lang, *et al.*, *Synchrotron Radiation* (2009)



SIS: Cave A

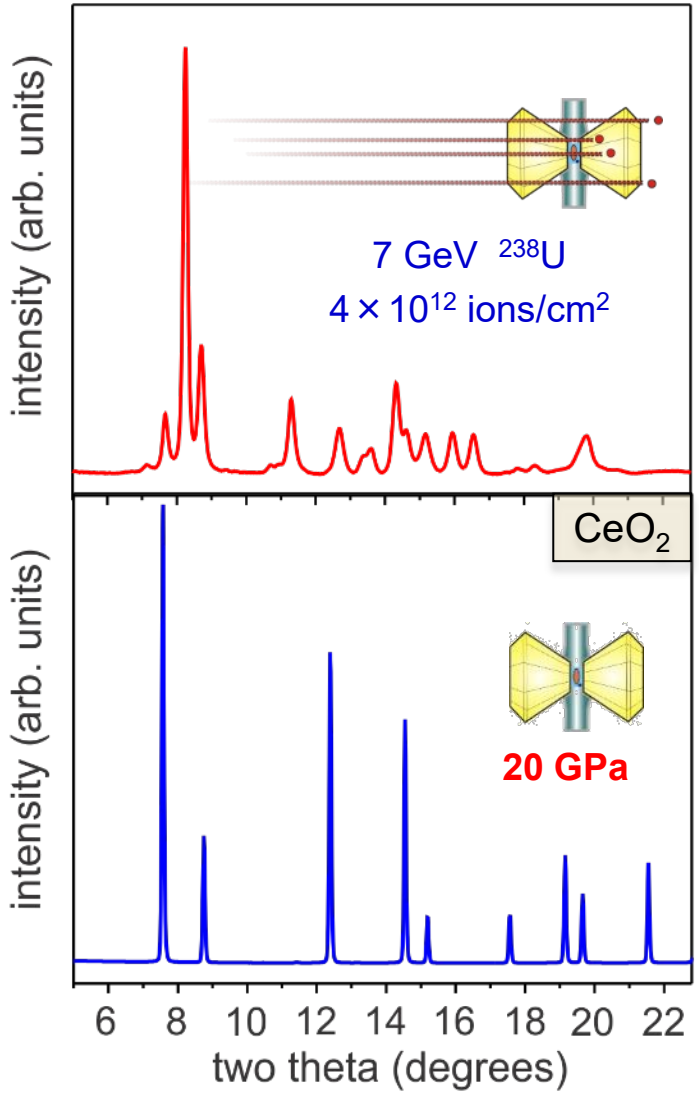
^{238}U (50 GeV)

10^{13} ions/cm^2

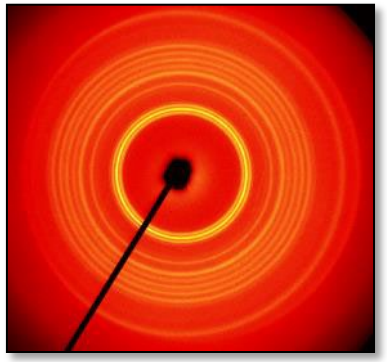
High-Pressure Irradiations

- ⇨ Relativistic heavy ions penetrate one diamond anvil of DAC (~2 mm)
- ⇨ Sample is exposed to high pressure (and high-T) when being irradiated
- ⇨ Successful to 1 Mbar and 10^{13} ions/cm^2

