Sample Environments

Presented at the

International School of Neutron Science and Instrumentation

5th Course: Neutrons for Chemistry and Materials Science Applications

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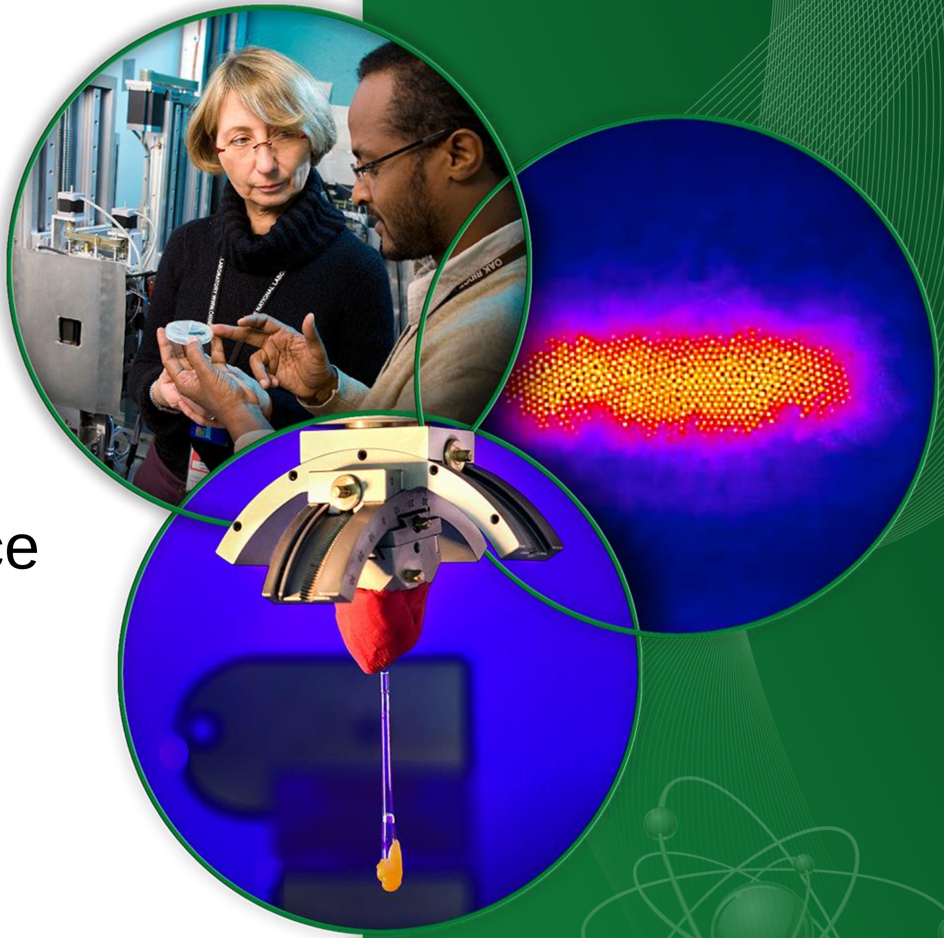
Oak Ridge National Laboratory

Erice-Sicily
July 4-13, 2018



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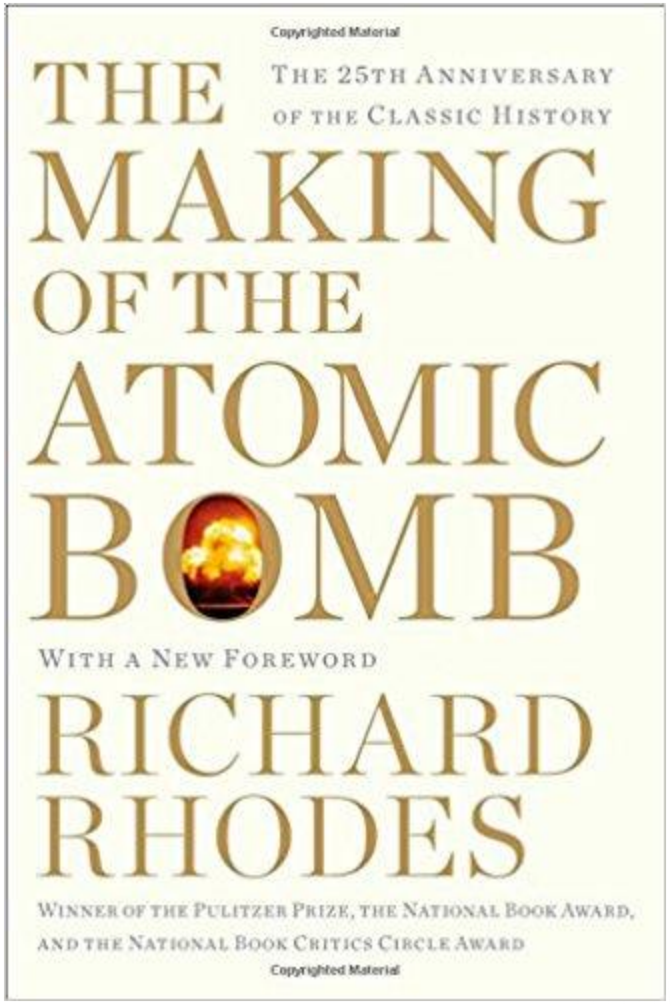
OAK RIDGE National Laboratory | HIGH FLUX ISOTOPE REACTOR | SPALLATION NEUTRON SOURCE



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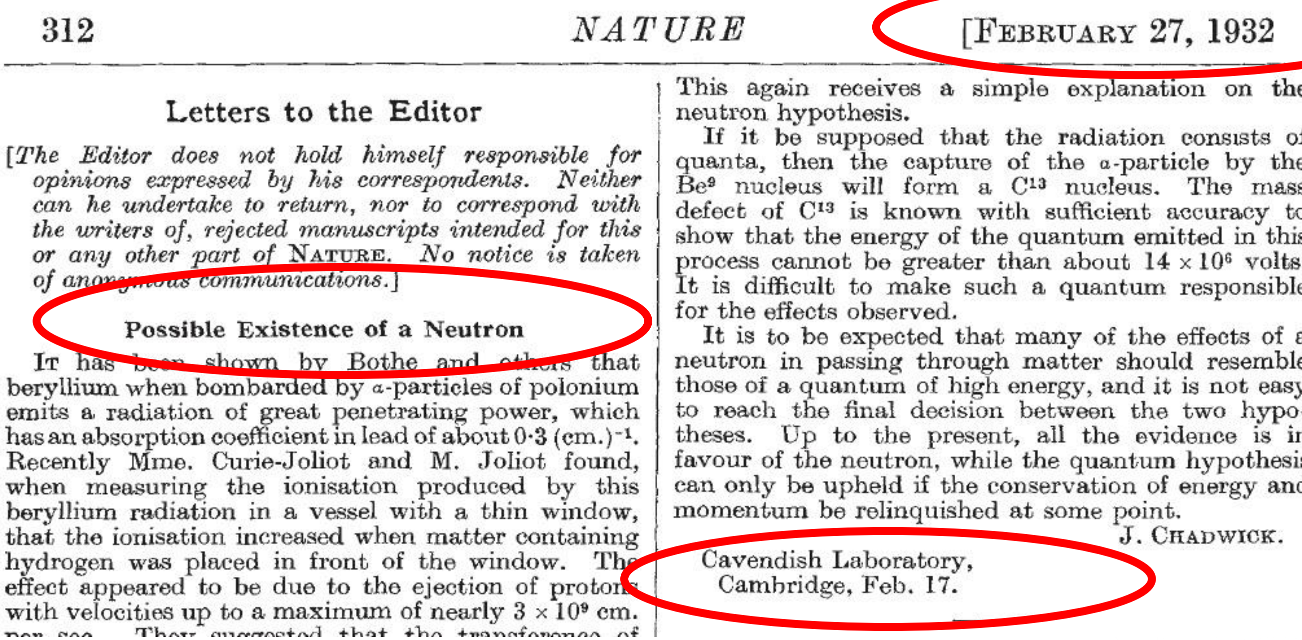


Letters to the Editor

[The Editor does not hold himself responsible for opinions expressed by his correspondents. Neither can he undertake to return, nor to correspond with the writers of, rejected manuscripts intended for this or any other part of Nature. *No notice is taken of anonymous communications.]*

Possible Existence of a Neutron

It has been shown by Bothe and others that beryllium when bombarded by α-particles of polonium emits a radiation of great penetrating power, which has an absorption coefficient in lead of about 0·3 (cm.)$^{-1}$. Recently Mme. Curie-Joliot and M. Joliot found, when measuring the ionisation produced by this beryllium radiation in a vessel with a thin window, that the ionisation increased when matter containing hydrogen was placed in front of the window. The effect appeared to be due to the ejection of protons with velocities up to a maximum of nearly 3×10^9 cm.

This again receives a simple explanation on the neutron hypothesis.

If it be supposed that the radiation consists of quanta, then the capture of the α-particle by the Be^9 nucleus will form a C^{13} nucleus. The mass defect of C^{13} is known with sufficient accuracy to show that the energy of the quantum emitted in this process cannot be greater than about 14×10^6 volts. It is difficult to make such a quantum responsible for the effects observed.

It is to be expected that many of the effects of a neutron in passing through matter should resemble those of a quantum of high energy, and it is not easy to reach the final decision between the two hypotheses. Up to the present, all the evidence is in favour of the neutron, while the quantum hypothesis can only be upheld if the conservation of energy and momentum be relinquished at some point.

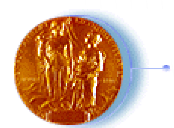
J. Chadwick.

Cavendish Laboratory,
Cambridge, Feb. 17.


Copyright Getty Images

James Chadwick

Nobel Prize in Physics 1935



Slide from Jaime Fernandez-Baca

How Did I Get Here?

450 km/hr and 3.2 km wide

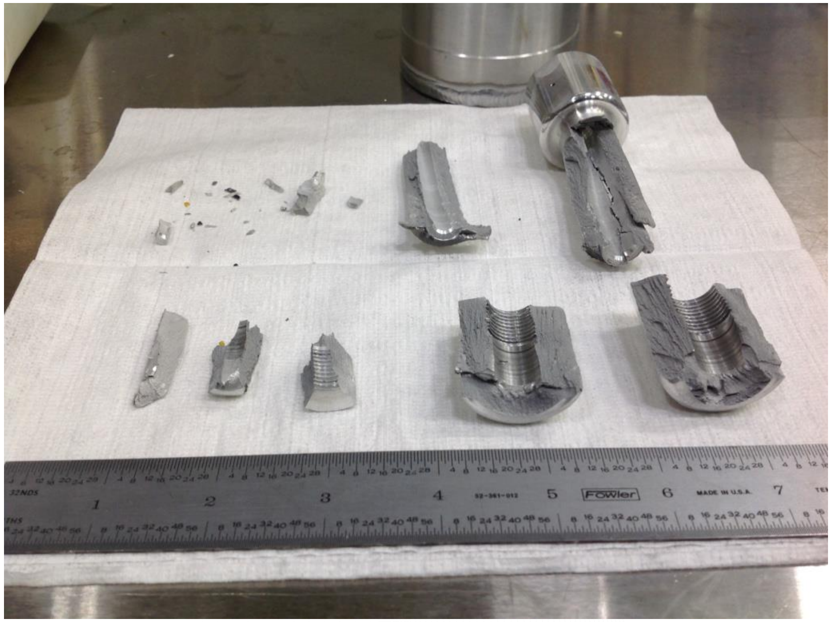
Image from Pacific Products Gallery

- BS Chemistry, minor Mathematics; Washburn University (1995)
- PhD Polymer Chemistry, University of Tennessee (2000)

Kansas area ~200,000 km^2
Kansas avg. population density ~10 people/km^2

Italy area ~300,000 km^2
Italy avg. population density ~200 people/km^2

How Did I Get Here



- ORAU post-doctoral program ORNL (2000-2004)
- ORNL staff (2004-present)

