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Title: Advanced Sample Environments for Neutron Diffraction Experiments

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Los Alamos NATIONAL LABORATORY EST. 1943

H. Matt Reiche, 214237 11/28/2011

Advanced Sample Environments for Neutron Diffraction Experiments

Abstract

A basic introduction to neutron scattering is presented. Current state-of-the-art of sample environments is compared with examples of Neutron Experiment User Facilities such as LANSCE, HFIR and SNS. A detailed explanation is given regarding the existing sample changer on the HIPPO flightpath at LANSCE. A robotic arm is the core of a new concept of a sample changer for HIPPO outlined in this presentation as well. A novel resistive furnace is presented using a graphite tube to reach temperatures in excess of 2300˚C. The last sample environment presented is a load frame combined with a furnace up to 1000˚C. Rotation in both furnaces allows for the study of in-situ texture development. As an application of the graphite furnace the formation of Uranium carbide is elucidated.

Advanced Sample Environments for Neutron Diffraction Experiments

H. Matt Reiche

December 5th, 2011

NM STATE UNIVERSITY

Los Alamos NATIONAL LABORATORY EST. 1943

LANSCE Los Alamos Neutron Science Center

Outline

1. Introduction to Neutron Diffraction
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3. Experimental Tools (HIPPO)
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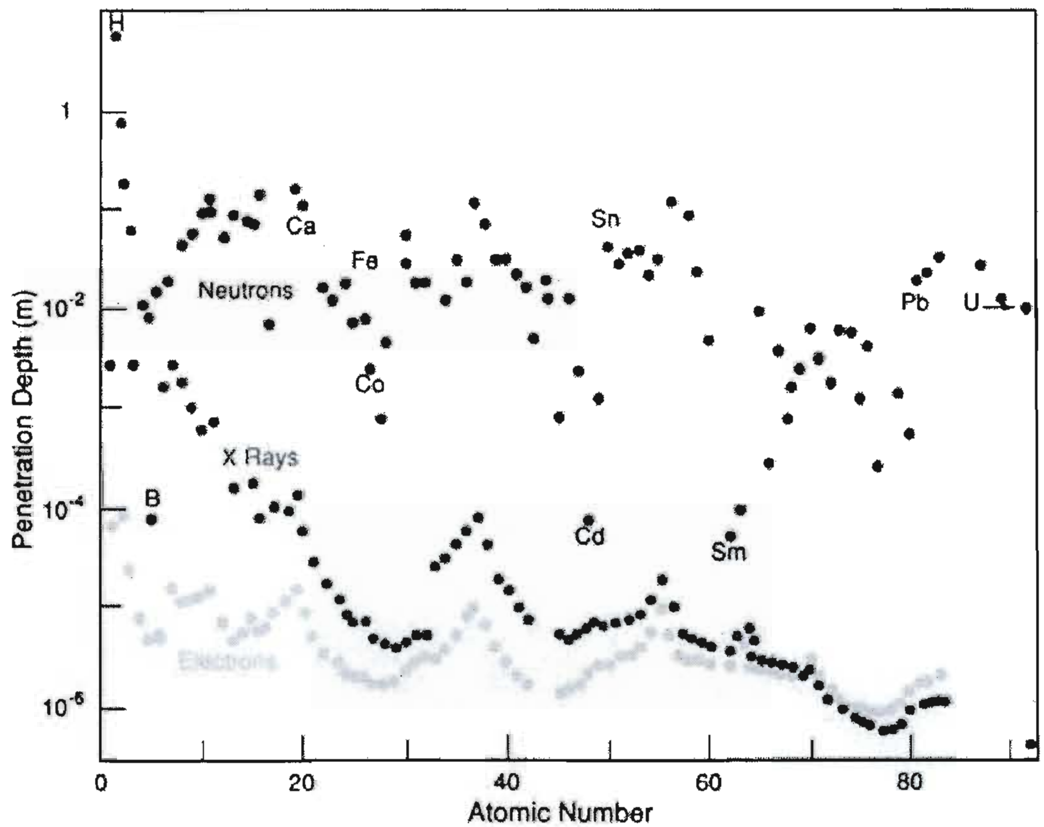
Neutron Properties

Properties:

- No charge
- Almost no electric dipole moment
- Spin-1/2
- Short range nuclear force(10^{-15}m)
- Average lifetime 888s

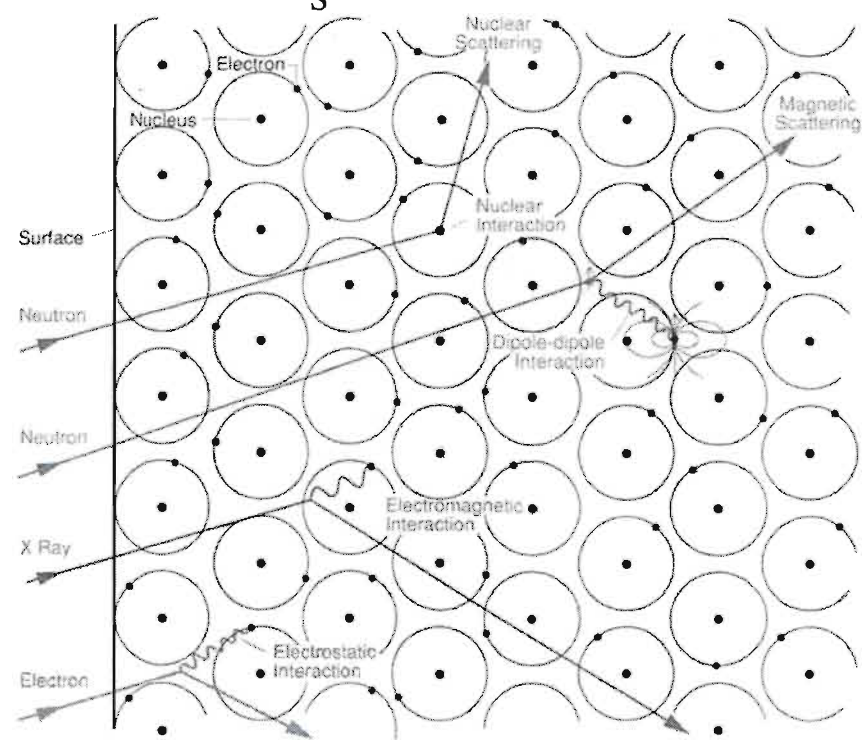


Neutron Radiation Properties

- Non- ionizing
- Neutrons tightly bound in nuclei
  - ➢ 190 MeV per n^0 from fission $v = 190{,}655{,}351 \frac{\text{m}}{\text{s}} = 0.64c \quad \lambda = 2.07^{-5}\,\text{Å}$
  - ➢ 30 MeV per n^0 from spallation $v = 75{,}758{,}754 \frac{\text{m}}{\text{s}} = 0.25c \quad \lambda = 5.22^{-5}\,\text{Å}$
- $\lambda_{thermal}$ ~$1.8*10^{-10}$m = 1.8 Å ->

$$E = 25meV \quad v = 2186\frac{\text{m}}{\text{s}} \quad T = 293K$$

$$E_{kin} = \frac{1}{2}mv^2 = \frac{p^2}{2m} = \frac{(\hbar k)^2}{2m} = \frac{h^2}{2m\lambda^2}$$



Neutron Scattering

Born approx. describing scattering of neutron by a single nucleus:

$$V(r) = \frac{2\pi\hbar^2}{m} b\delta(r)$$

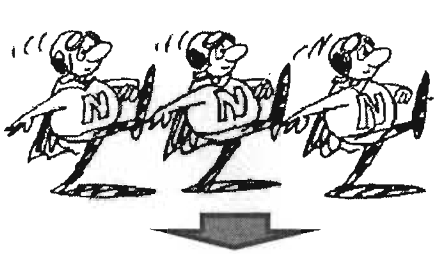
- *b* is material dependent coefficient called *Scattering Length*

$$b = b_{coh} + b_{inc} \frac{2}{\sqrt{\vec{I}(\vec{I}+1)}} \vec{s} \cdot \vec{I}$$

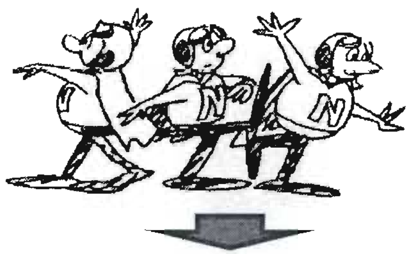
neutron spin

nucleus spin

$$b_{coh} = < b > \qquad b_{inc} = \left(< b^2 > - < b >^2\right)^{1/2}$$



Bragg peaks



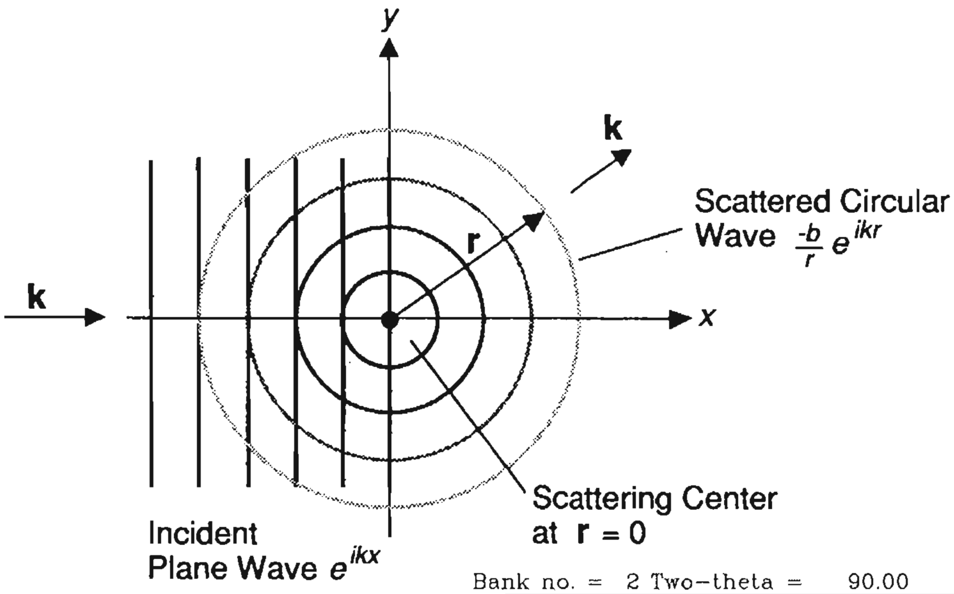
Background

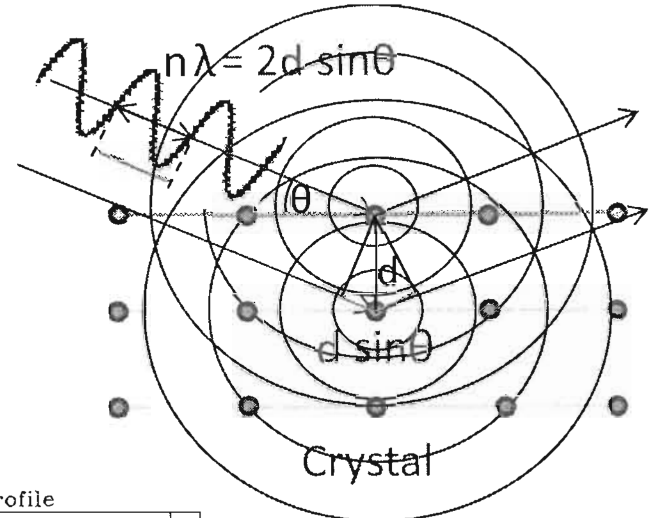
	c	I	b_{inc}	b_{coh}
$^{24}_{53}Cr$	10%	-3/2	6.87	-4.2
$^{24}_{52}Cr$	84%	0	0	4.9

Neutron Scattering Types

- Coherent scattering
  - Elastic → Equilibrium Structure
  - Inelastic → Phonons
- Incoherent scattering
  - Elastic → "Background"
  - Inelastic → Atomic Diffusion