Example: Phase Transitions Induced under Coupled Extremes



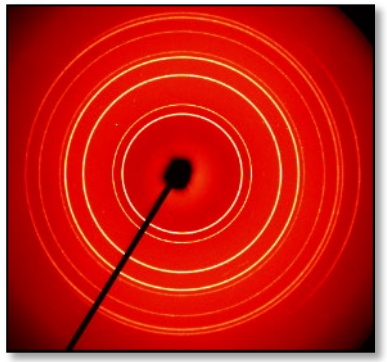
Dense high-pressure phase



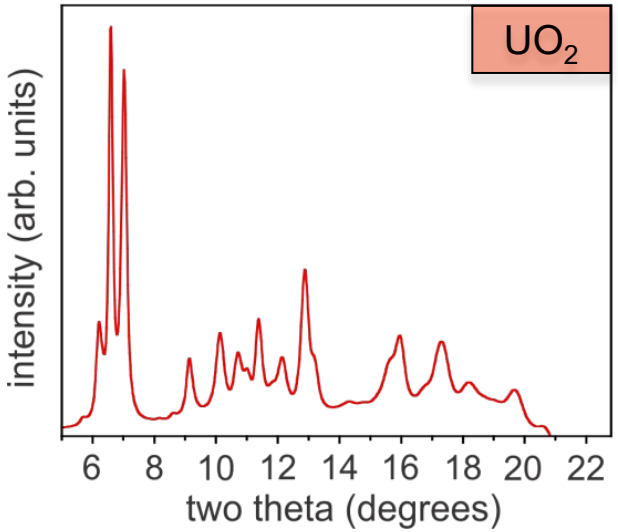
Cotunnite structure

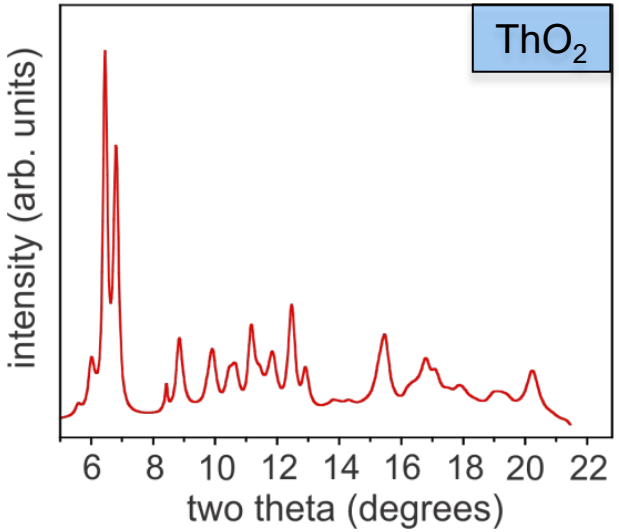
↑

Fluorite structure



Low density
Ambient phase





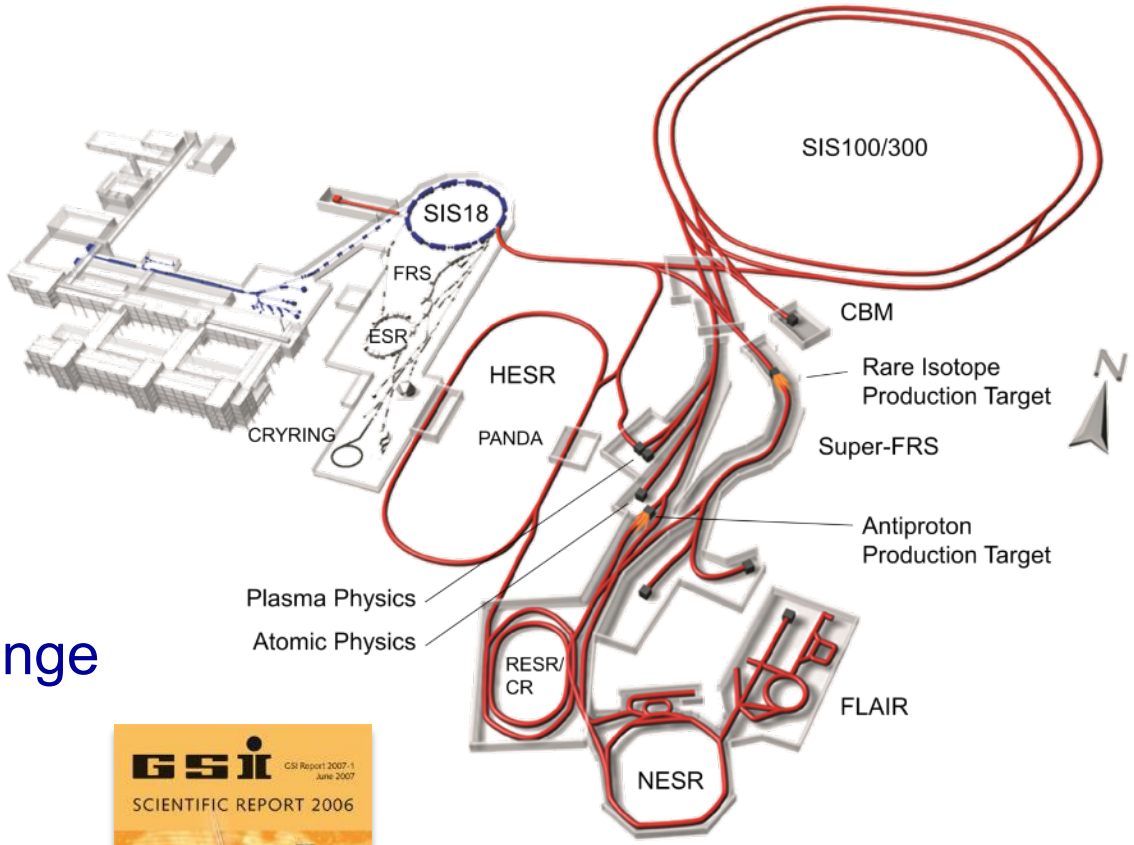
Ion-Induced fluorite-to-cotunnite Transition

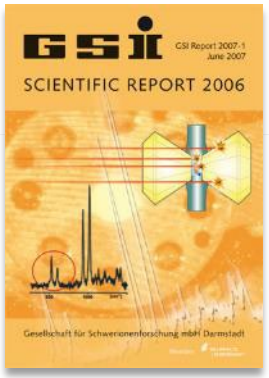
- ⇨ Ions effectively form high-pressure phase
- ⇨ Transition complete at 20 GPa (no ions: ~40 GPa)
- ⇨ Consistently observed for CeO_2, UO_2, and ThO_2

M Lang, *et al.*, unpublished data

Conclusions and Outlook

- Swift heavy ions:
  - processing of bulk material (large volume)
  - can simulate effects from fission fragments
  - induce complex structural modifications
- Accelerator-based characterization:
  - synchrotron X-rays and spallation neutrons
  - complementary probes
  - information on atomic arrangement and long-range
  - sensitive to both cation and anion sublattices
- Unique future experimental capabilities :
  - FAIR facility being currently constructed
  - European Spallation Source





FAIR

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Advanced Photon Source

Thank you!

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