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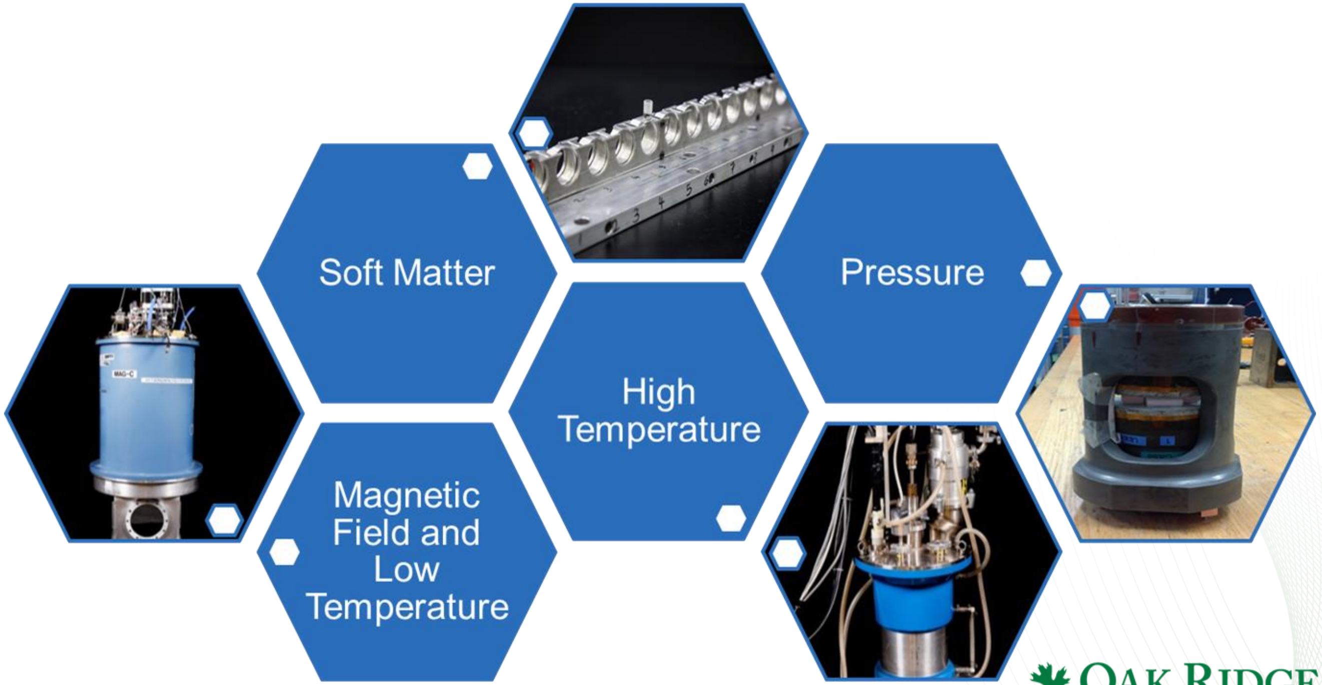
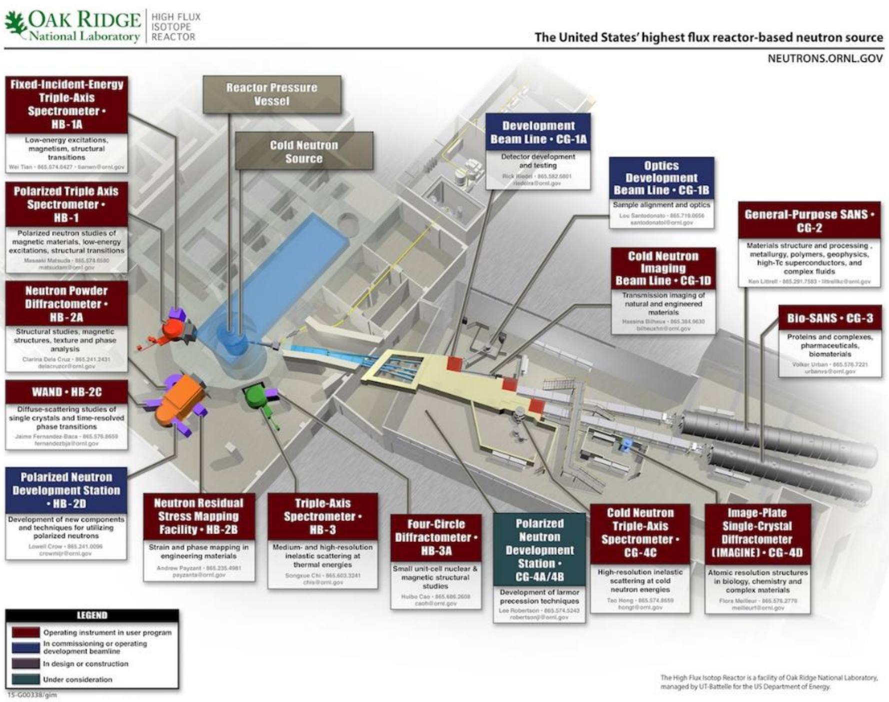
What is Sample Environment?

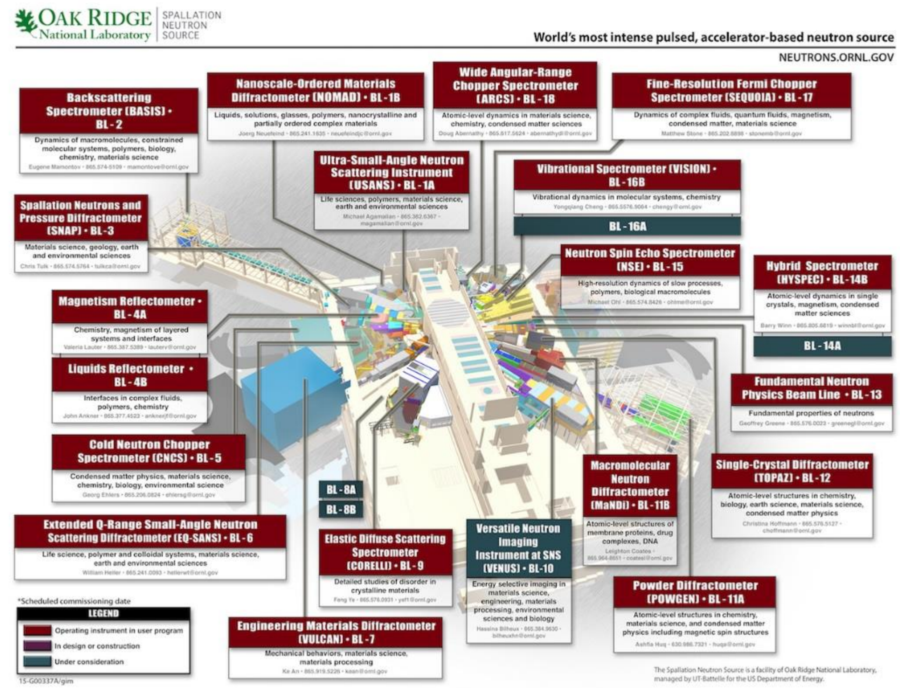
- Sample environment is an integral part of the neutron scattering experiment where neutrons are used as an investigative probe
- Equipment is used to precisely and accurately control experimental parameters such as temperature, pressure and magnetic fields



HFIR/SNS Sample Environment Group

31 Instruments in the User Program



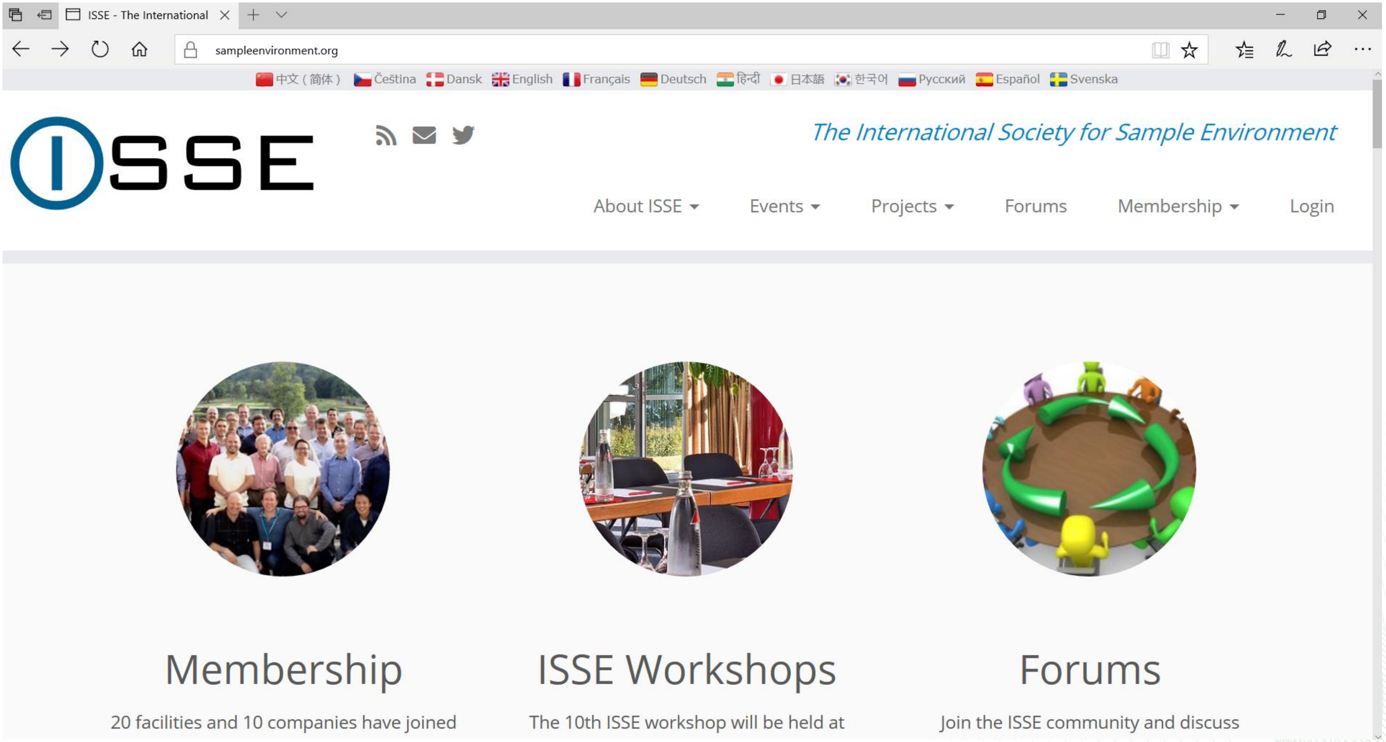


- ~1200 neutron scattering experiments per year
- ~70% experiments are supported by the Sample Environment Group

The International Society for Sample Environment (ISSE)

- The ISSE is a platform to promote scientific and technical developments of sample environment at scattering facilities

http://sampleenvironment.org



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Outline

- What to consider when designing sample environments
- Overview of commonly used equipment
  - Low Temperature
  - Magnets
  - High Throughput Sample Changers
  - Pressure
  - High Temperature

In General…

- When designing equipment, one must choose the right materials
  - Equipment must satisfy material physical properties
  - Minimize amount of material in the incident and detected neutron beam paths
  - Choose neutron-friendly materials: minimize neutron interaction (avoid incoherent background or coherent scattering from the equipment itself)
- There are always trade-offs and compromises: Yield strength of material allows for high pressures but has high neutron absorption cross section

Sample Environment Design Considerations From a Facility Point of View

- Build in as much flexibility as possible in the sample area: Experiments are continuously more complex
- Don't skimp on the utilities: Electrical power, crane coverage, compressed air, chilled water, Helium recovery
- Remember your neighbors: Stray magnetic fields, neutron background
- It is important to define requirements up front
- Project management: budget, scope and time

How to Choose the Right Materials When Designing Sample Environments

- Material physical properties must satisfy the application
- What application is the material going to be used in:
  - Vacuum (usually operate in high vacuum region, 1 x 10^{-3} torr to 1 x 10^{-7} torr)
  - High temperature (1200 °C to 1600 °C)
  - Low temperature (0.030 K to1.5 K)
  - Magnetic fields (1.0 T to 14 T)
  - High pressure (1.0 bar to 90 GPa)
  - Chemical resistant (pH < 7.0, acidic conditions)
  - Radiation field (Rad/hr in the direct beam)

How to Choose the Right Materials When Designing Sample Environments

Neutron Properties

- Neutrons have no electric charge: can penetrate into materials to be scattered by the nucleus
- Neutrons have a magnetic moment and therefore are sensitive to the magnetic field of the atoms
- Neutrons have an intrinsic energy that makes them sensitive to inter-atomic vibrations
- Scattering and absorption cross-sections depend on the isotope: isotope labelling and contrast variation take advantage of the scattering differences between Hydrogen and Deuterium

Taking Advantage of the Highly Penetrating Nature of Neutrons

- Cross section *σ*, is the effective area of a nucleus to an incident neutron
- Scattering length *b*, is a measure of the strength of the neutron-nucleus interaction
  - Related to the cross section *σ* by $\sigma = 4\pi b^2$
- Transmission = $e^{-t\sigma\eta}$
  - t = sample thickness in cm
  - σ = total neutron cross section in barns (1 barn = 10^{-24} cm^2)
  - η = number density [mass density (g/cm^3) x Avogadro's number (mol^{-1})] / [formula weight (g/mol)]
  - Neutron scattering lengths and cross sections
    - https://www.nist.gov/ncnr/planning-your-experiment/sld-periodic-table

Example: Sample Can That is 1 mm Thick Aluminum

- Mass density = 2.70 g/cm^3
- Formula weight = 26.982 g/mol
- Avogadro's number = 6.02×10^{23} mol^{-1}
- t = 0.1 cm
- $\sigma = 1.73 \times 10^{-24}$ cm^2 (v = 2200 m/s, λ = 1.8 Å)
- Transmission = 0.99 (99%)

✓Neutron Properties

✓Physical Properties (vacuum applications)