Neutron Diffraction

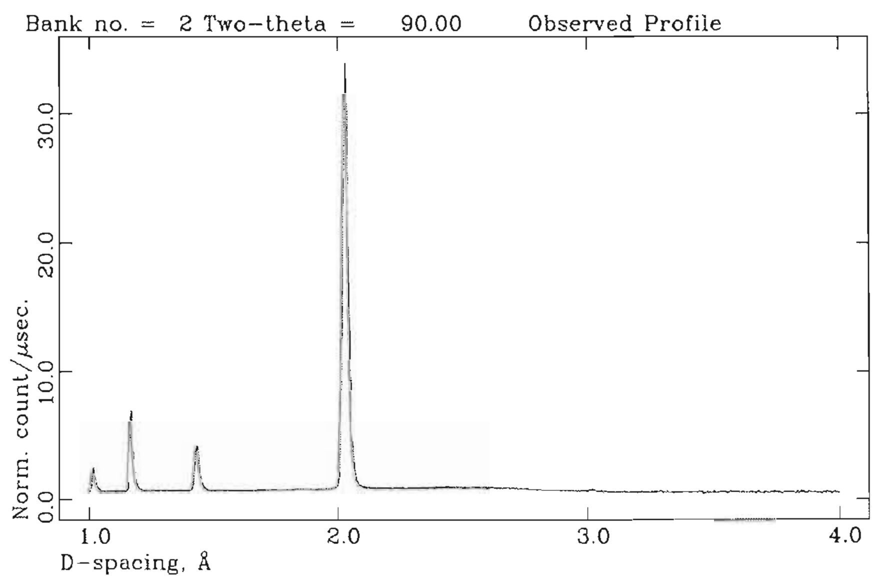




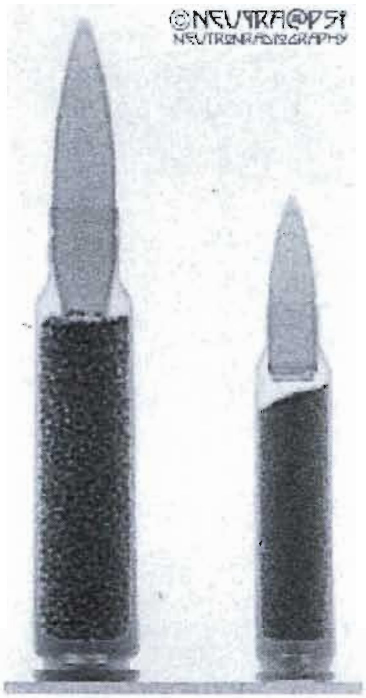
Bragg's Law

$$\left|\vec{k}\right| = \frac{2\pi}{\lambda} = \frac{2\pi mv}{h}$$

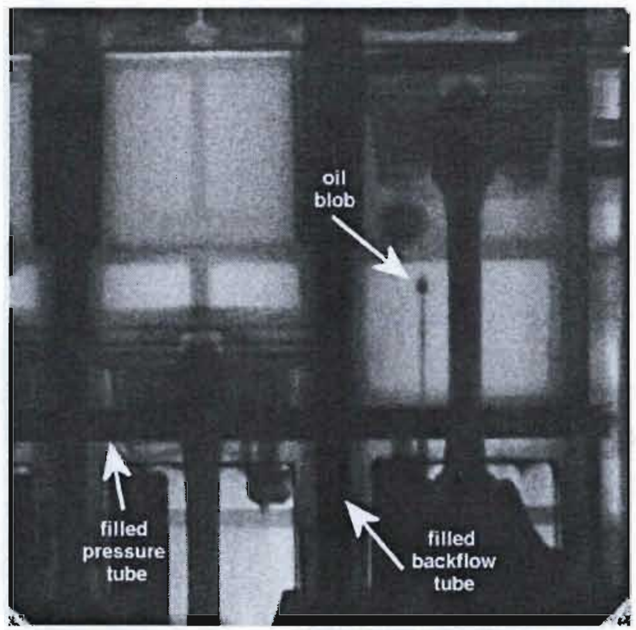
$$\lambda = \frac{h}{mv}$$



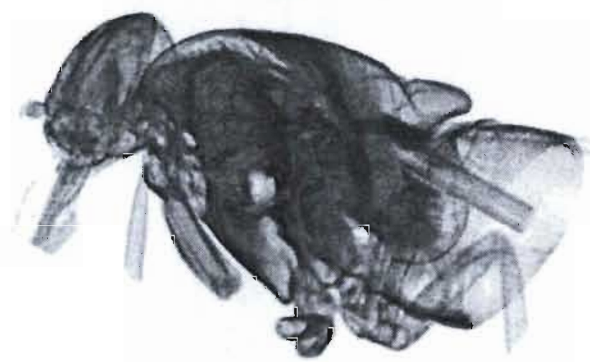
Neutron Radiography and Tomography



E. Lehmann,
Paul Scherrer Institute,
Villigen, Switzerland



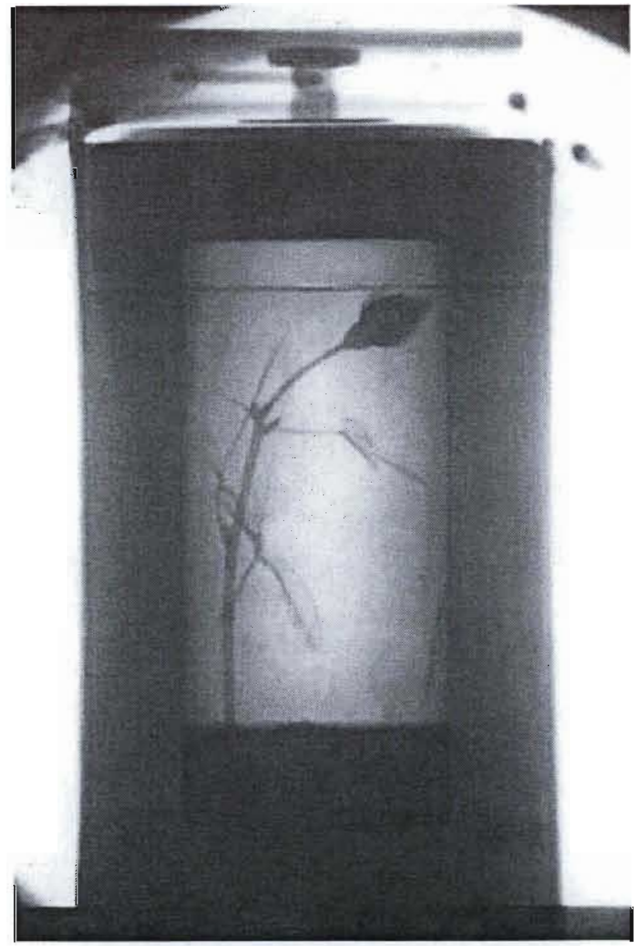
Burkhard Schillinger, FRM2
Antares beamline,
Technische Universität
München



Reconstruction by Martin
Dawson, HZB Berlin

Neutron Radiography and Tomography

A rose in a lead container used for transporting radioactive materials.



Vogel, Priesmeyer, 2006

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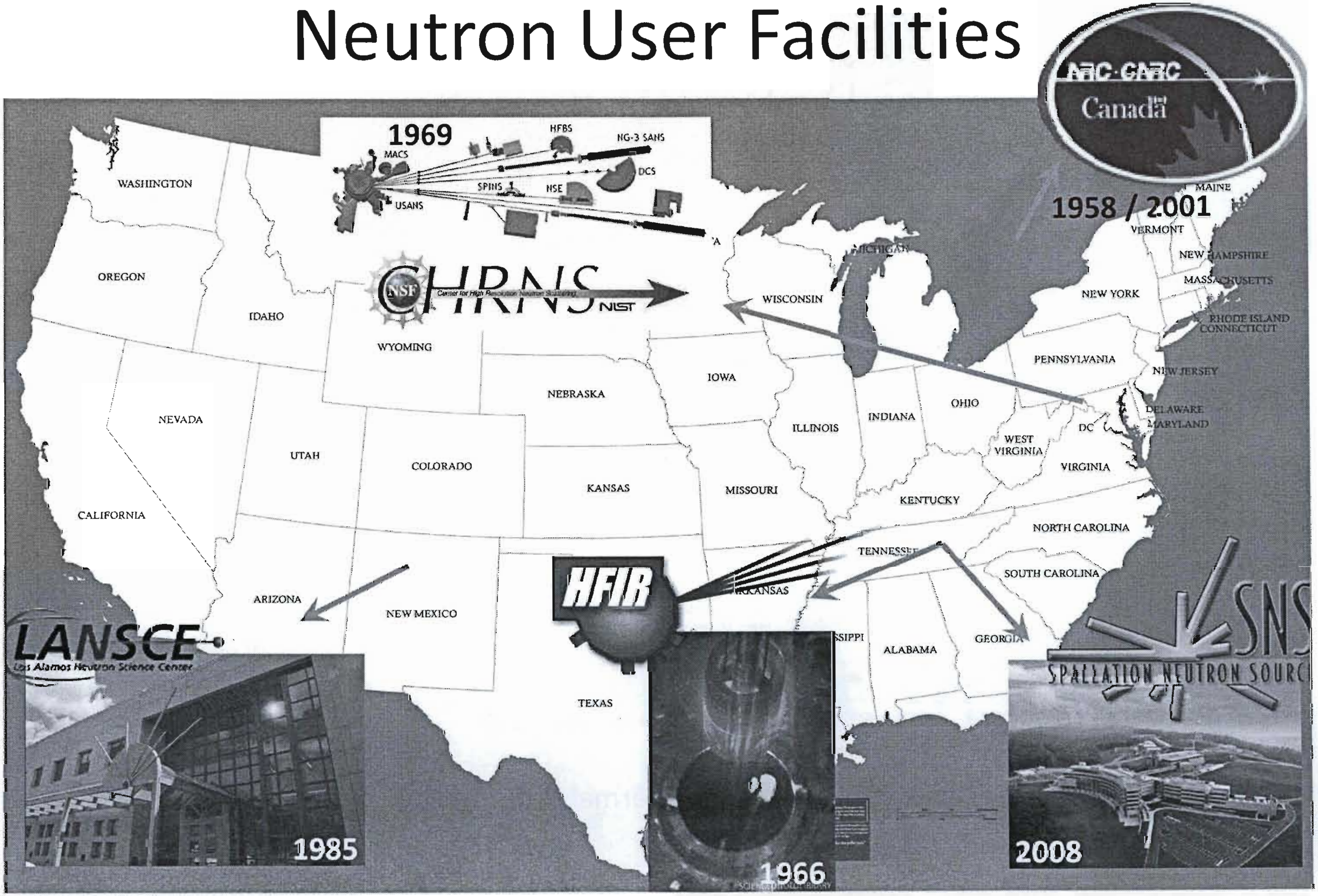
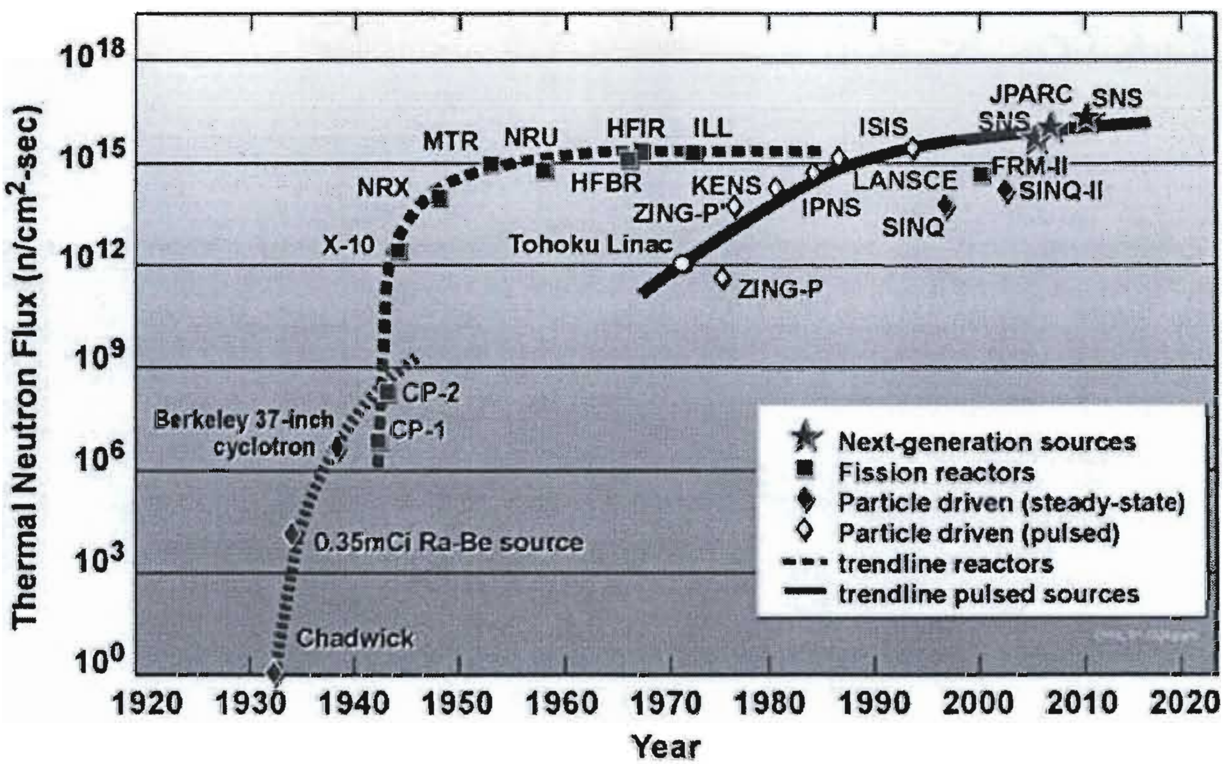
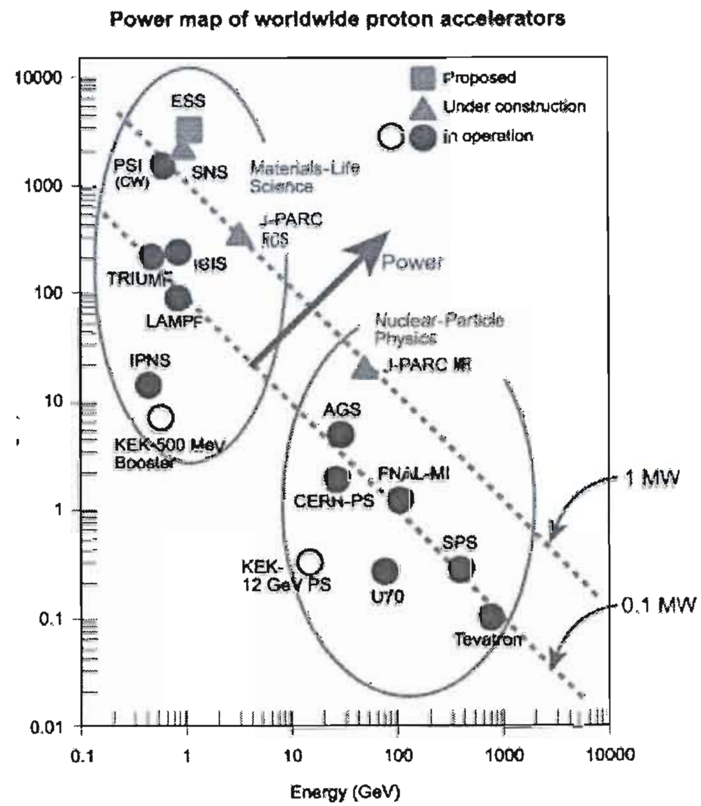
Neutron User Facilities