Neutron Friendly Materials For Neutron Powder Diffraction Experiments

- One does not want the sample environment equipment to add to the neutron signal
- Choose null-scattering materials (minimize coherent scattering)
- Vanadium
  - bound coherent scattering length -0.3824 fm
  - Can use up to 1200 °C (melting point 1910 °C, but can recrystallize and embrittle at lower temperatures)

✓Neutron Properties

✓Physical Properties (high temperature applications)

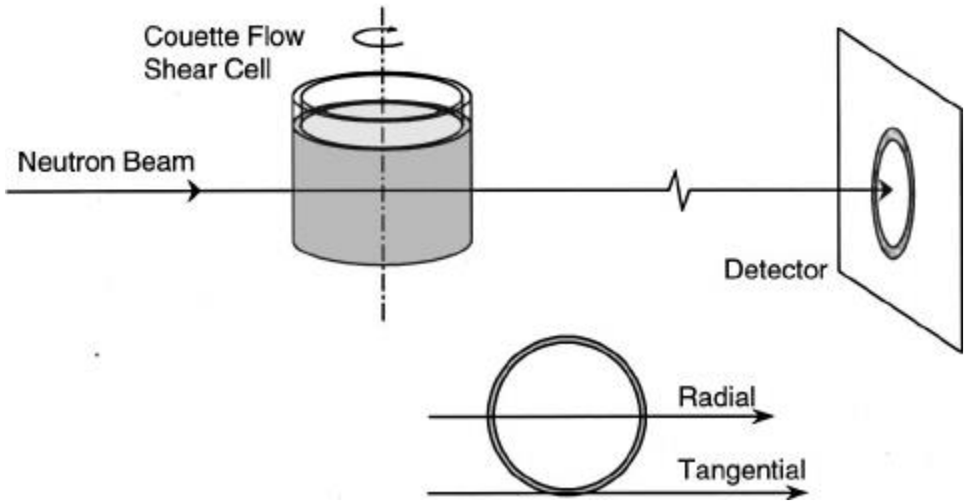
Neutron Friendly Materials For Neutron Powder Diffraction Experiments

- One does not want the sample environment equipment to add to the neutron signal
- Choose null-scattering materials (minimize coherent scattering)
- TiZr
  - Ti bound coherent scattering length -3.438 fm
  - Zr bound coherent scattering length 7.16 fm
  - Alloy typically 66% Ti and 34% Zr
  - Applied pressures ~1300 bar

✓Neutron Properties

✓Physical Properties (pressure applications)

Many Factors Contribute to the Detected Neutron Signal on a SANS Experiment



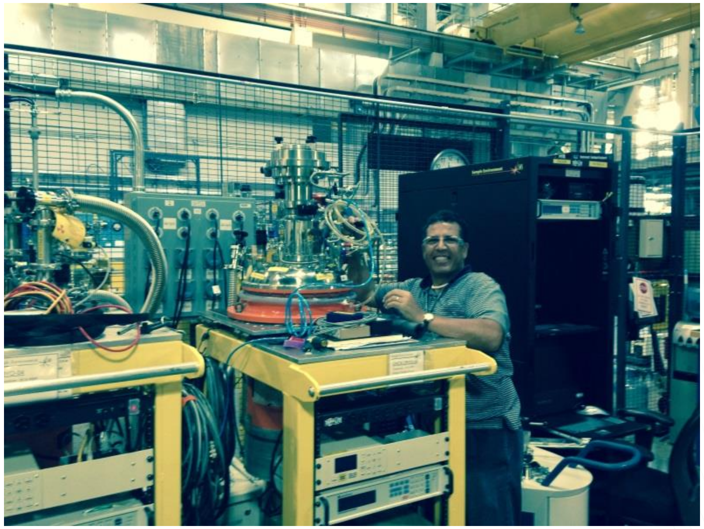
K. Krishnan et.al.; **Journal of** Rheology; 46(2); March 2002

- $I_{cor} = I_{sam} – T \times I_{emp} – (1 – T) \times I_{blk}$
  - I_{cor} = corrected intensity
  - I_{sam} = intensity from sample
  - I_{emp} = intensity from empty cell
  - I_{blk} = blocked beam
  - T = transmission of sample
  - Background signal from sample environment adds scattering

Low Temperature

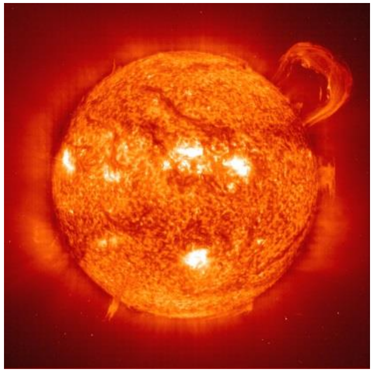
Low Temperature Equipment:

- Closed cycle refrigerators 4 K - 300 K
- Liquid Helium Cryostats 1.5 K - 300 K
- 3He inserts 0.3 K - 300 K
- Dilution refrigeration inserts 0.03 K - 300 K



What Do We Mean by Low Temperature?

Sun~ 6,000 K



X 20

Room temp ~ 300K

X 30,000

D/R system~0.010 K

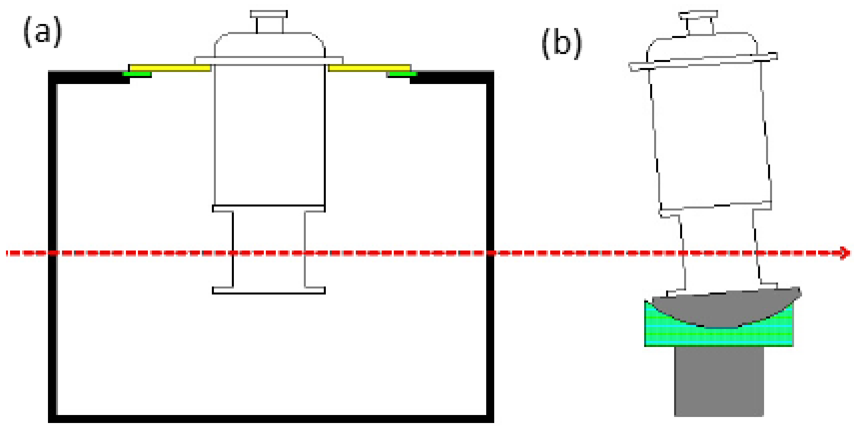
- Mass of Sun: ~ 2 x 10^{33} g
- Mass of typical neutron scattering experiment samples: milligrams-grams

Low Temperature

Helium Closed Cycle Refrigerators (CCR)

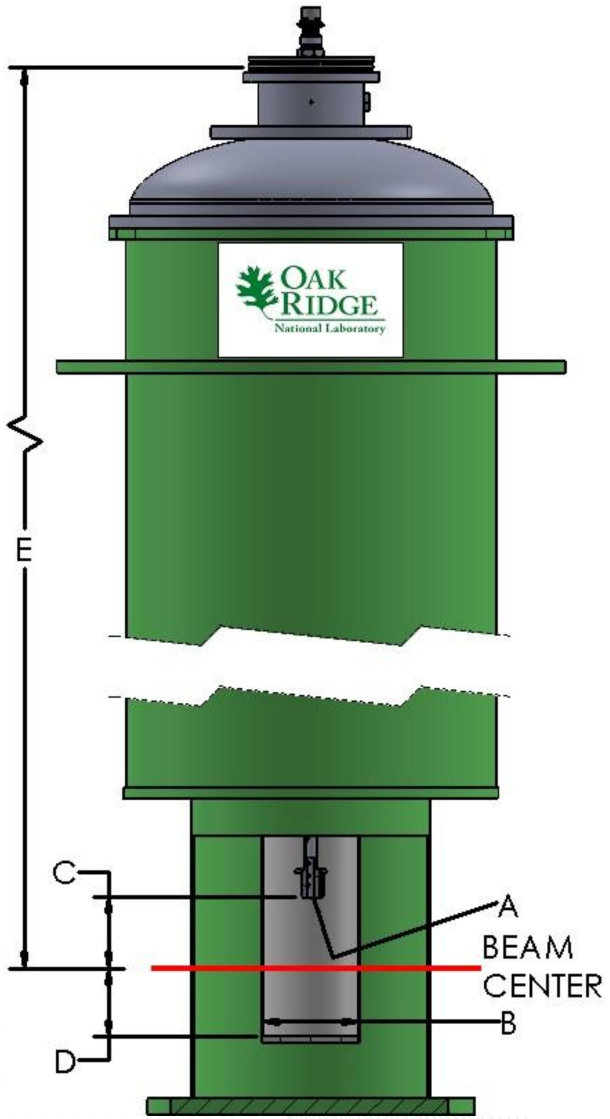
- Temperature range 4.0 K - 300 K
- Direct cooling: Sample mounted directly to the cold-head, rapid cooling (40 min), entire CCR must be removed from instrument vacuum boundary to change sample
- Indirect cooling: Sample is held in a helium exchange gas chamber, chamber is cooled by 2nd stage, sample can be changed without warming up entire CCR

Low Temperature

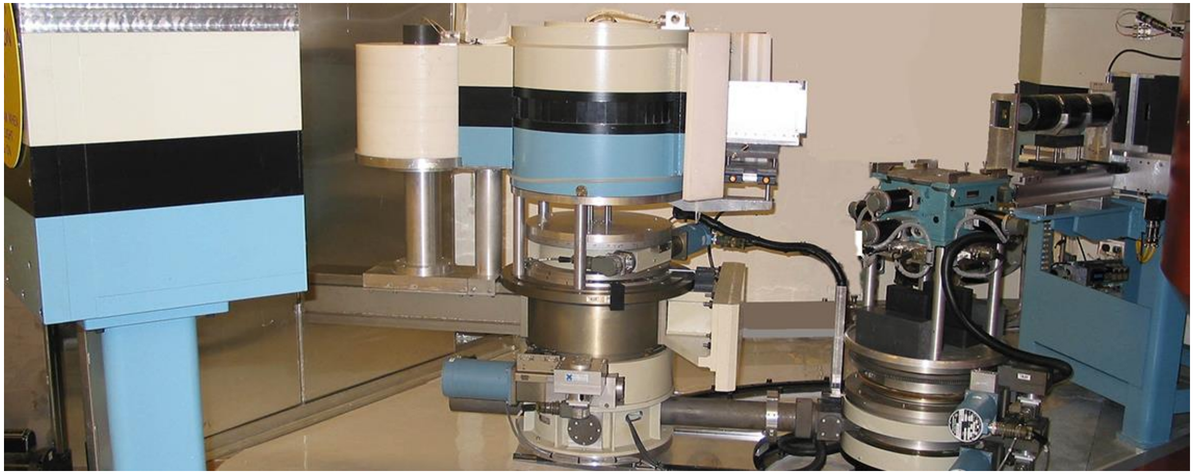


Closed Cycle Refrigerators (CCR)

- Flange mount or tail mount
- Flange diameter defines maximum diameter allowed (700 mm typical)
- Define flange: bolt holes, vacuum boundary, etc.
- Tail mount: distance from beam center to bottom of tail (50 mm typical)
- Outer Vacuum Chamber (tails) diameter 350 mm
- Distance from stick flange to beam center 1092 mm
- 100 mm bore



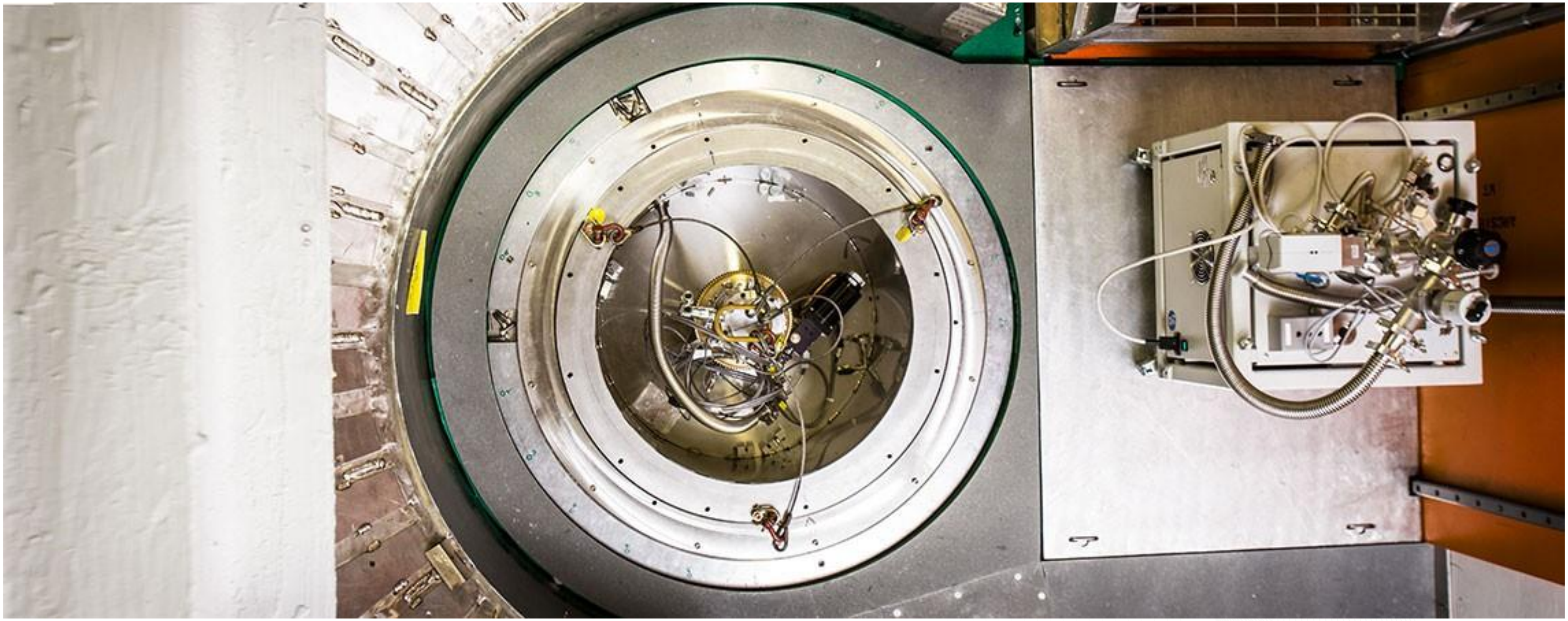
Tail Mounted



HB1-A Beamline at HFIR

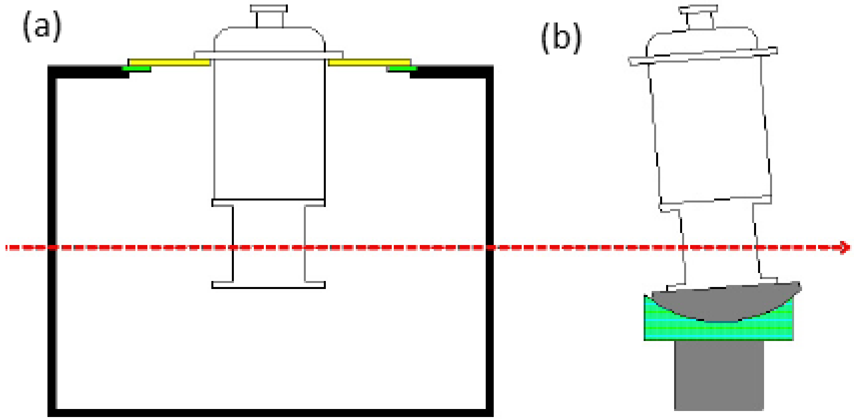
CG-2 Beamline at HFIR

Flange Mounted



ARS Beamline at SNS

Low Temperature



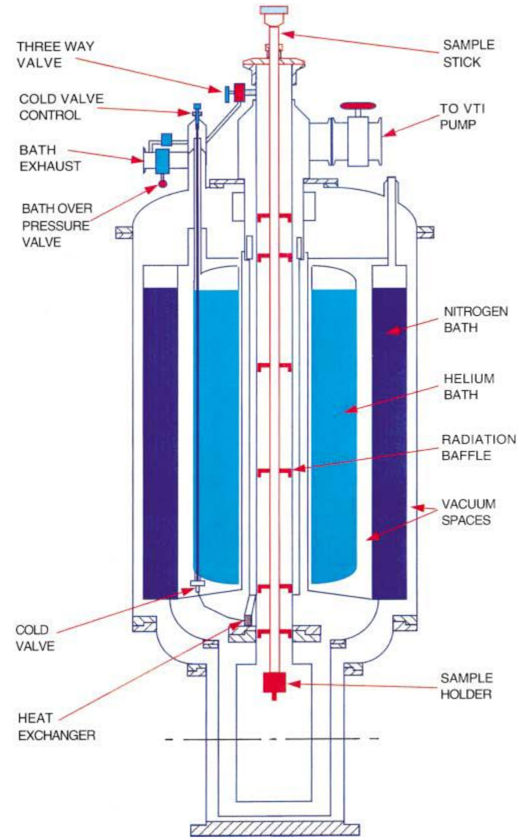


Image from I.F. Bailey; **A Review of Sample Environments in Neutron Scattering;** Z. Kristallogr. **218** (2003)

Liquid Helium Cryostats

- Temperature range 1.5 K - 300 K
- Flange mount or tail mount
- Sample is held in a helium exchange gas chamber (IVC) at a helium pressure of 10 mbar
- Liquid helium exhausts through a heat exchanger connected to IVC
- Exhausting cold helium gas passes through entire length of annular space cooling the IVC
- Be careful not to use too much helium exchange gas!

Alfred Leitner on Superfluid Helium

https://www.youtube.com/watch?v=sKOlfR5OcB4

Low Temperature

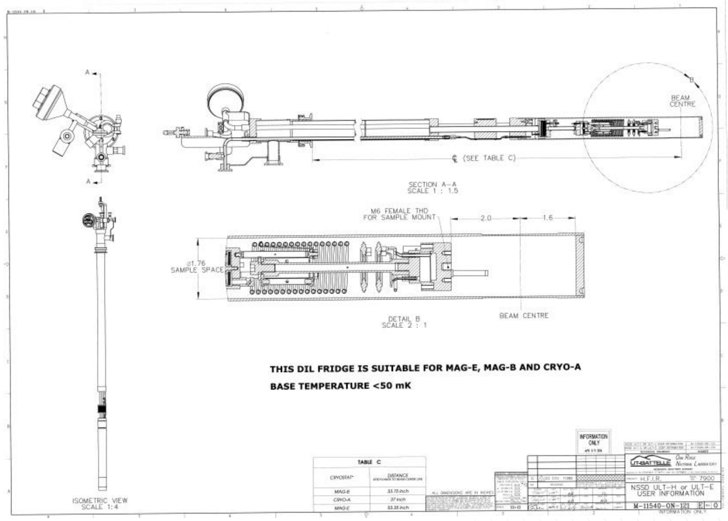
3He Insert

- Temperature range 0.3 K - 80 K (up to 300 K with VTI)
- Achieve a base temperature less than 0.3 K for more than 40 hours
- Maintain a base temperature less than 0.35 K for more than 6 hours with a 50 µW heat load
- Temperature stability of ± 0.003 K below 1.2 K

Dilution Refrigeration Insert

- Temperature range 0.03 K - 1.5 K (up to 300 K with VTI)
- Cooling power at least 40 µW at 0.1 K

image from Oxford Instruments



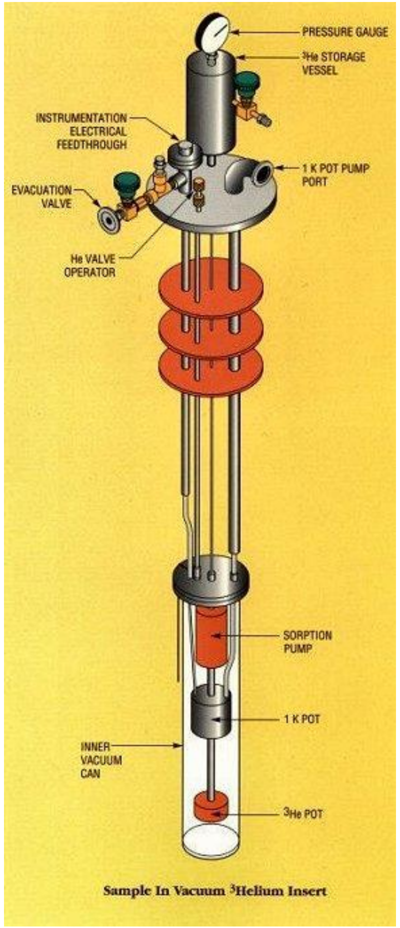
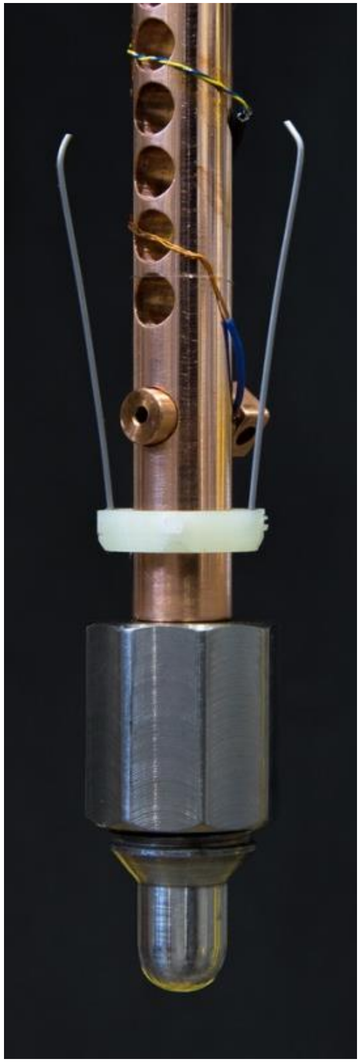


image from Janis Research Company, Inc.

Sensors for Low Temperature



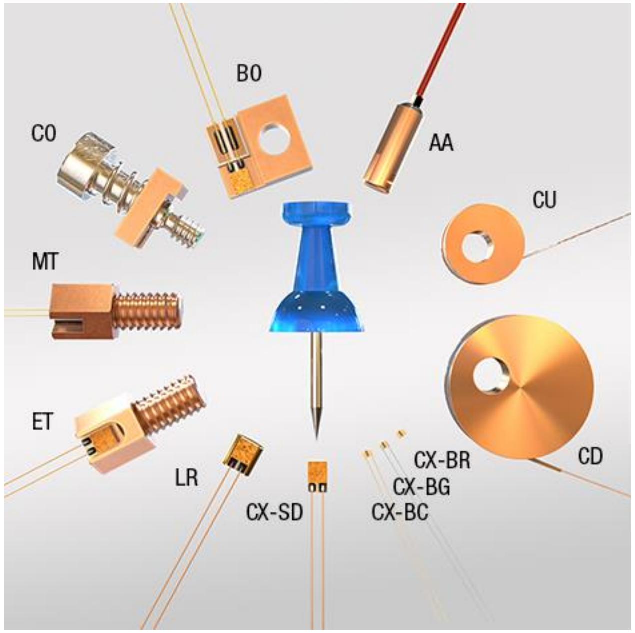


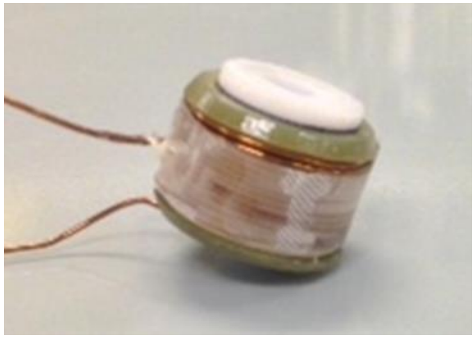
Image from Lake Shore Cryotronics

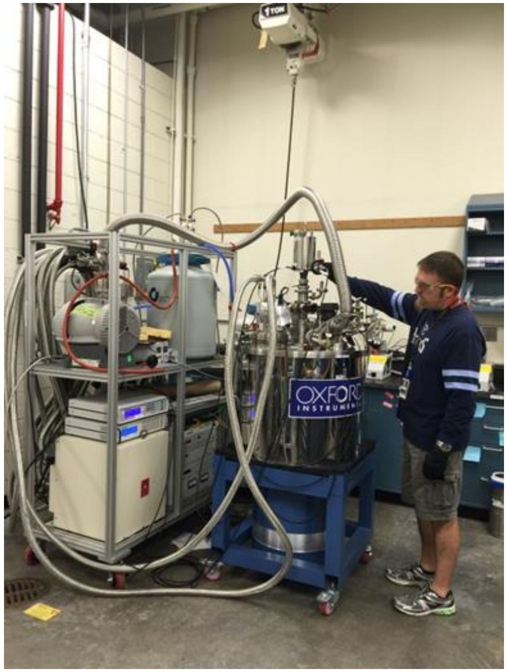
- **Cernox®**
- Temp. range 0.10 K to 420 K
- Good in a radiation field
- Good in a magnetic field at temperatures above 1 K
- **Ruthenium Oxide (Rox™)**
- Temp. range 0.010 K to 40 K
- Good in a radiation field
- Good in a magnetic field at temperatures below 1 K

Magnets

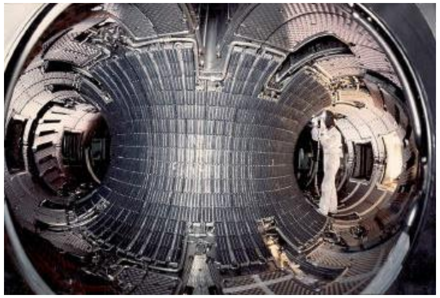
Magnetic Field Equipment:

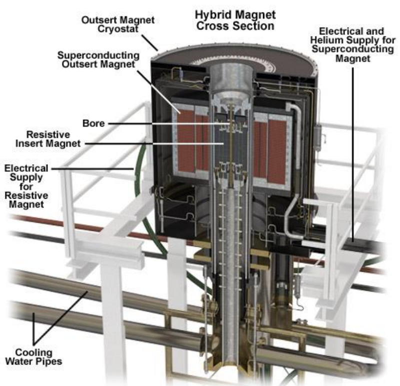
- 0.5-3 T electromagnet: specialized for Reflectometry or SANS
- 3-11 T superconducting cryomagnet, horizontal field
- 5-11 T superconducting cryomagnet, vertical field, symmetric or asymmetric
- 30 T pulsed magnet