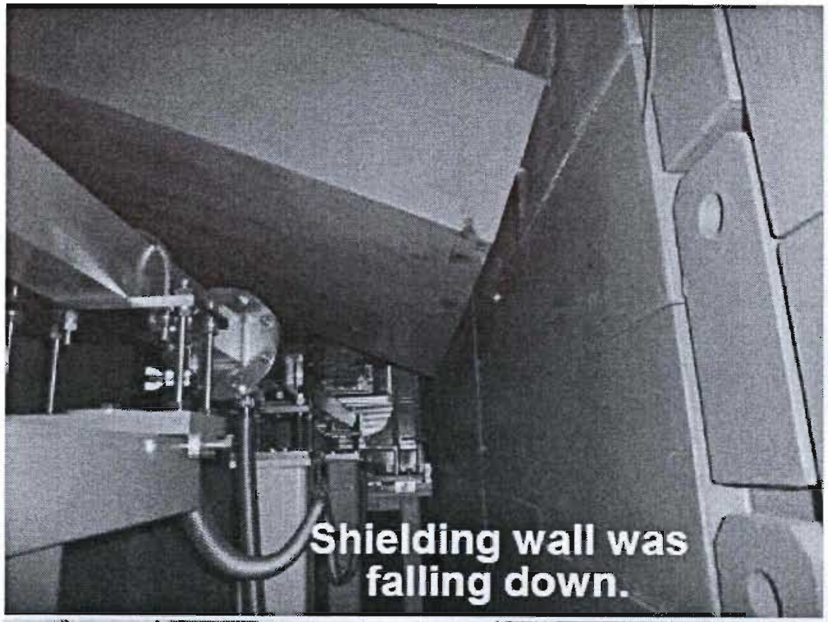
Historic Development of Neutron Sources

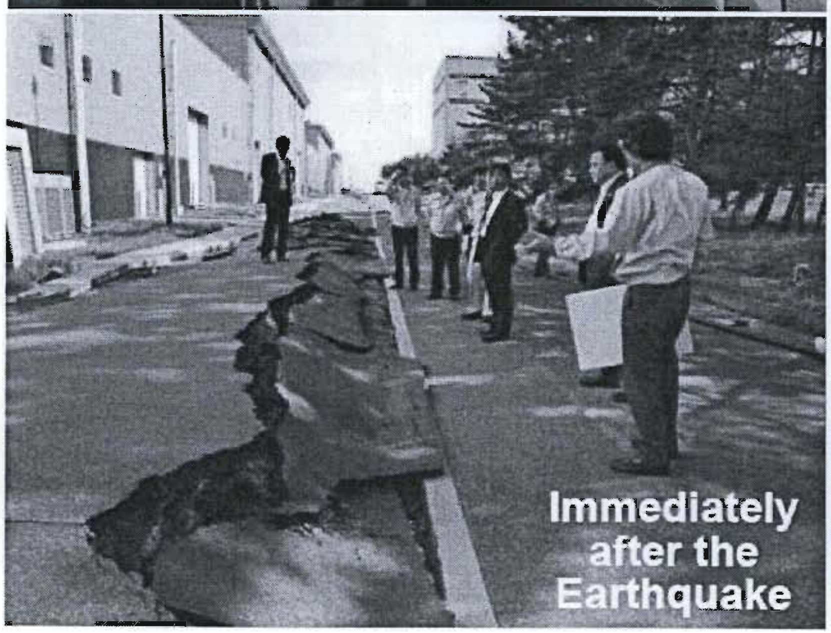


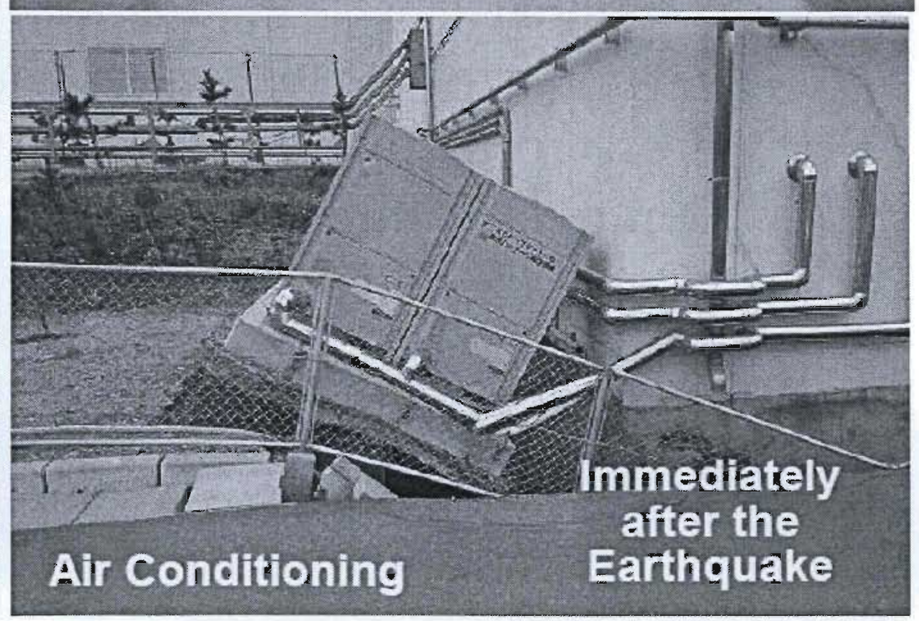


Peak Thermal Neutron Flux

J –PARC after Earthquake



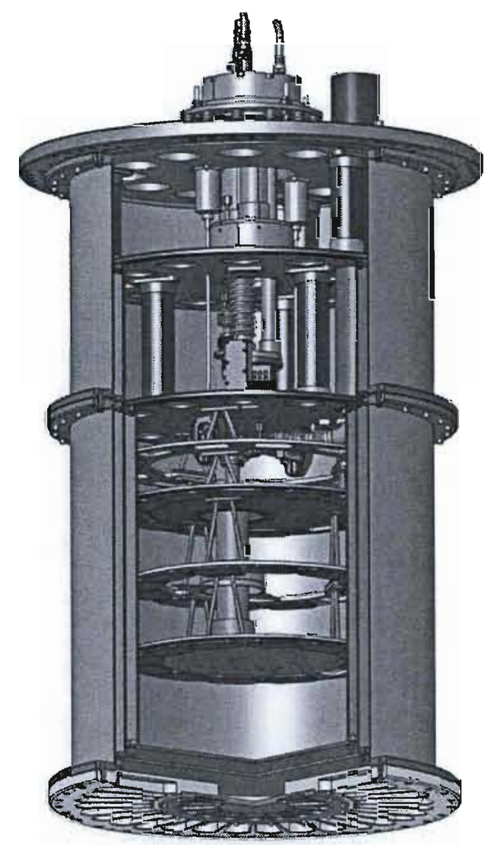


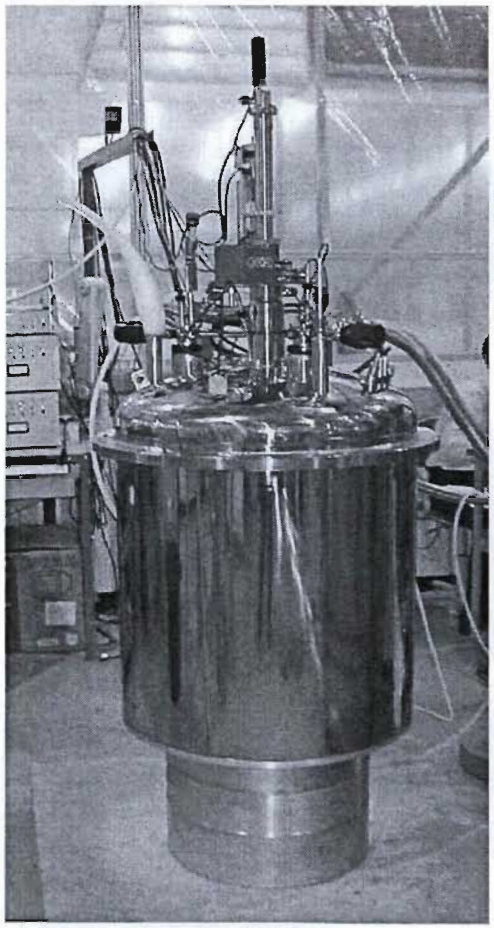


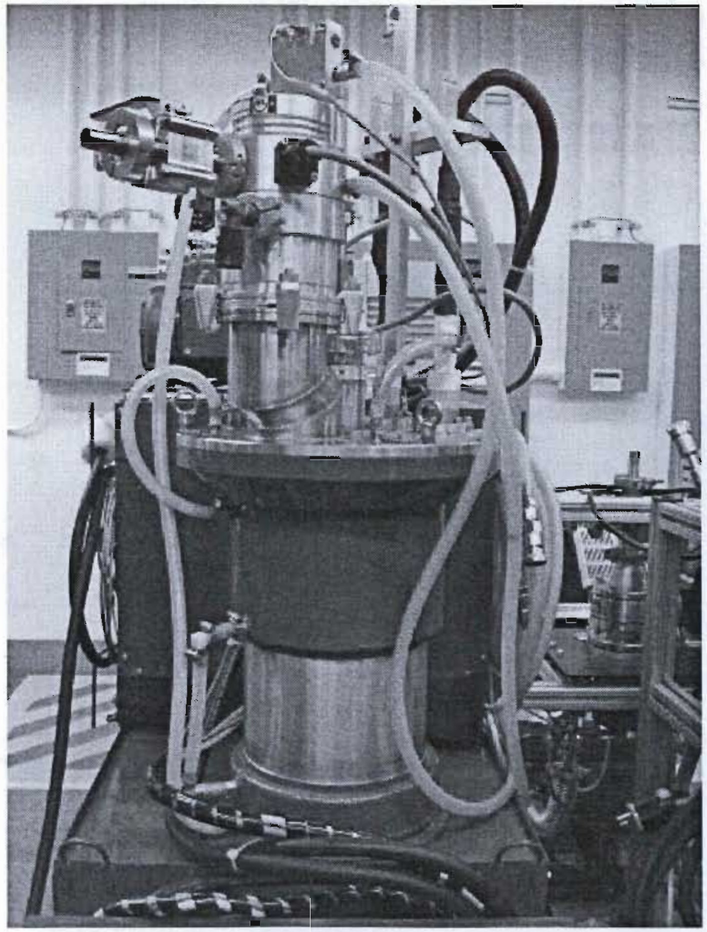
Sample Environments:

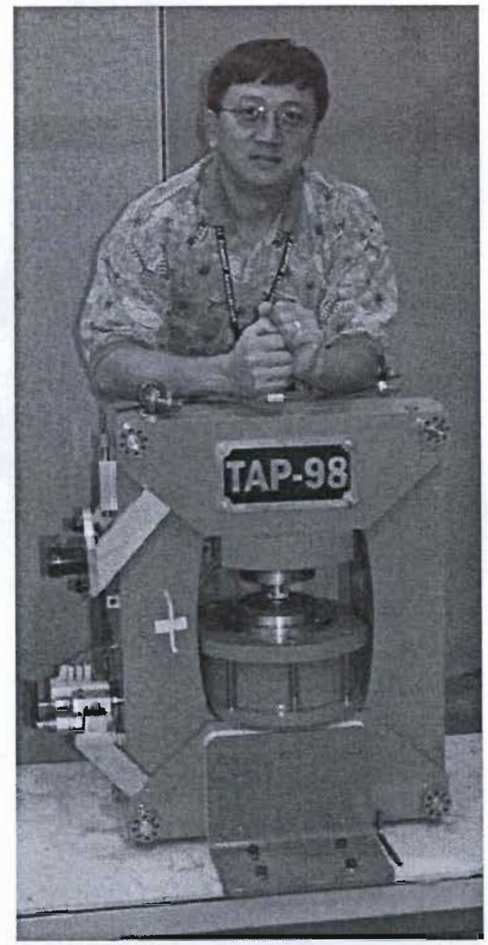
Any combination

- Cryogenic Temperatures:
  - Liquid He Cryostats ("Orange Cryostat") ~ 1.5 K
  - Closed Cycle Refrigerators (Joule expansion cycles) ~ 2-4K
  - Dry Dilution Fridge ~ 25mK
- High Temperature/ Furnace:
  - ILL-Furnace (V: ~1100°C; Nb: ~1800°C)
- Magnetic Fields:
  - Pulsed
  - Continuous: <16T superconducting (SNS)
- Pressure: Vacuum - 20GPa = 200kbar
- Controlled Atmosphere:
  - Specific gas composition and pressure
  - Humidity level

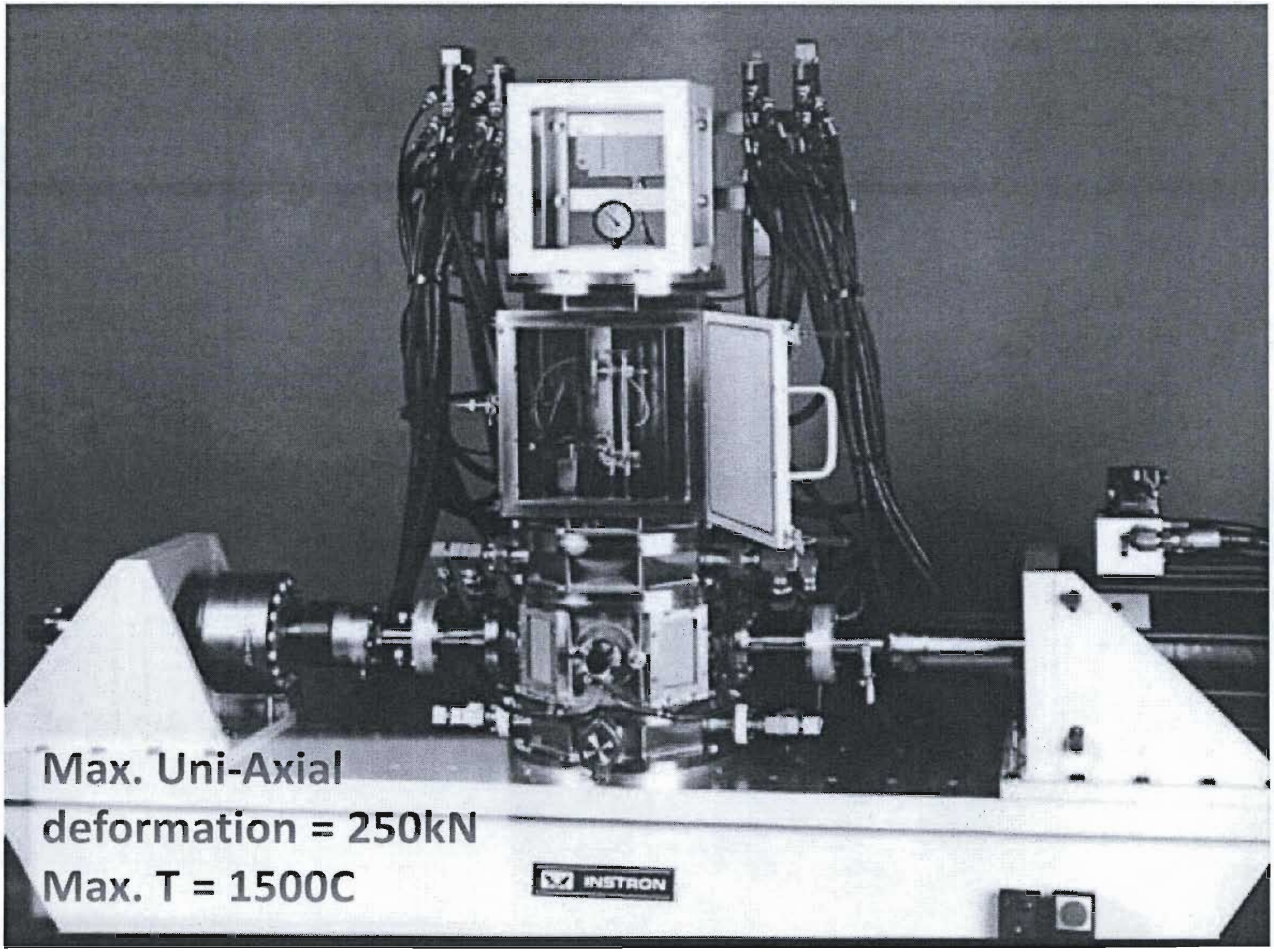







TAP-98

SMARTS Furnace



Equivalent

Facility	Beamline
SNS, TN, USA	POWGEN3
ISIS, UK	ENGIN-X
ILL, France	XX

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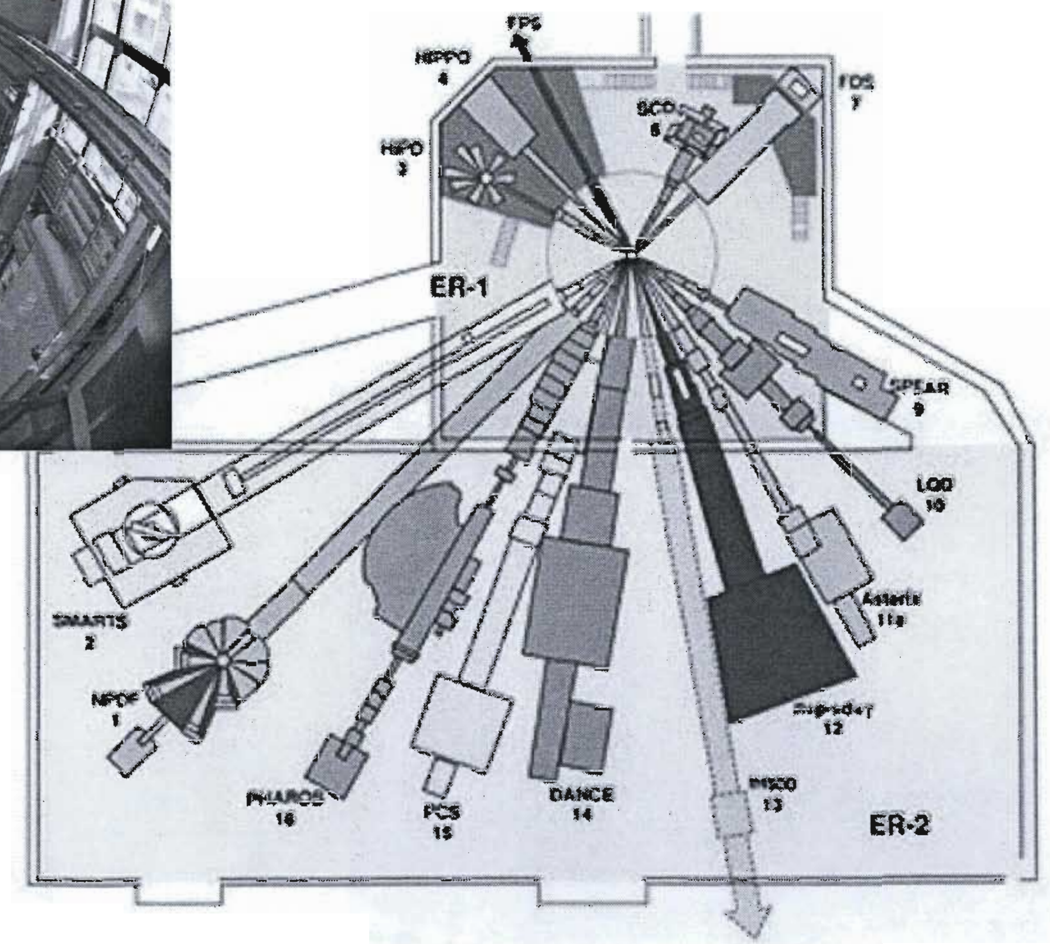
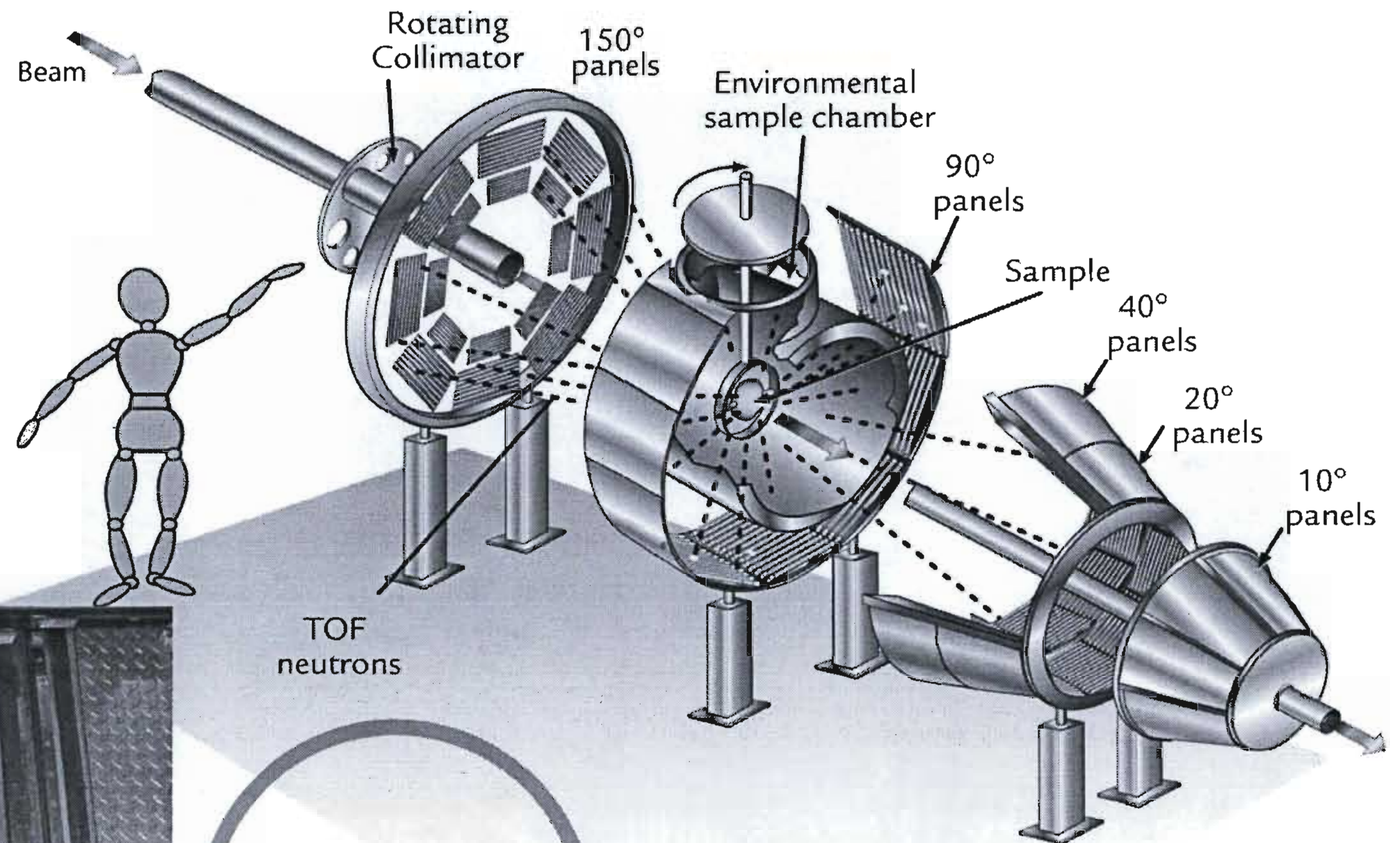
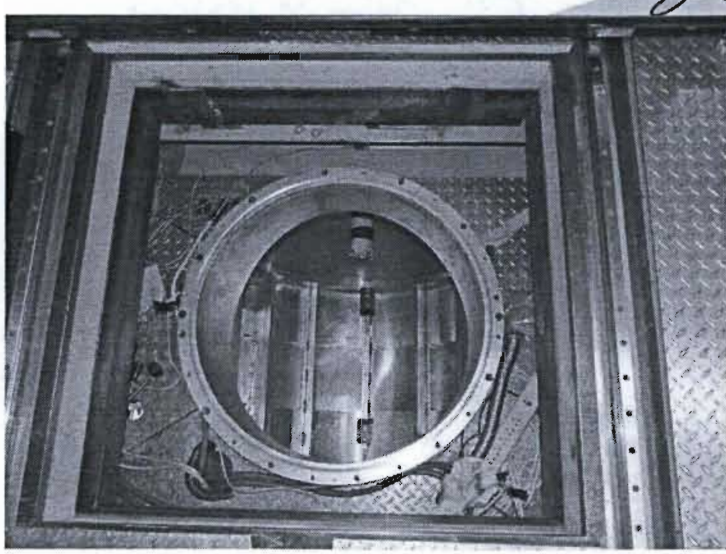
Lujan Center at LANSCE