Examples of Magnetic Fields





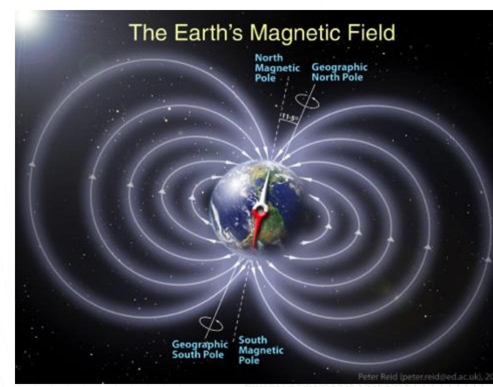
[T (Tesla)]

	Tesla	
10^{13} G	1 GT (10^9)	Neutron Stars
	1 kT (10^3)	
1 MG	100 T	Pulsed Fields (100 T); 10 ms - 1 ms
		"DC" Fields (Hybrid: 45 T, SC: 32 T)
	10 T	SCM for thermonuclear reactors (~ 10 T)
		SCM for maglev, generators, motors (~ 5 T)
	1 T	Permanent Magnets (~ 1.4 T)
		SCM for MRI (~ 1 T)
1 kG	100 mT	
	10 mT	
	1 mT (10^{-3})	
1 G	100 μT	
	10 μT	
	1 μT(10^{-6})	
1 mG	100 nT	
	10 nT	
	1 nT (10^{-9})	
1 μG	100 pT	Heart (~100 pT)
	10 pT	
	1 pT (10^{-12})	Brain (~500 fT)
1 nG	100 fT	
	10 fT	
	1 fT (10^{-15})	Lower limit of SQUID sensitivity

Field Produced by:
Structural Steel Beams (30 G = 3 mT)
Large Steel Equipment (10 G = 1 mT)

Motor Vehicles, Elevators (1 G = 0.1 μT)
Earth (25 μT - 65 μT)

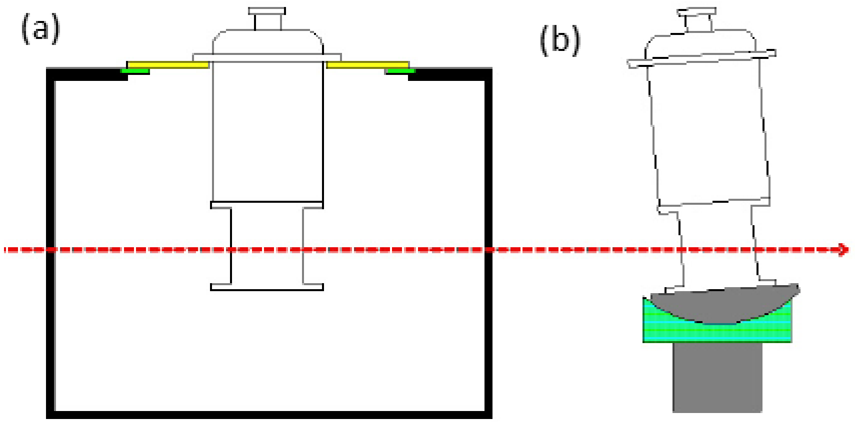
Objects Affected:
50 G: Magnet Power Supplies
20 G: Magnetic Storage Media
10 G: Computers, Watches and Clocks, Credit Cards
5 G: Pacemakers, Cathode Ray Tubes, Photomultiplier Tubes, EPA Limit for occupational exposure
1 G: Electron Mcroscopes, RGAs



Magnets

Overall Physical Dimensions and Weight

- Flange mount or tail mount
- Flange diameter defines maximum diameter allowed (700 mm typical)
- Define flange: bolt holes, vacuum boundary, etc.
- Tail mount: distance from beam center to bottom of tail
- Maximum overall height (2200 mm typical)
  - Crane access: below the hook to mounting surface
  - Movement around the facility: through doors, etc.
- Total weight (including cryogens 450-1000 kg) not to exceed crane capacity



Magnets

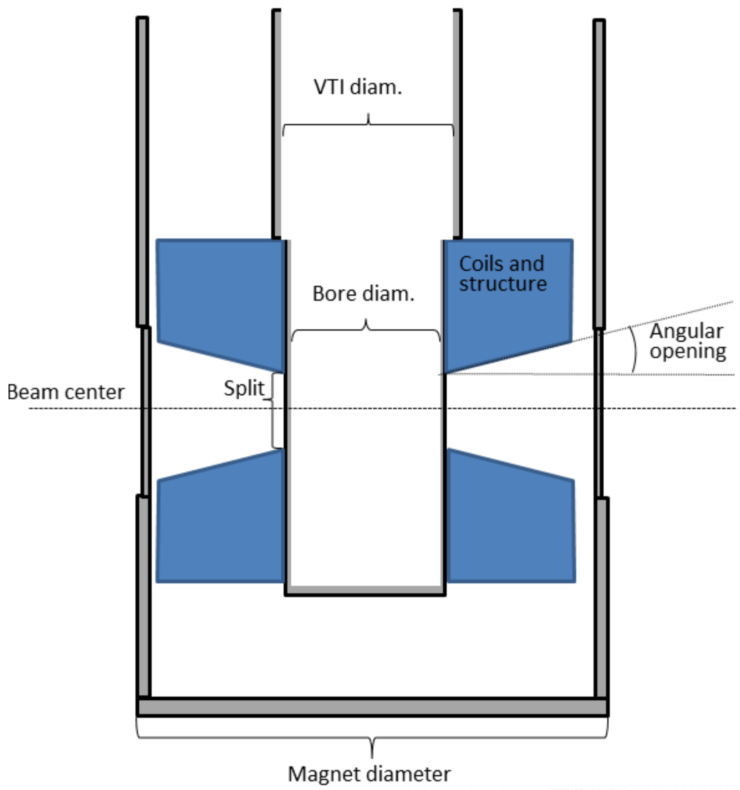
Real Estate and Utilities

- Ancillary equipment such as power supply, Helium recondensing equipment, vacuum pumps, etc. can take up several square meters of space around the instrument
- Routing of vacuum lines, power and signal cables can be a little tricky
- Electrical power (U.S.):
  - 60 Hz at 110 V and 20 A for instrumentation
  - 60 Hz at 208 V and 30 A for power supply
  - 60 Hz at 480 V and 30 A for cold head compressor
- Chilled water

Magnets

Split Pair Magnet Dimensions

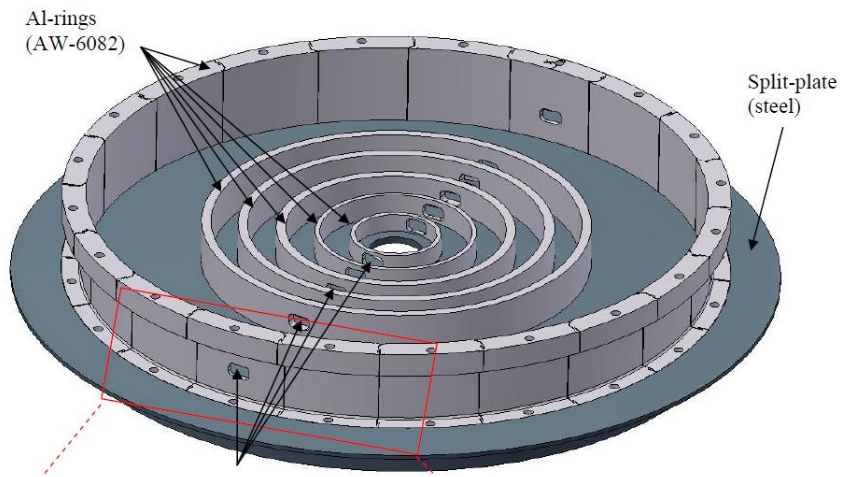
- Magnet diameter increases as magnetic field increases
  - Roughly 600 mm for 14 T uncompensated
  - Diameter can double for compensated
- Angular opening should be chosen to work with detector geometry (a few degrees is typical)
- Bore diameter should be matched to beam width available and low temperature inserts (VTI, DR; 50 mm typical)
- Split is the vertical height available for neutron beam between the magnet poles

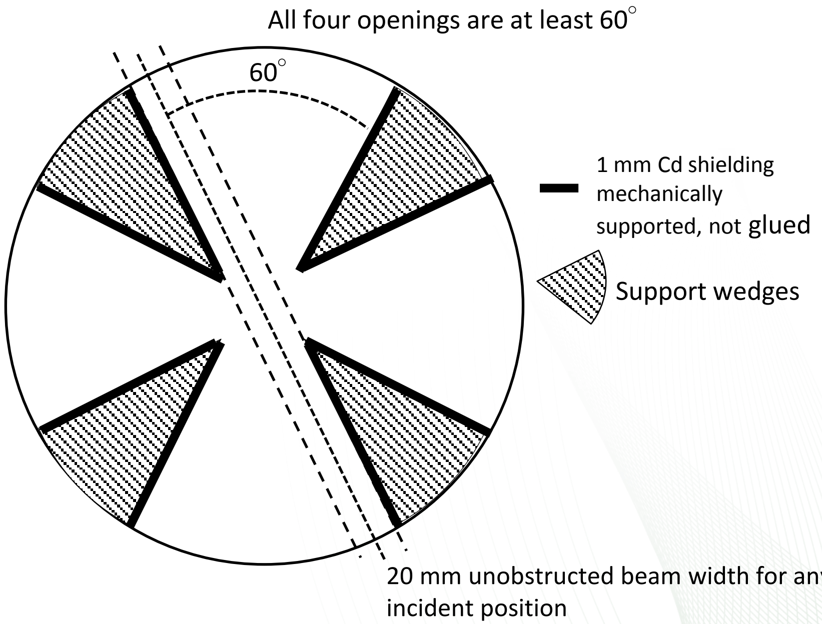


Magnets

Split Pair Magnet Support

- Provide structural support for the weight plus force from magnetic field
- Goal is to minimize the amount of Aluminum in the incident and scattered neutron beams
- Positioning of wedges to take up "dark angle" (where there is no detector coverage)

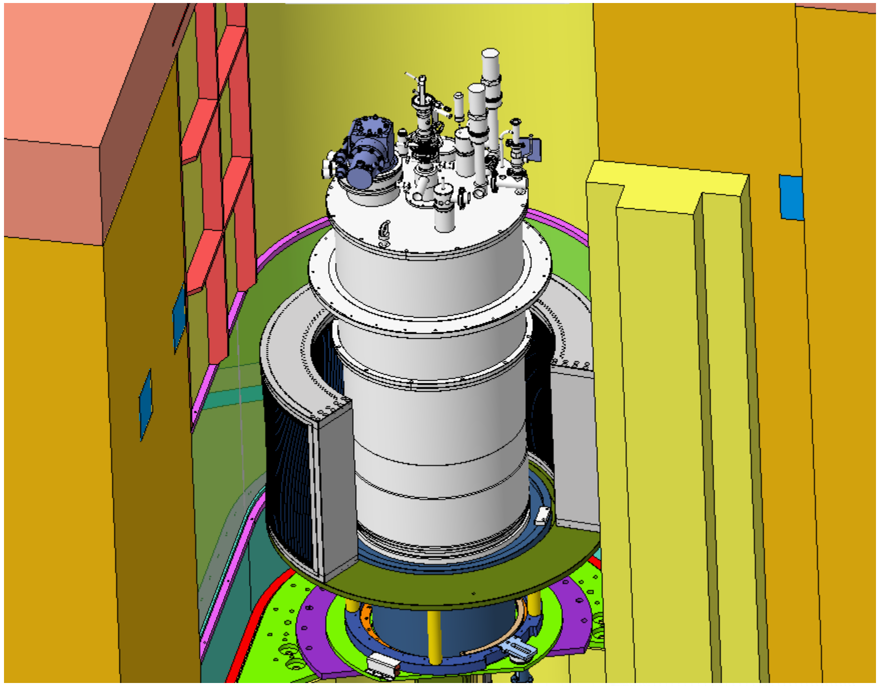


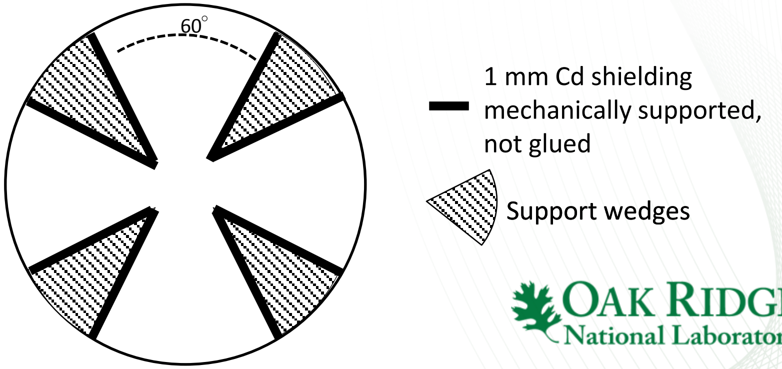


Magnets

14 T Vertical Field Magnet for SNS

- Working with Oxford Instruments
- 14 T symmetric and 12 T asymmetric
- Designed for CNCS (BL-5), HYSPEC (BL-14B), CORELLI (BL-9), ARCS (BL-18) and SEQUOIA (BL-17)
- $\pm$ 10° out of plane covers all of HYSPEC, ~ 60% of CNCS detectors and middle banks of ARCS and SEQUIOA detectors
- Total path-length of Aluminum < 6.5 mm
  - 16 T FatSam magnet path-length of Aluminum: 12.25 mm in direct neutron beam and 79.25 mm in scattered neutron beam