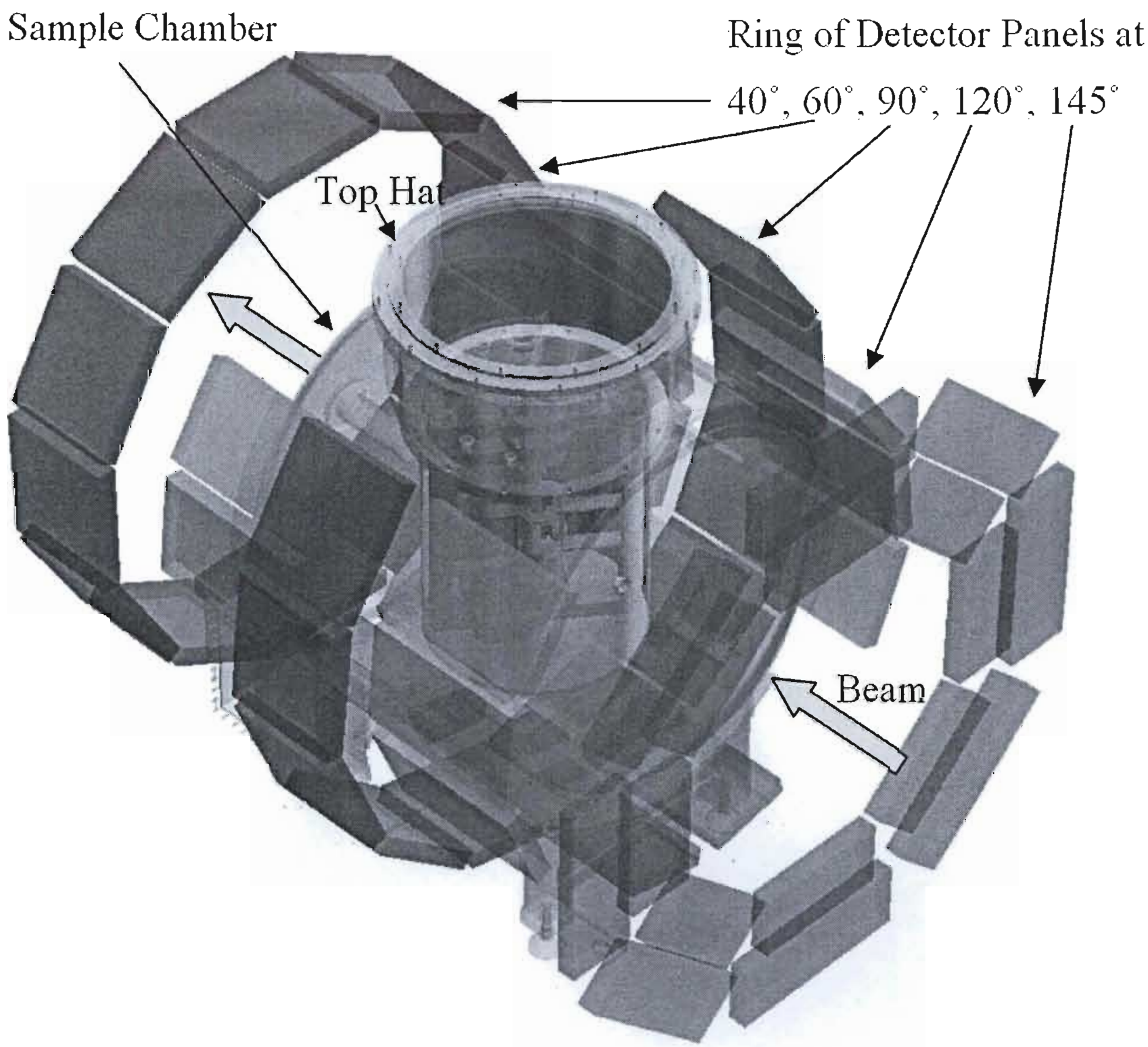
800 MeV protons
With H_2 cold source

High Pressure – Preferred Orientation





HIPPO: New Detector Arrangement



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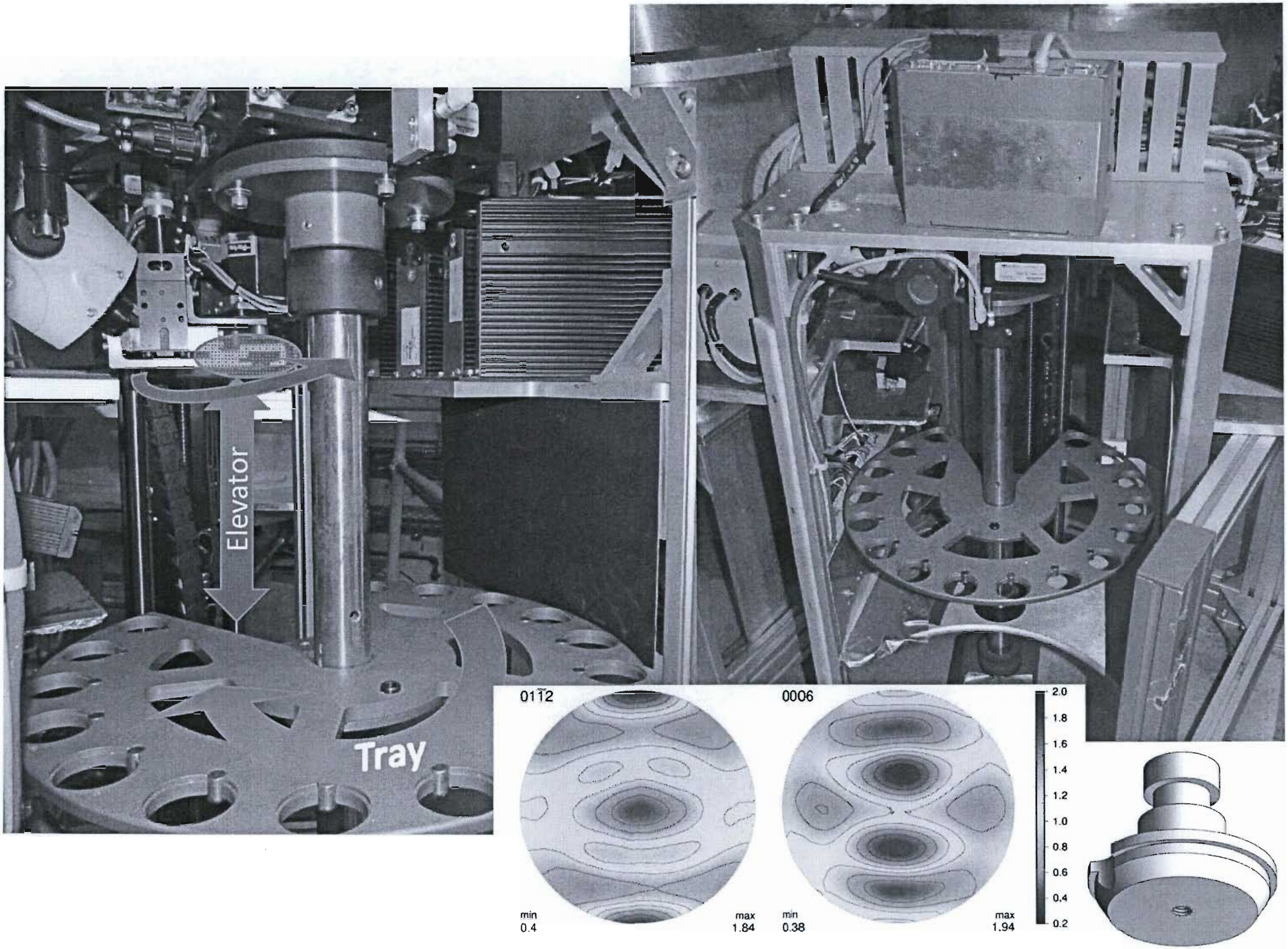
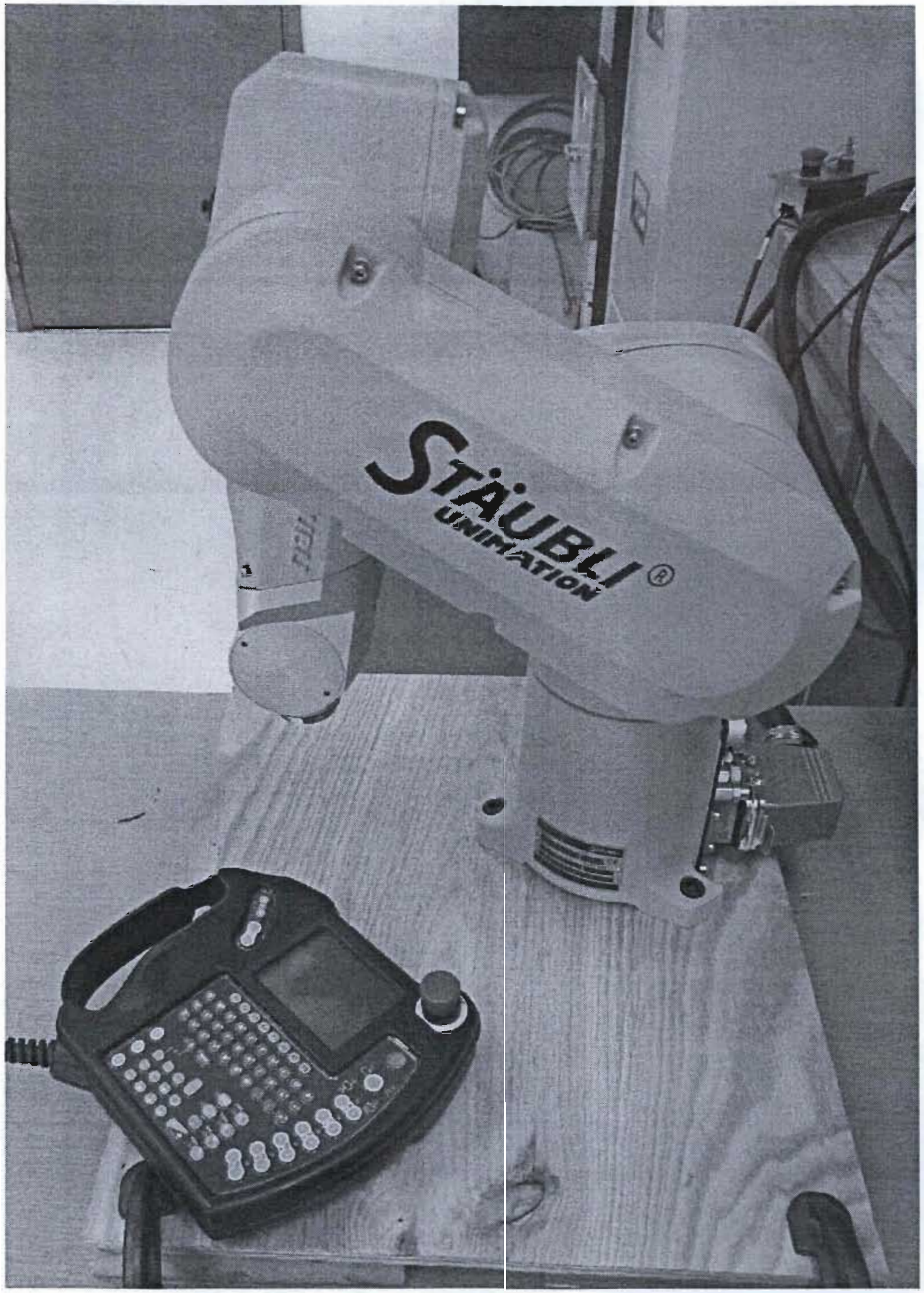
HIPPO Sample Changer

- H.M. Reiche, S.C. Vogel, *A versatile automated sample changer for texture measurements on the high pressure-preferred orientation neutron diffractometer*, REV SCI INSTRUM 81 (2010) 93302
- Need: average measuring time 15min/orientation -> 45min/sample
- Availability: No commercial available sample changer for neutron diffraction experiment. Custom build versions:
  - Shah, 1991, Physica B
  - Rix el al., 2007, Rev. Sci. Instrum,

- ❑ No preferred orientation / texture
- ❑ space limitations
- ❑ **=> Custom Solution necessary!**


Elevator
Tray
01$\bar{1}$2
0006
min 0.4
max 1.84
min 0.38
max 1.94
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2


STÄUBLI
UNIMATION

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High Temperature Furnace

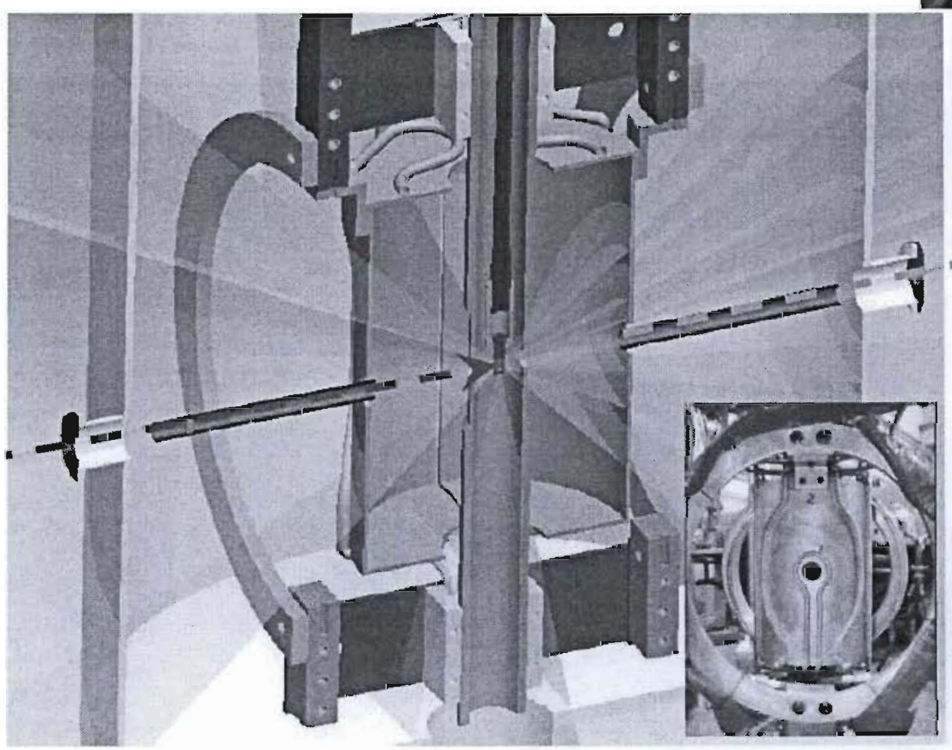
- Heat distribution:
  1. Conduction $\vec{q} = -k\nabla T$
  2. Convection
  3. Radiation
- Sample protection from Oxidation:
  1. Cover Gas
  2. Vacuum
- Safety Interlock
  1. Water, Vacuum loss
  2. Over Temperature at Multiple Locations