Magnets

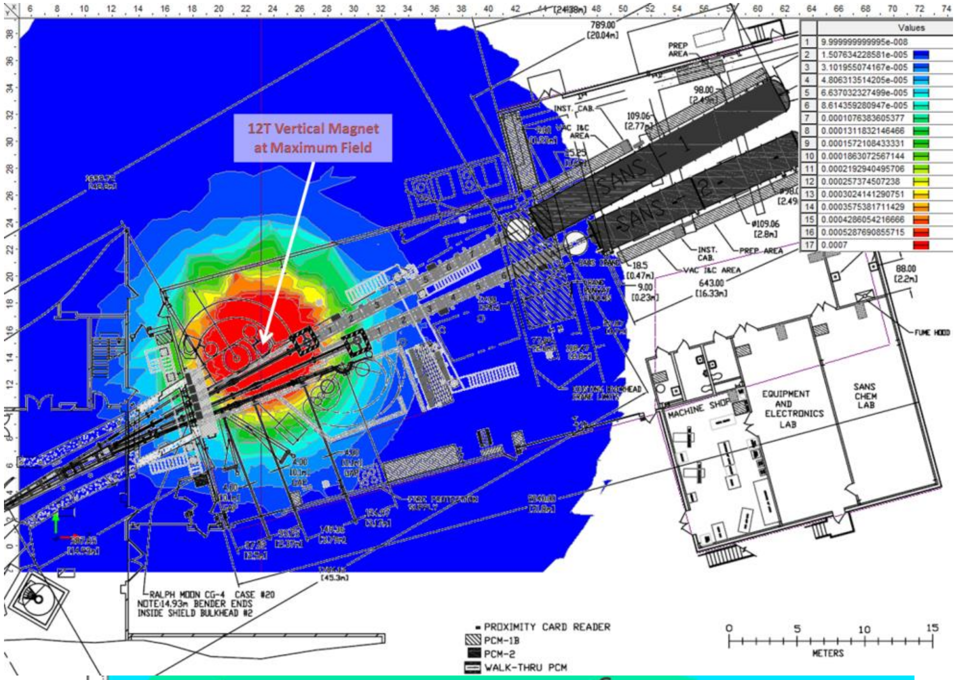
26 T High Field Magnet at Helmholtz Zentrum Berlin (HZB)

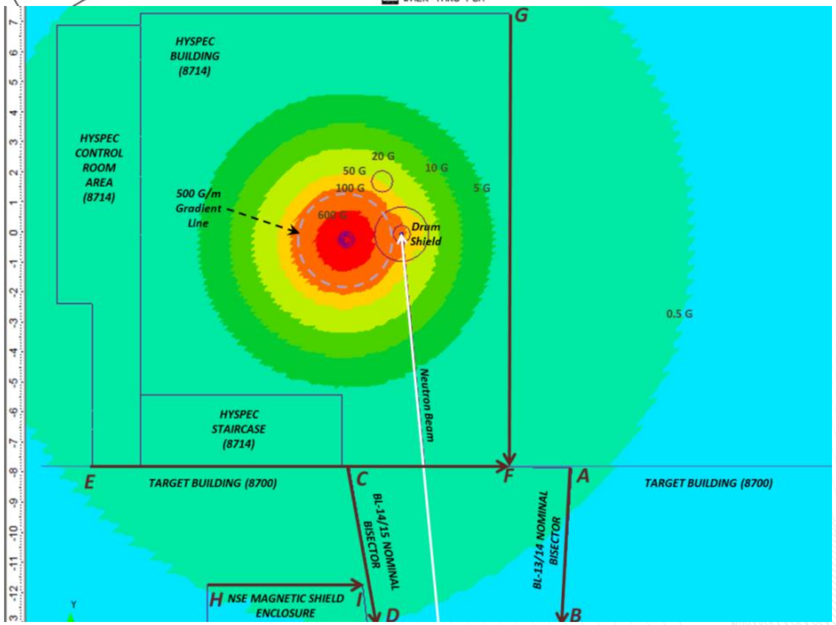
image from: https://www.helmholtz-berlin.de/quellen/ber/hfm/hfm/index_en.html
Research Company, Inc.

Magnets

Stray Fields

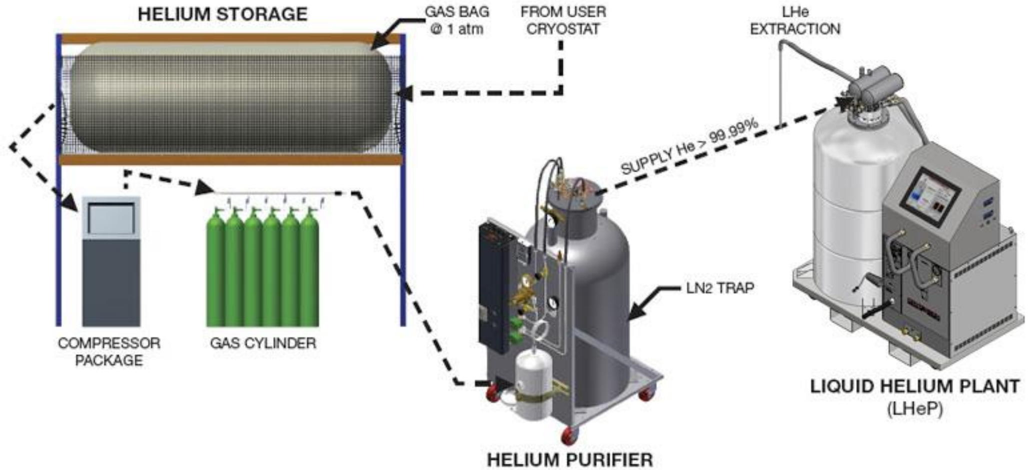
- Stray field plots usually provided by vendor
- Intensity decreases by the cubed of the distance
- What impact in the immediate area
    - 1 gauss: vacuum gauges, etc.
    - 5 gauss: pacemakers
    - 10 gauss: computers
    - 20 gauss: magnetic storage media
    - 50 gauss: magnet power supply, stepper motors

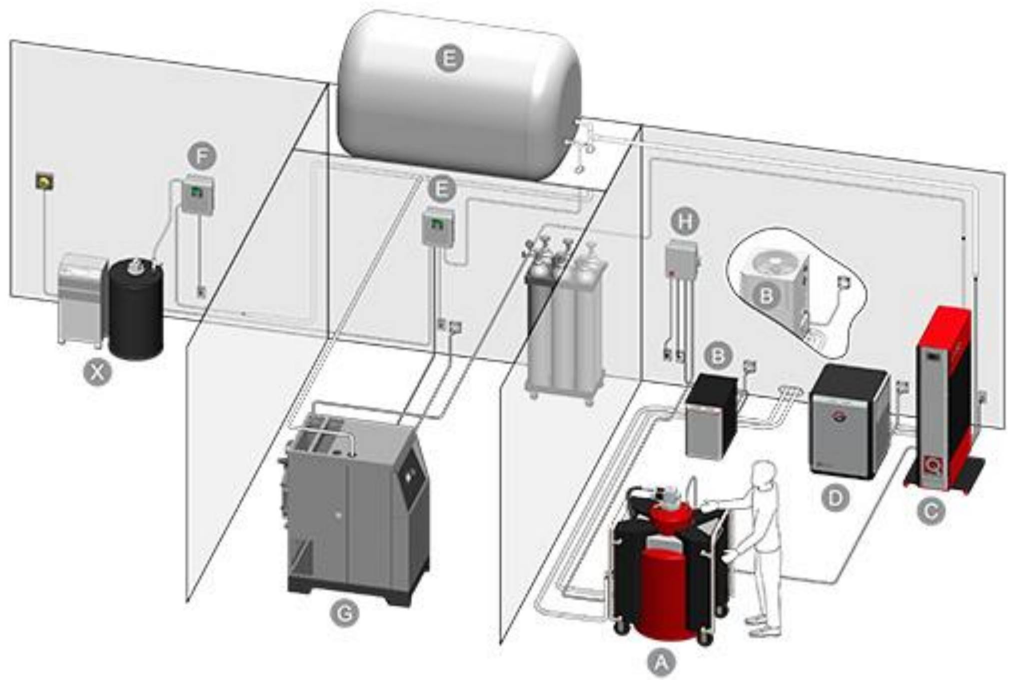




Cryogen Free?

- In the U.S., helium supply is not a problem **(at present)**
- Liquid Helium usage about 15,000 Liters per year
- At a cost of $14.25 per Liter, $213,750 total expense
- Liquid Helium recovery system capital cost of $1,400,000
- 10 years for recovery of capital and operating costs
- Solar panels to provide power?

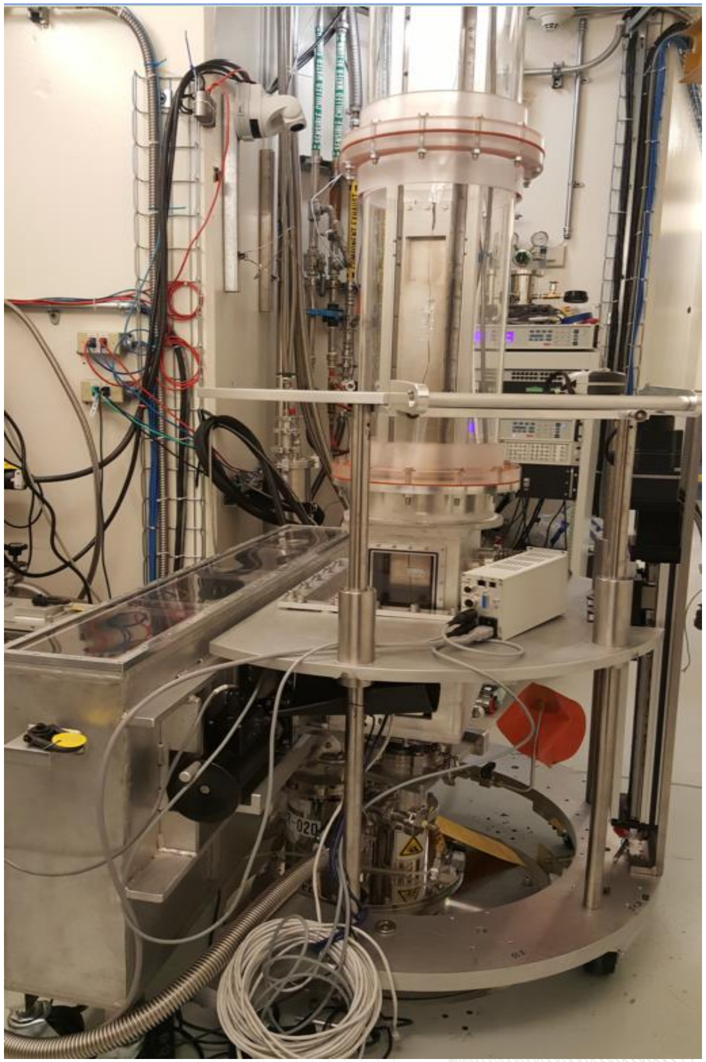




High-Throughput Sample Changers

VISION (BL-16B) Sample Changer

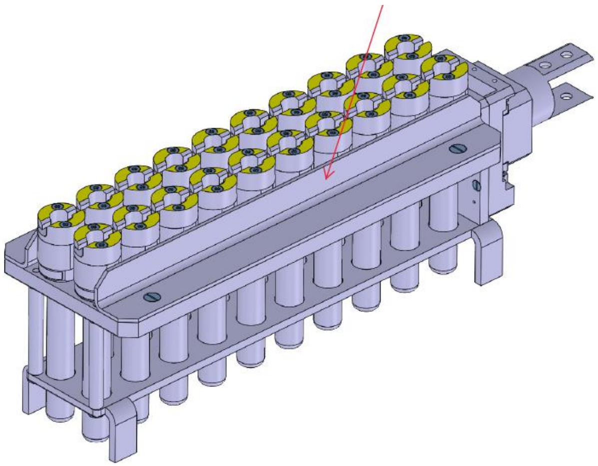
- Complete initial cool-down in 4 hours
- Sample temperature <10 K
- Sample changes every 25 minutes
- 48 sample load capacity
- Up to 5 samples pre-cooled
- 7 mbar exchange gas

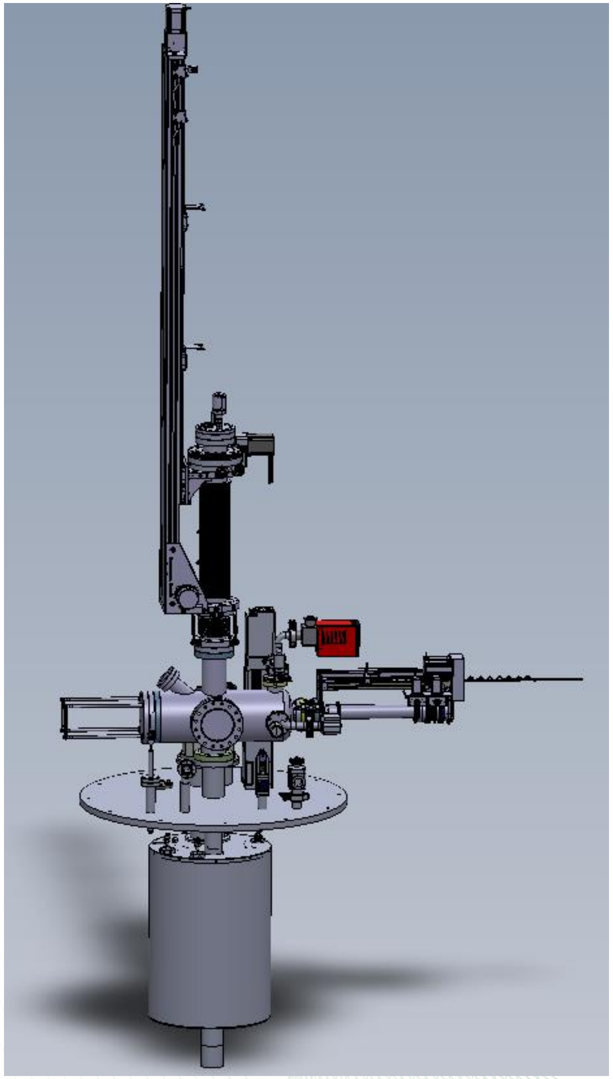


High-Throughput Sample Changers

NOMAD (BL-1B) Sample Changer

- Working with Janis Research Company, LLC
- 20 Sample capacity
- Temperature range 10 K – 800 K
- Auto exchange gas system
- Low background Vanadium tails





Pressure

Pressure Equipment:

- 400 - 1300 bar V and TiZr cells for diffraction
- 6 kbar and 1.5 - 300 K Helium gas cells
- 4 GPa and 3.5 K Palm Cubic Anvil
- 1 - 3 GPa and 0.3 - 1.5 K Clamp cells for inelastic
- 10 - 40 GPa and 15 - 300 K Diamond Anvil Cells for diffraction

What Is Considered High Pressure?

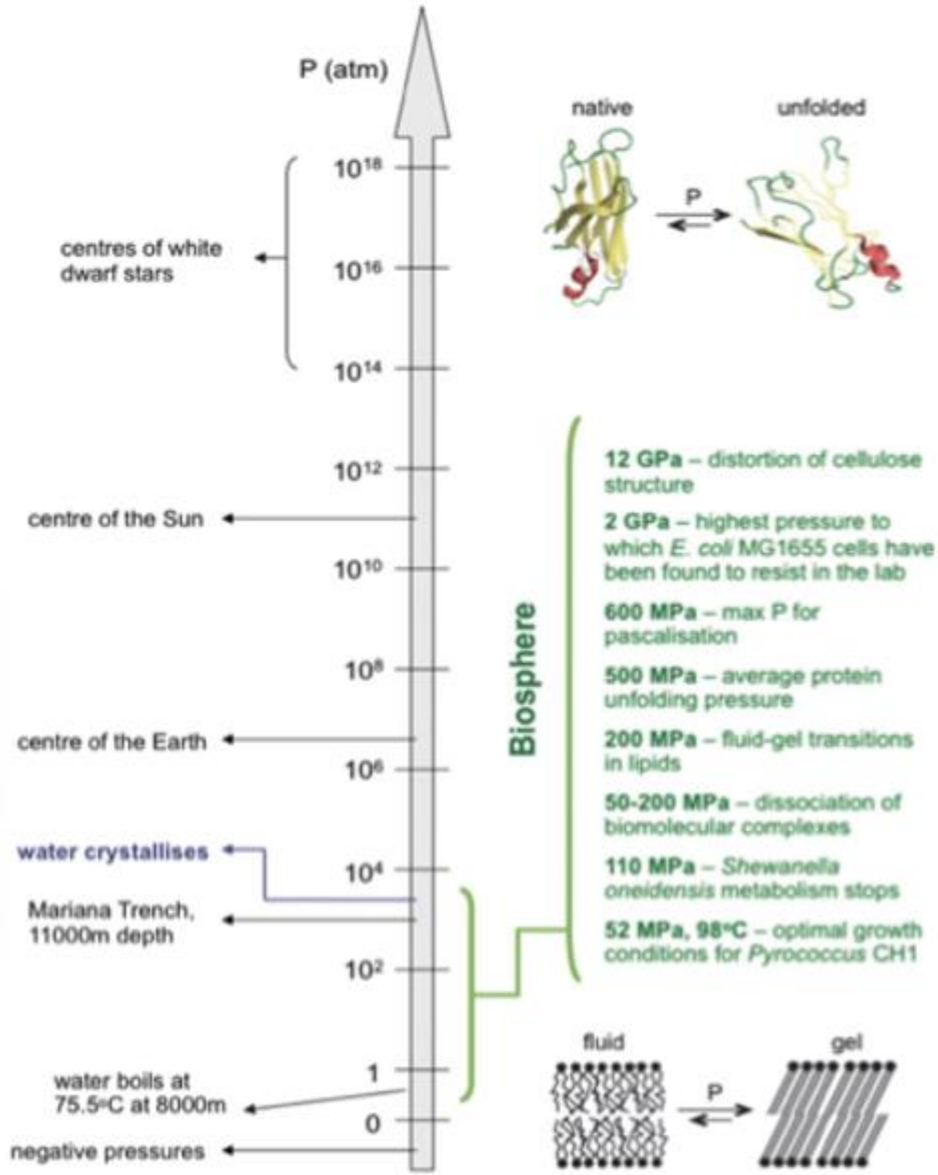
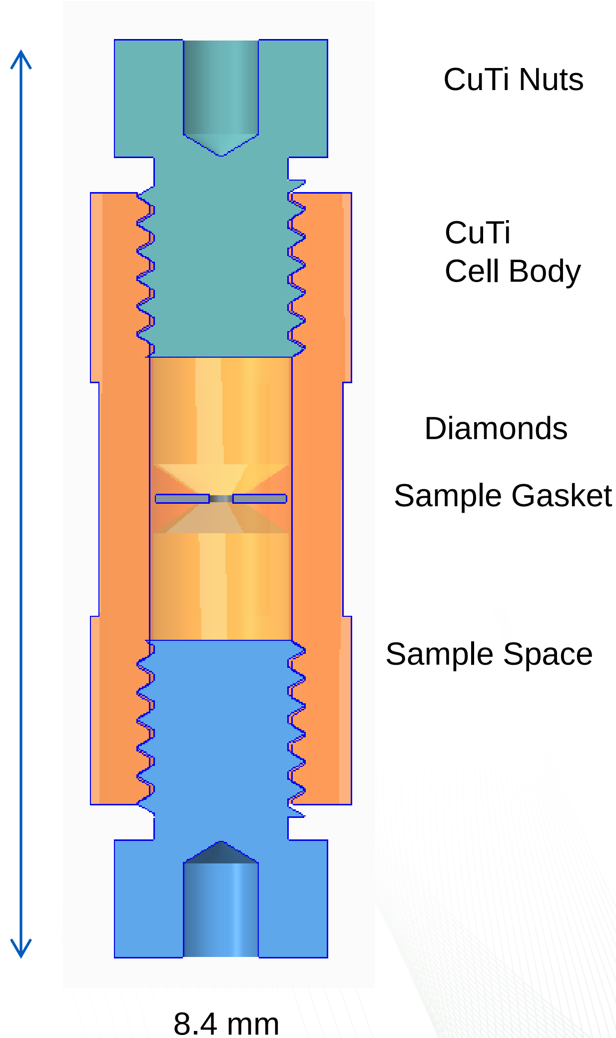


Image from: Meersman & McMillan, Chem. Comm. 2014, 50, 766-765.

Pressure

Clamp Cell

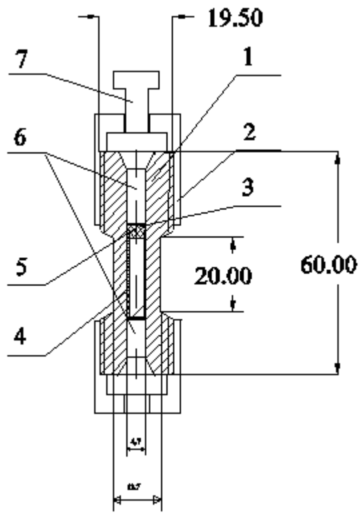
- Max. Load : 3.5 Tons for a wall thickness of 2mm.
- Sample Space: max. 0.3 mm diameter and 0.2 mm height.
- Pressure will depend on the culet size of the diamonds
- For a culet size of 1.7mm, Max Pressure approx.: 10GPa
- Disadvantages: Peaks from the Material
- Choices of Material for High Pressure Cells:
  - NiCrAl yield strength 2 GPa
  - CuBe yield strength 1.2 GPa
  - CuTi yield strength 1.2 GPa
  - Maraging Steel yield strength 0.8 GPa
  - TiZr yield strength 0.7 GPa



Pressure

Clamp Cell Design for Inelastic Scattering

- 500 mm^3 sample volume for inelastic scattering
- Fit in bore of magnet
- Non-magnetic material
- High thermal conductivity material to cool below 4 K
- Cell components have similar coefficients of thermal expansion
- Yield strength of material (NiCrAl) sufficient to pressurize up to 2.0 GPa
- Compatible with our dilution refrigeration insert and 8 T magnet at SNS
- Successful experiments at 2 GPa, 0.070 K and 7 T



1- Body-nonmagnetic HNU (Ni-Cr-Al) alloy
2- Clamping nut-nonmagnetic Ti alloy
3- Extrusion ring-CuBe alloy
4- Capsule for sample (teflon or lead)
5- Capsule cap (teflon or lead)
6- Piston of a cell- nonmagnetic HNU alloy
7- Piston for pushing out the sample and for generating pressure- nonmagnetic HNU alloy

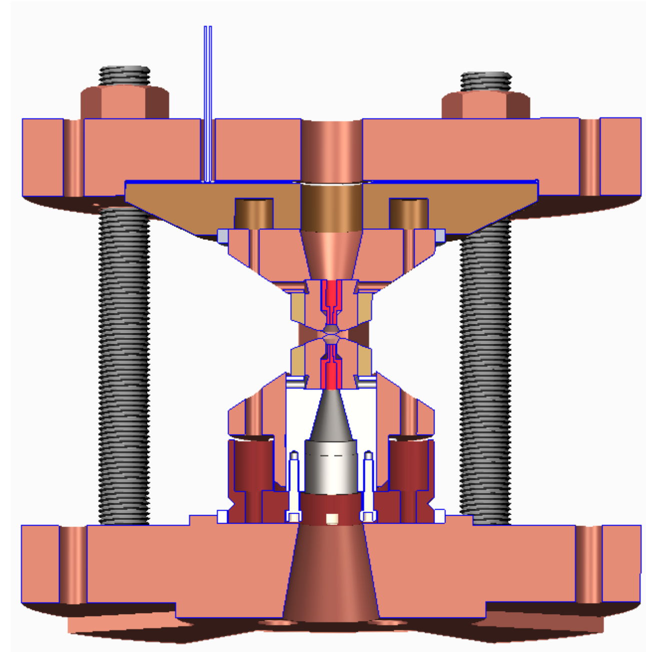
Pressure

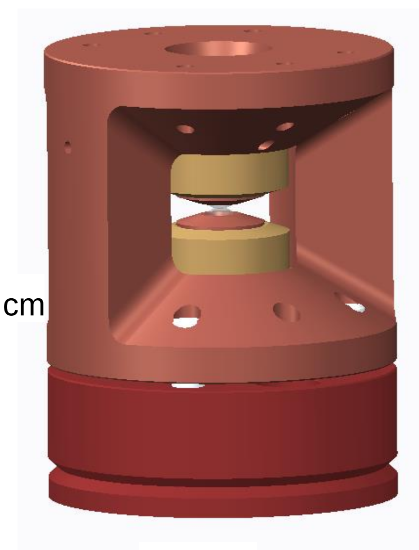
Diamond Anvil Cell (SNAP)

- Max recorded Pressure for Neutrons: 94GPa.
- Reliable Pressures up to 40 GPa
- DAC can be made of Steel with Diamond anvils. For low temperatures, CuBe is used.
- Sample volume of 0.4 mm^3 (1st gen cell 0.06 mm^3) 1.3 mm diameter and 0.3 mm gasket thickness
- Beamline Background reduction and collimation is of utmost importance for diamond anvil cell measurements.