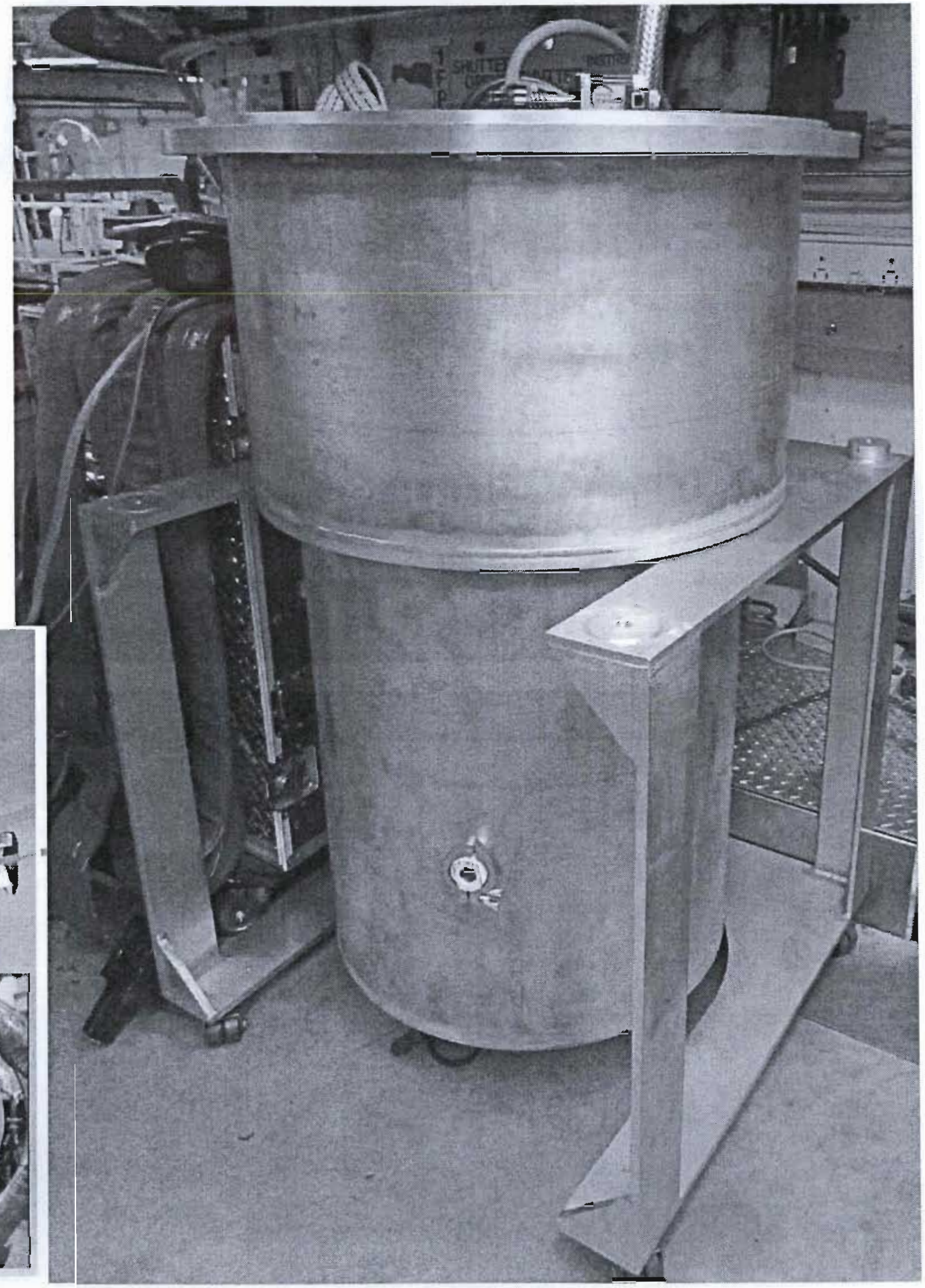
Beam Alignment

Alignment with Image Plates:

1. ^{157}Gd irradiated with Neutrons
2. β-particles with 5μm range in silver halide
3. Capture reaction results in semi-stable excitation state
4. De-excited by laser stimulation

- Max T=2300°C
- Max Rate=10°C/s
- 360° Sample Rotation
- 50mm Sample Height Adjustment





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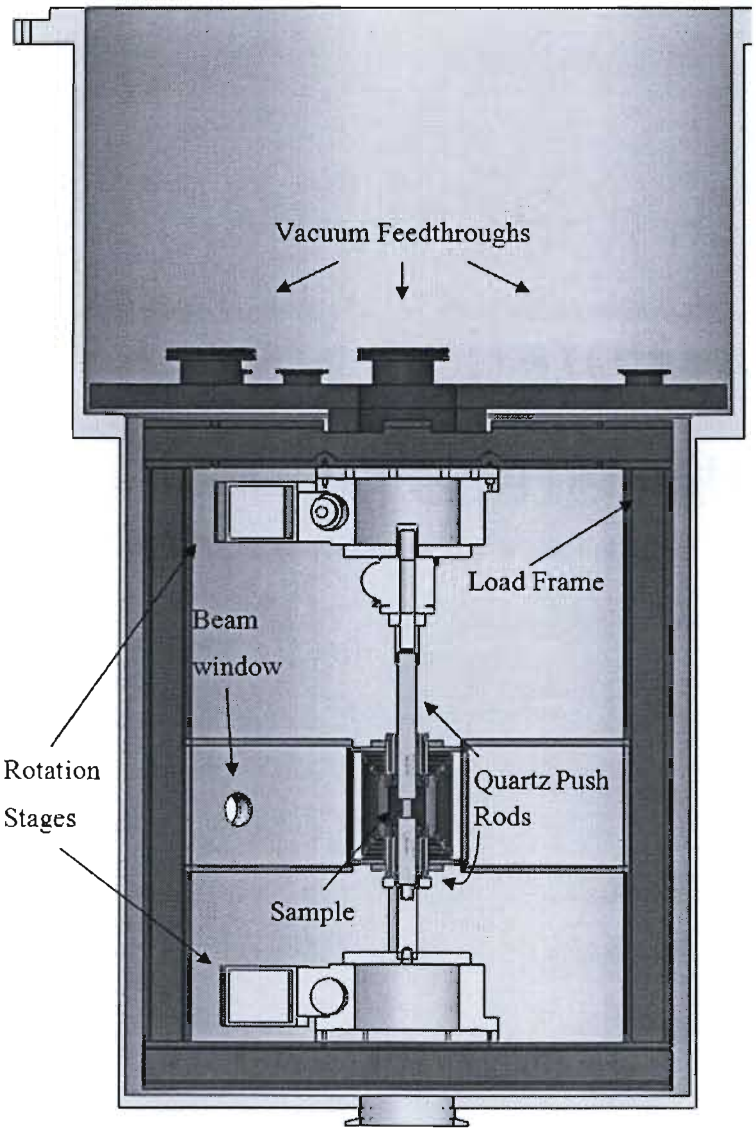
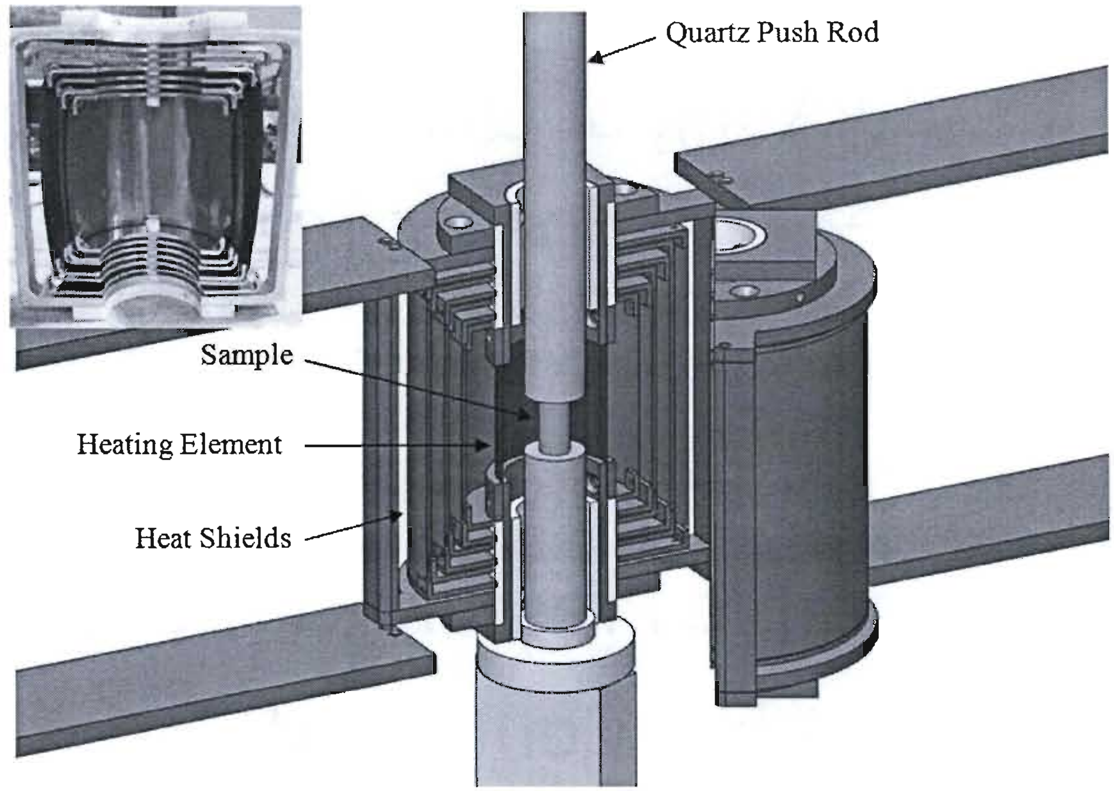
Load Frame with Furnace and Rotation

- Buckling of Quartz
- Max. T = 1000°C (Quartz Push Rods, Vanadium)
- Max. uni axial stress = 2.7kN (Rotation Stage)
- Sample Deformation via Motor Steps
- Sample Pressure via Load Cell
- LabView Controlled

Load Frame





Fused Quartz Push Rods:
- Amorphous -> no Bragg Reflexes
- High Temperature Resistance (~1100°C)
- High Compressive Strength (1100MPa)

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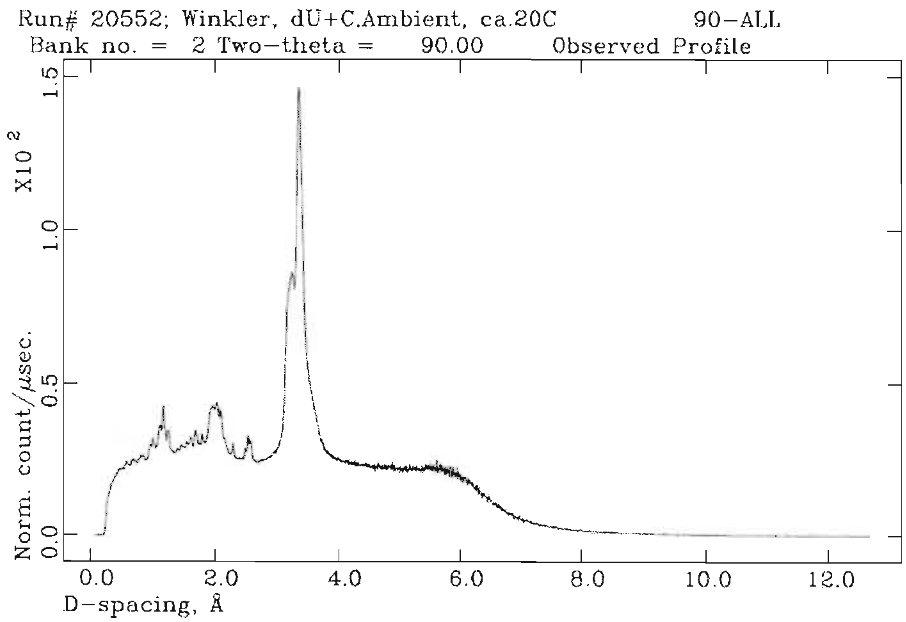
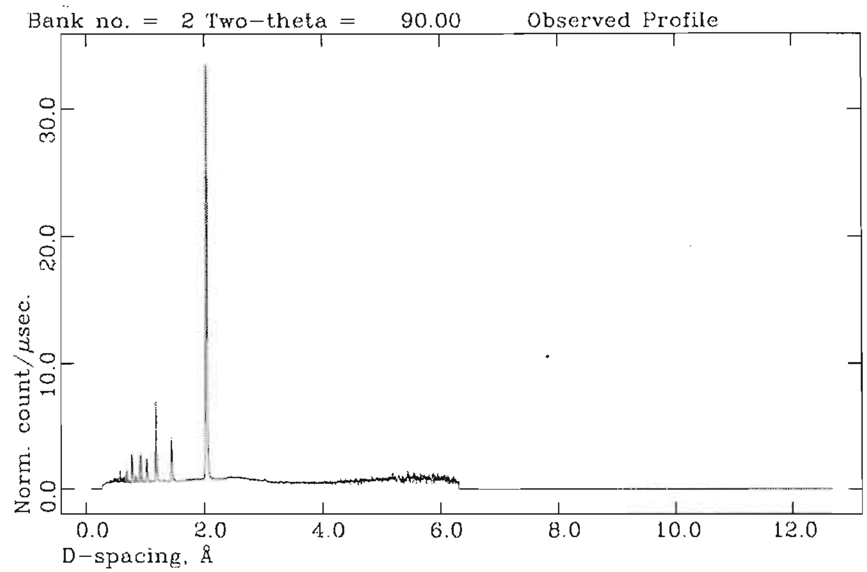
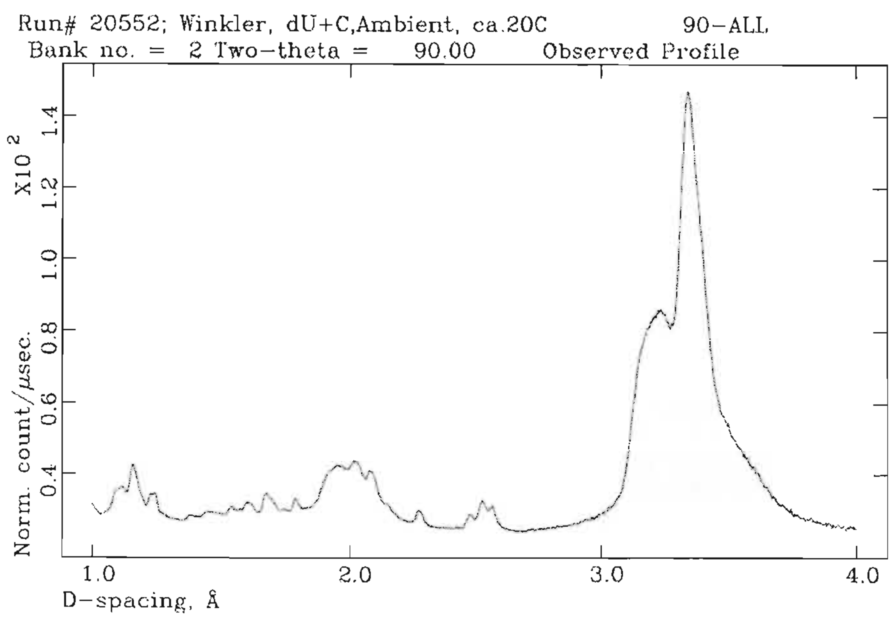
Uranium Carbide