3 kbar Gas Loading System

- Enables gaseous samples
- Hydrostatic gas medium
- Hydrogen compatible



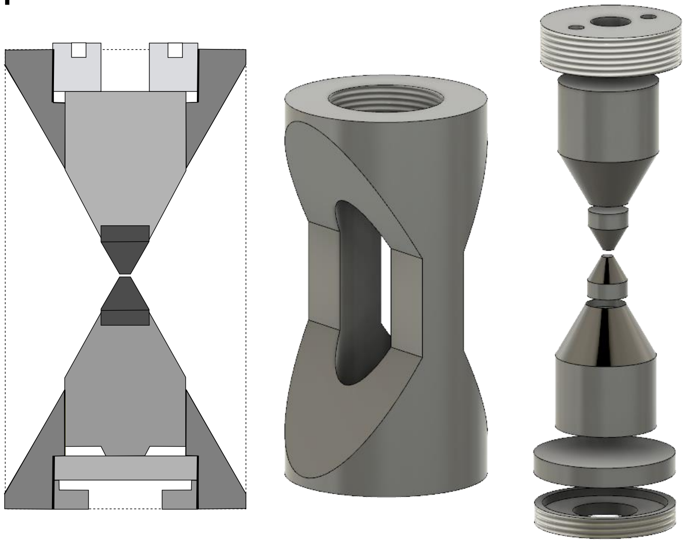


Pressure

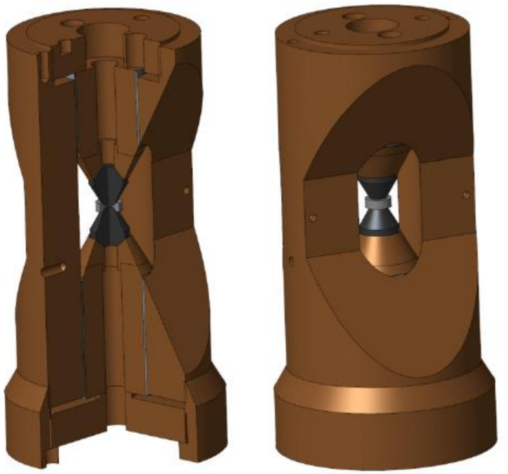
Clamped diamond anvil cell with Versimax® anvils:

- Reliable pressures up to 10 GPa
- VISION, CORELLI, HB-3A, IMAGINE, NOMAD
- Opening aperture of 120º.
- Pressure is applied in press and clamped in via a simple spring.
- Cell can be cooled to ~5 K.
- Sample volume is ~1 mm^3.

PCD anvil and gasket



Original Vascomax design [1]



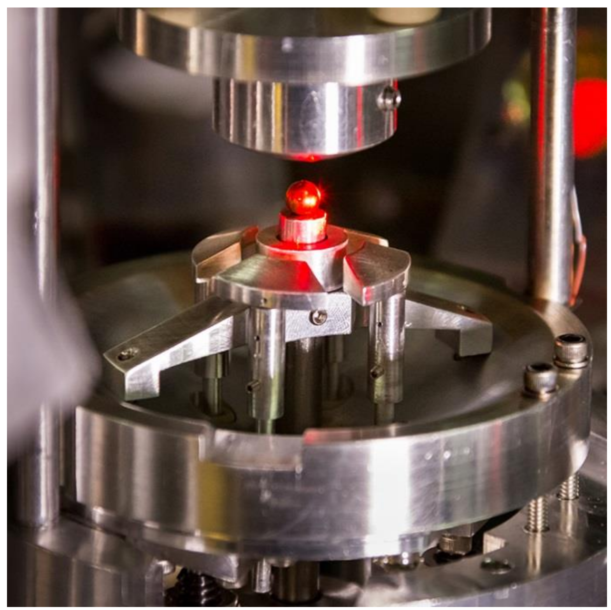
Optimized CuBe design [2]

[1] B. Haberl et al, High Pressure Research 37, 495 (2017).
[2] B. Haberl et al, submitted to Re. Sci. Instr. (2018).

High Temperature

High Temperature Equipment:

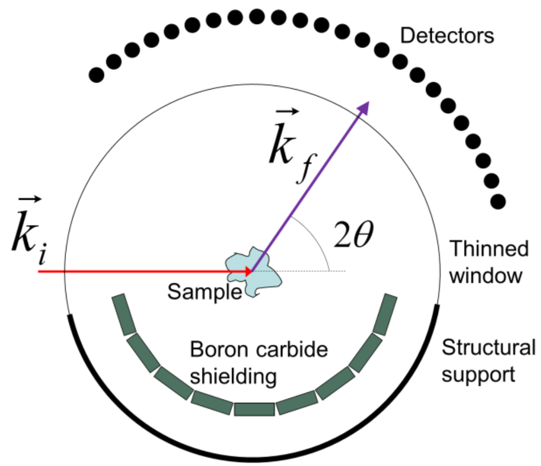
- 1200 °C Vanadium ILL
- 1600 °C Niobium ILL
- 1200 °C or 1600 °C MICAS2
- 1500 °C Controlled Atmosphere
- 500 - 2000 °C Electrostatic Levitator

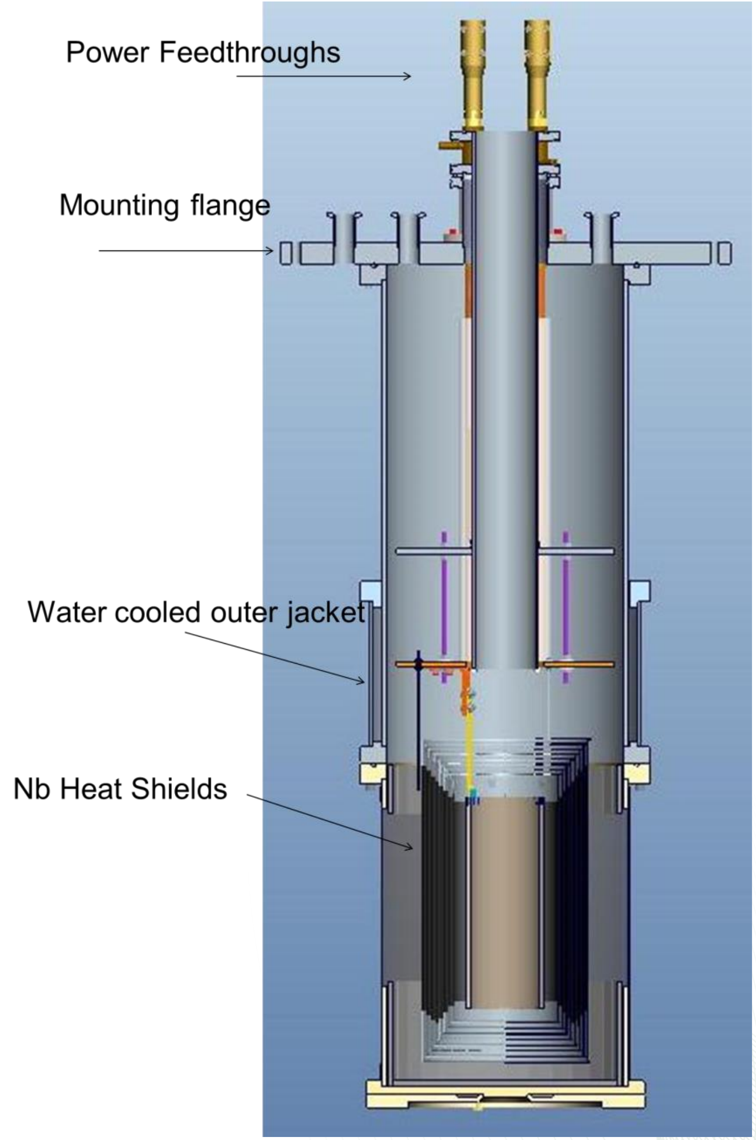


High Temperature

Radiative Heating Furnace

- Customize Outer Vacuum Chamber for detector coverage
- Minimize background using a thin (0.05 mm) Niobium window instead of Aluminum on Outer Vacuum Chamber
- Use of Boron Carbide to prevent multiple scattering





Container-less Sample Environment

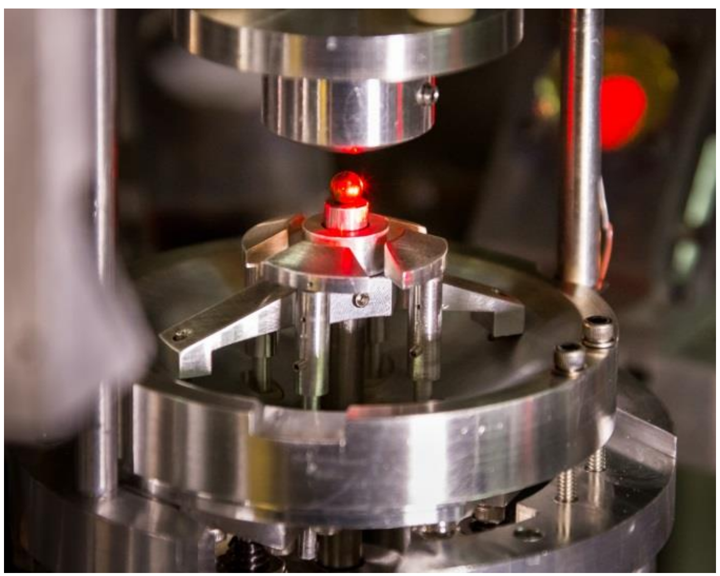
Neutron Electrostatic Levitator

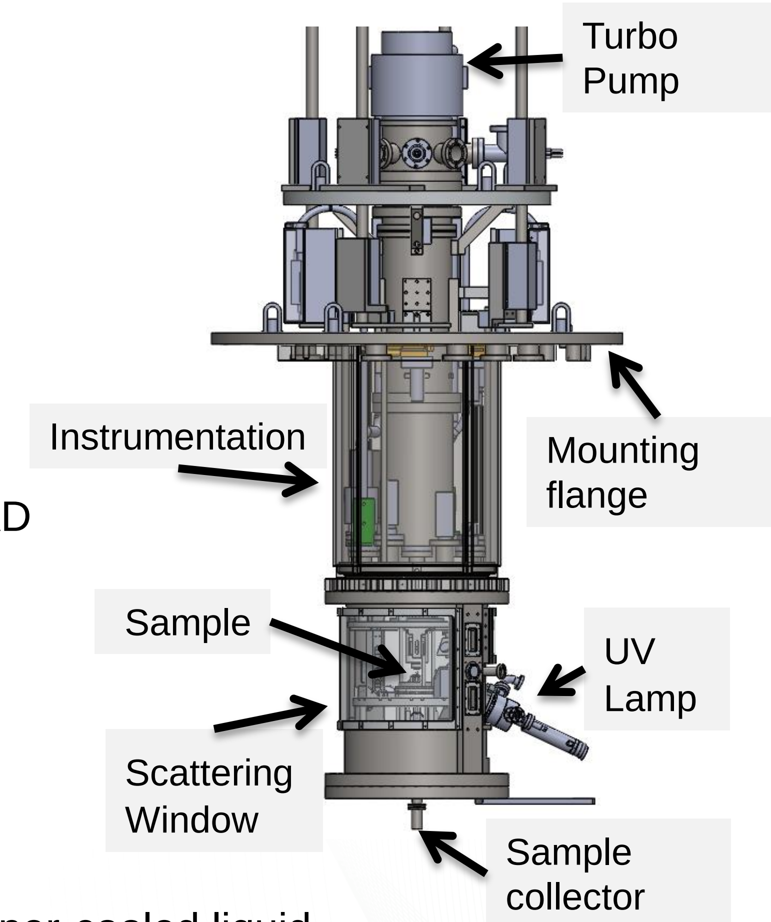
Container-less sample environment: no obstruction or reactivity from container

High temperature (500 – 2000 °C), high vacuum (10^{-7} torr), high voltage (30 kV, 14 mm)

Sample carousel holds up to 29 samples: spherical shape preferred, diameter 3-5 mm, mass 100-400 mg

Successful commissioning and experiments on NOMAD and ARCS





Access to super-cooled liquid regime

No container to induce nucleation

Take Home Message

- When designing equipment, one must choose the right materials
  - Equipment must satisfy material physical properties
  - Minimize amount of material in the incident and detected neutron beam paths
  - Choose neutron-friendly materials: minimize neutron interaction (avoid incoherent background or coherent scattering from the equipment itself)
- There are always trade-offs and compromises: Yield strength of material allows for high pressures but has high neutron absorption cross section

Take Home Message

- Build in as much flexibility as possible in the sample area: Experiments are continuously more complex
- Don't skimp on the utilities: Electrical power, crane coverage, compressed air, chilled water, Helium recovery
- Remember your neighbors: Stray magnetic fields, neutron background
- It is important to define requirements up front
- Project management: budget, scope and time