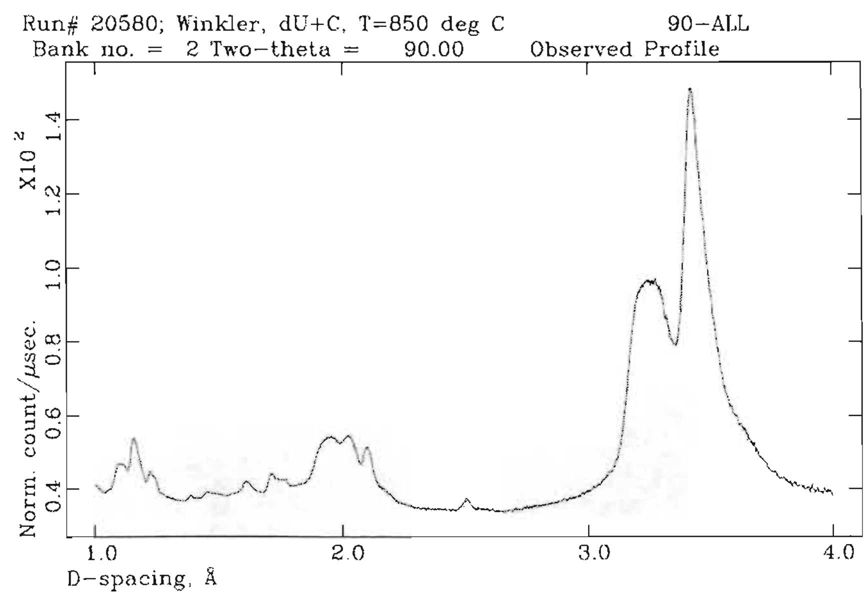
- Significance:
  - Nuclear Fuel Rod in Reactor
- Melting Point:
  - U: 1132°C
  - UC: 2790°C

Diffraction Pattern

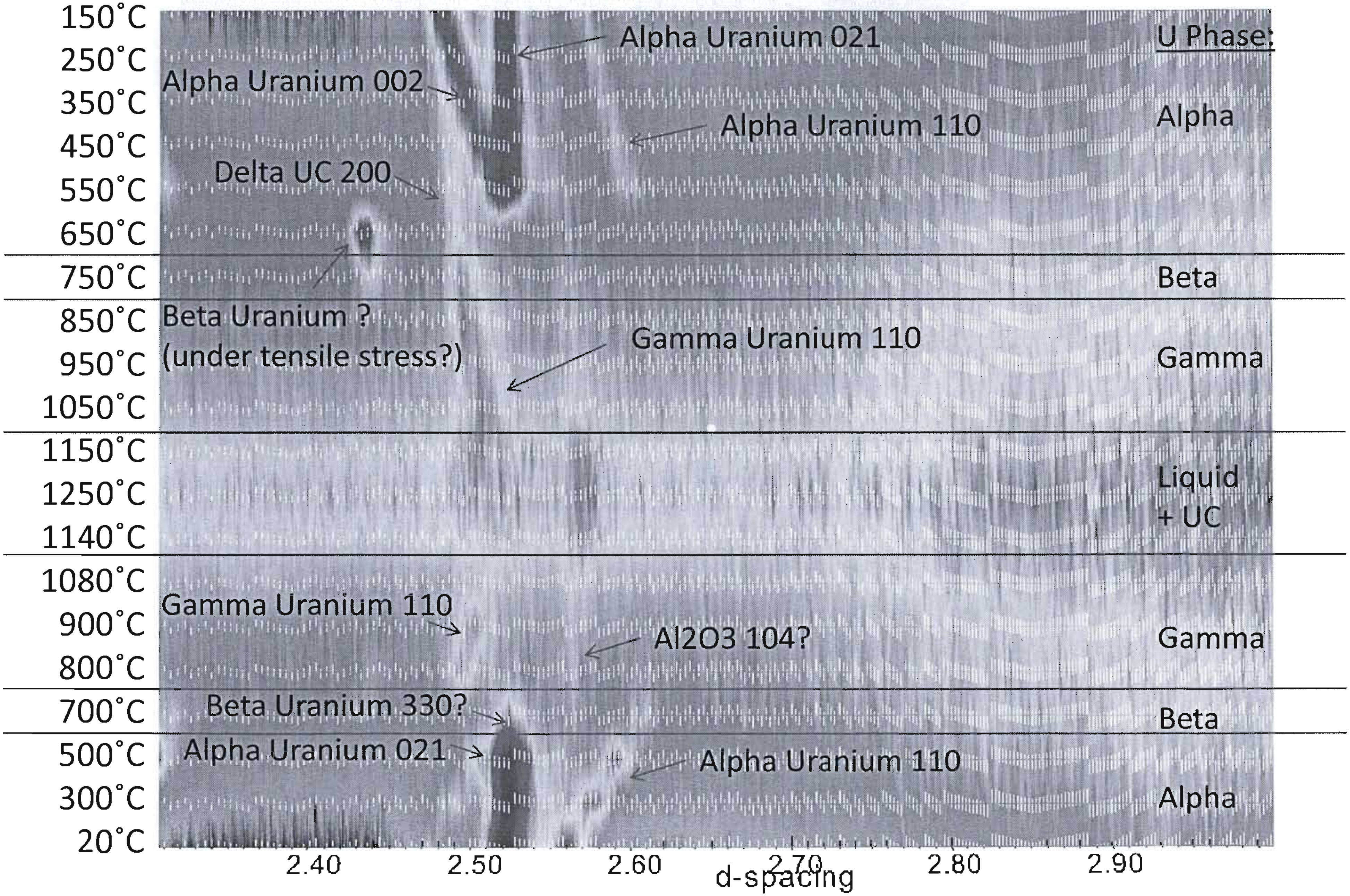








Overview of Phase Changes in Uranium



References

- **V. *SEARS , Neutron News, Vol. 3, No. 3, 1992***
- ***L. Liang* - Neutron applications in earth, energy and environmental sciences, Springer, 2009**
- **Birkbeck College, University of London**
- **Vogel, Priesmeyer, Rev. in Mineralogy & Geochem., Vol 64, 2006**

Acknowledgements

Special Thanks to:

- Our Creator
- Dr. Sven Vogel
- Dr. Heinz Nakotte
- Dr. Steve Stochaj

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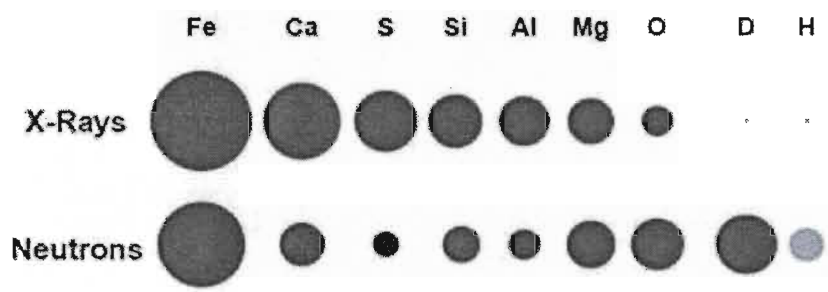
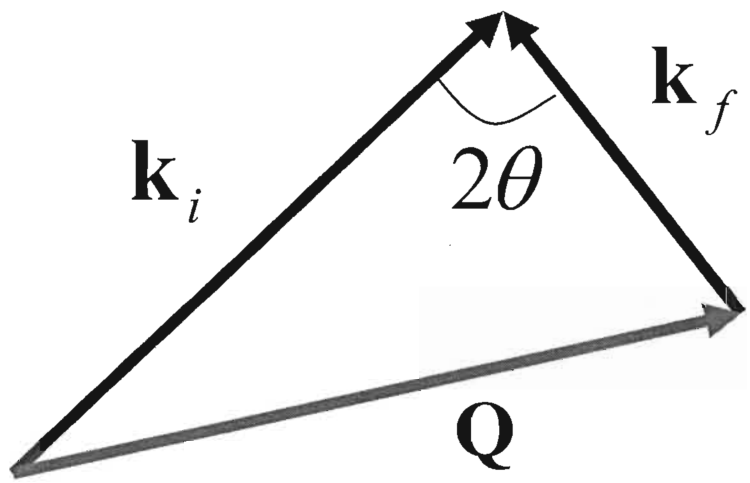
NM STATE UNIVERSITY

Neutrons vs. X-rays



Atom	Bound Coh. Scatt.	Incoherent scatt.
H	-3.7	80.3
D	6.7	2.1
C	6.6	0.0
O	5.8	0.0
N	9.4	0.5
S	2.8	0.0

Nuclear vs. Magnetic Scattering



$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$$

$$\hbar\omega = E_i - E_f$$

Nuclear scattering

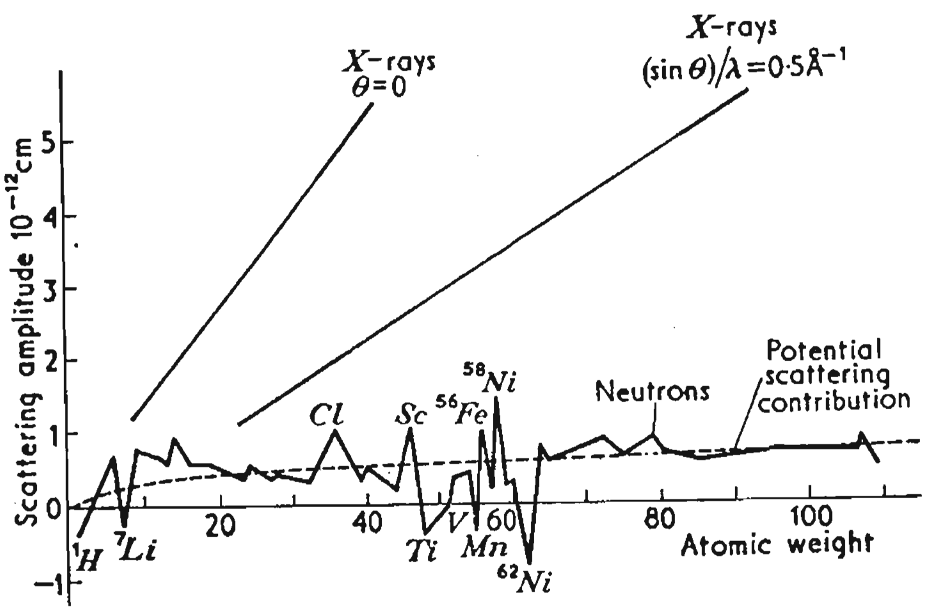
$$\mathrm{S}(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, e^{-i\omega t}\,\frac{1}{N}\left\langle \rho_{\mathbf{Q}}(0)\rho_{-\mathbf{Q}}(t)\right\rangle$$

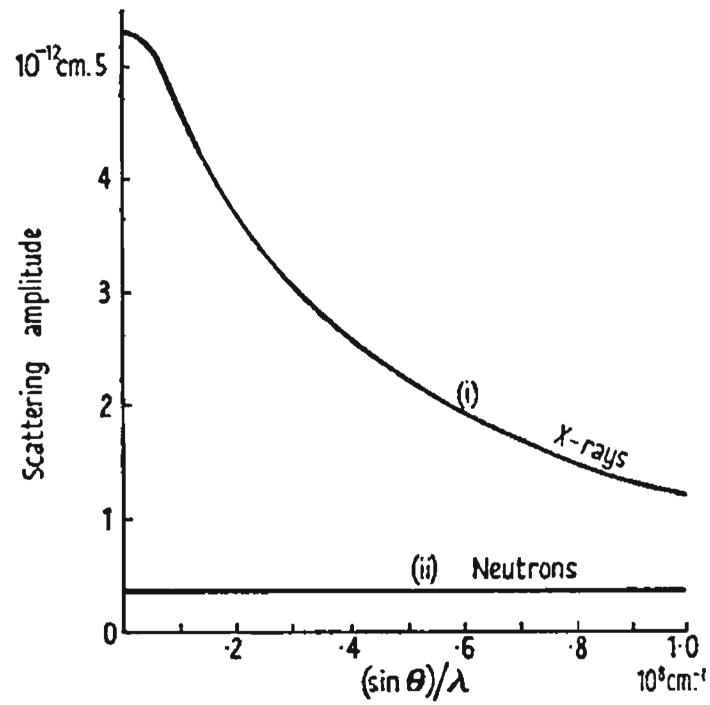
Magnetic scattering

$$\mathrm{S}^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, e^{-i\omega t}\,\frac{1}{N}\sum_{\mathbf{RR'}} e^{i\mathbf{Q}\cdot(\mathbf{R}-\mathbf{R'})} < S^{\alpha}_{\mathbf{R}}(0)\,\mathrm{S}^{\beta}_{\mathbf{R'}}(t) >$$

Scattering Factors f, cont'd

- For x-rays the magnitude of f is proportional to Z
- For neutrons nuclear factors determine f, thus no regular with Z (different isotopes can have different f s)





For neutrons conventionally f = b
(Scattering length - constant for an element)

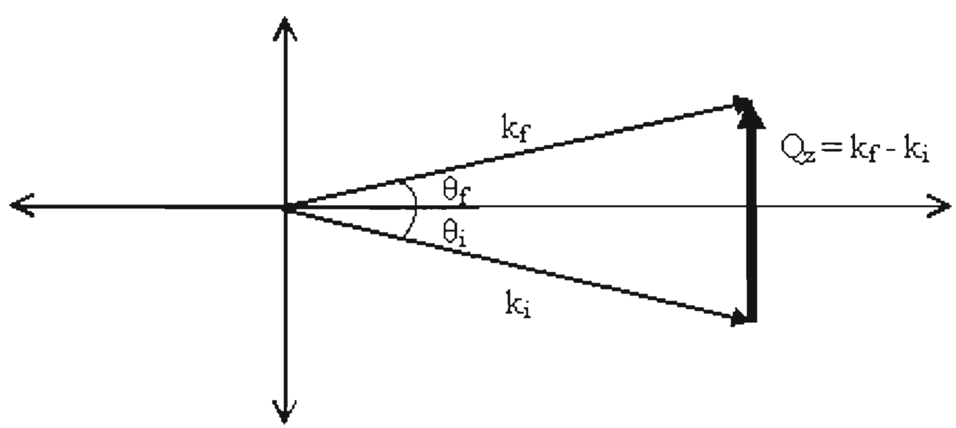
Neutron Reflectivity

Index of refraction:

$$n = 1 - \delta + i\beta$$

Snell's Law:

$$n_i \cos \theta_i = n_{refr} \cos \theta_{refr}$$



$$\overrightarrow{Q} = \overrightarrow{k_f} - \overrightarrow{k_i}$$

$$\left|\overrightarrow{k_i}\right| = \left|\overrightarrow{k_f}\right| = \frac{2\pi}{\lambda}$$

$$\left|Q_z\right| = \frac{4\pi}{\lambda} \sin \theta$$

Intensity recorded by detector from a powder

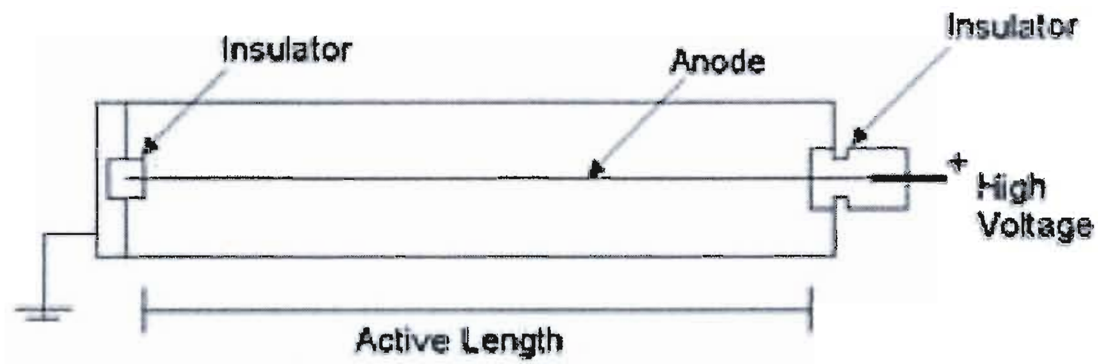
$$I \propto S(Q) = \lambda^3[I_o][SF][G][M][TF][LF][AF][PO][EE]|AA^*|$$

- I_o = Incident intensity
- [SF] = Overall scale factor (det. efficiency, everything else you forgot)
- [G] = Geometrical factors of instrument and sample (e.g., density)
- [M] = Multiplicity of reflection [# cooperating planes, e.g. 8 (111)]

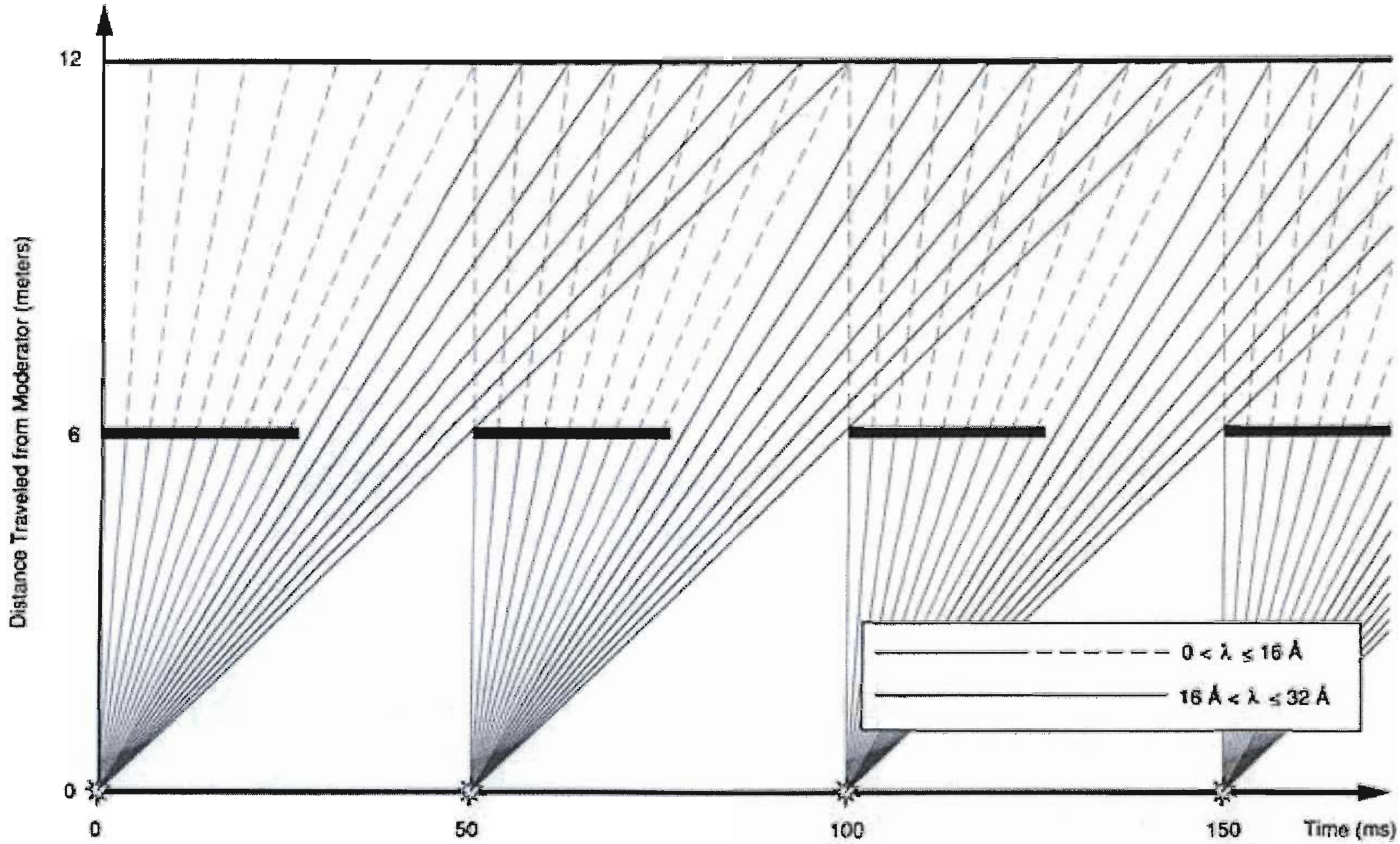
- [TF] = Debye Thermal Vibration Factor = e^{-2W} = $e^{-\frac{C}{\theta_D} f\left(\frac{\theta_D}{T}\right)\left(\frac{\sin\theta}{\lambda}\right)^2}$
- [LF] = Lorenz geometrical factor LF = $1/(2\sin^2\theta\cos\theta)$ [λ fixed]
  LF = $d^4 \sin\theta$ [TOF]; LF = $1/\sin^2 2\theta$ [plate geom., λ fixed]
- [AF] = Absorption factor AF = $e^{-A\lambda}$ [varies as 1/v] [AF very large for x-rays, small for neutrons except Gd, B, Li, Cd, …]
- [PO] = Preferred Orientation factor (compensates for non-random crystallite orientation in sample)
- [EE] = Primary extinction correction [non-uniform illumination of all reflecting planes]
- [AA*] = Complex square of scattering amplitude

Neutron Detectors

- ${}^{3}He + n \rightarrow {}^{3}H + {}^{1}H + 764$ keV



Frame overlap problem at pulsed neutron source



Hyer and Pynn 1990

Neutron Guides

- Refractive Index

$$n = 1 - \frac{\lambda^2}{2\pi} N \cdot b_{coh}$$

- Critical Angle

$$\varepsilon = \lambda \sqrt{\frac{N \cdot b_{coh}}{\pi}}$$

- Concave(!) lenses (Eskildsen et al., 1998)

Time Of Flight

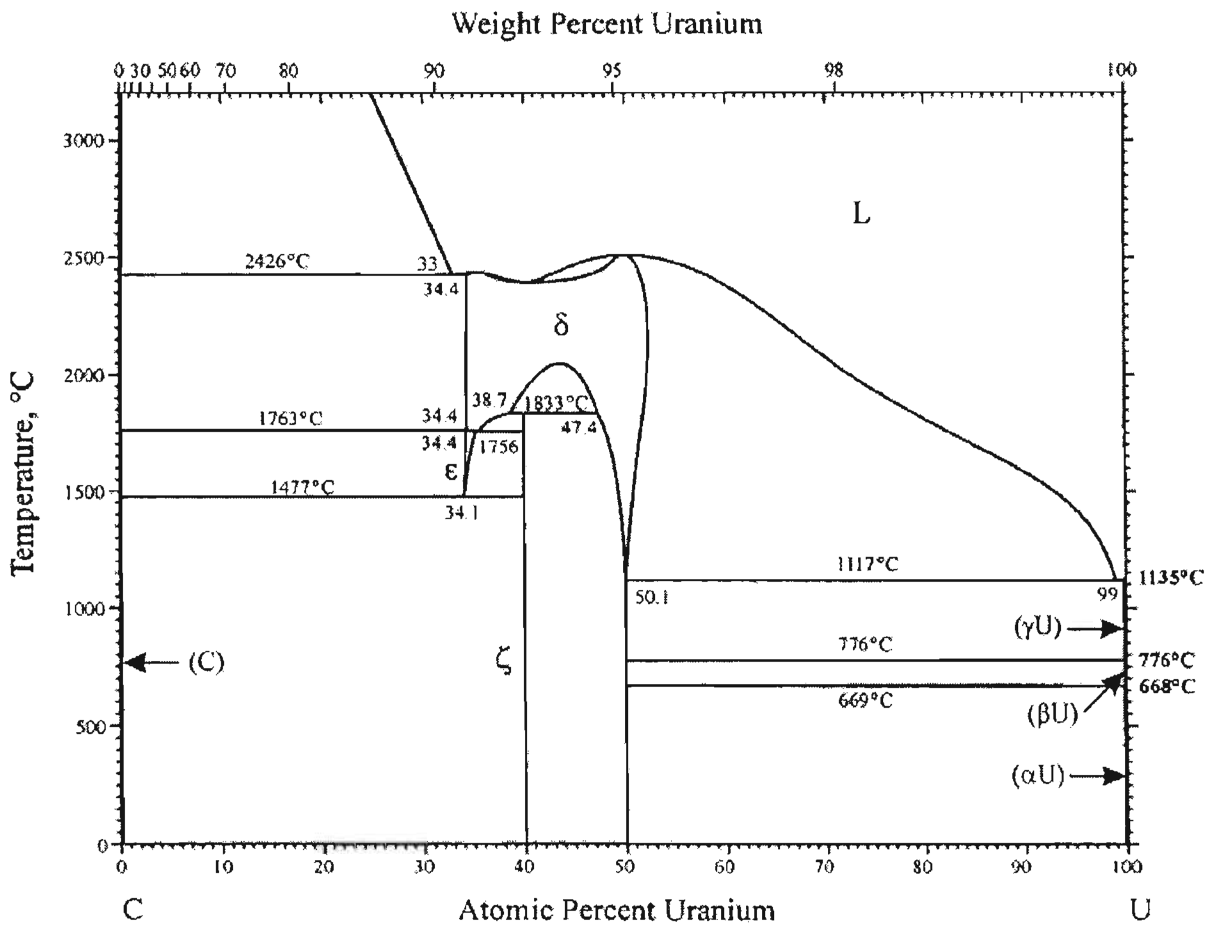
Equating quantum mechanical momentum (de Broglie relation) with classical mechanics momentum, we find:

$$p_{QM} = \hbar k = mv = p_{CM} \leftrightarrow \frac{h}{\lambda} = m\frac{L}{t} \leftrightarrow \lambda = \frac{ht}{mL}$$

With Bragg's law, this becomes

$$\lambda = 2d\sin\theta = \frac{ht}{mL} \leftrightarrow d = \underbrace{\frac{h}{2mL\sin\theta}}_{\text{const}} \cdot t$$

U-C Phase Diagram



Okamoto, 2005, JPEDAV

Health Physics

- 74 Bq/g (74 disintegrations per second per gram), the limit accepted for shipping material as “non-radioactive.”