The Dynamic Nuclear Polarization Program at ORNL

PSTP2024

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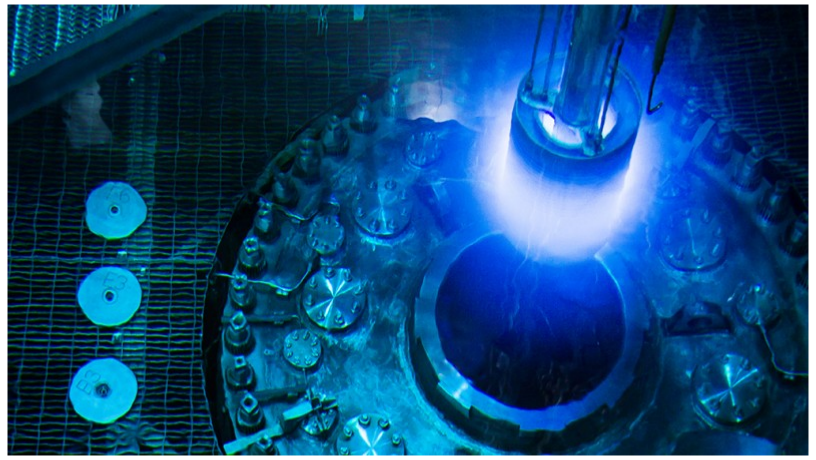
ORNL

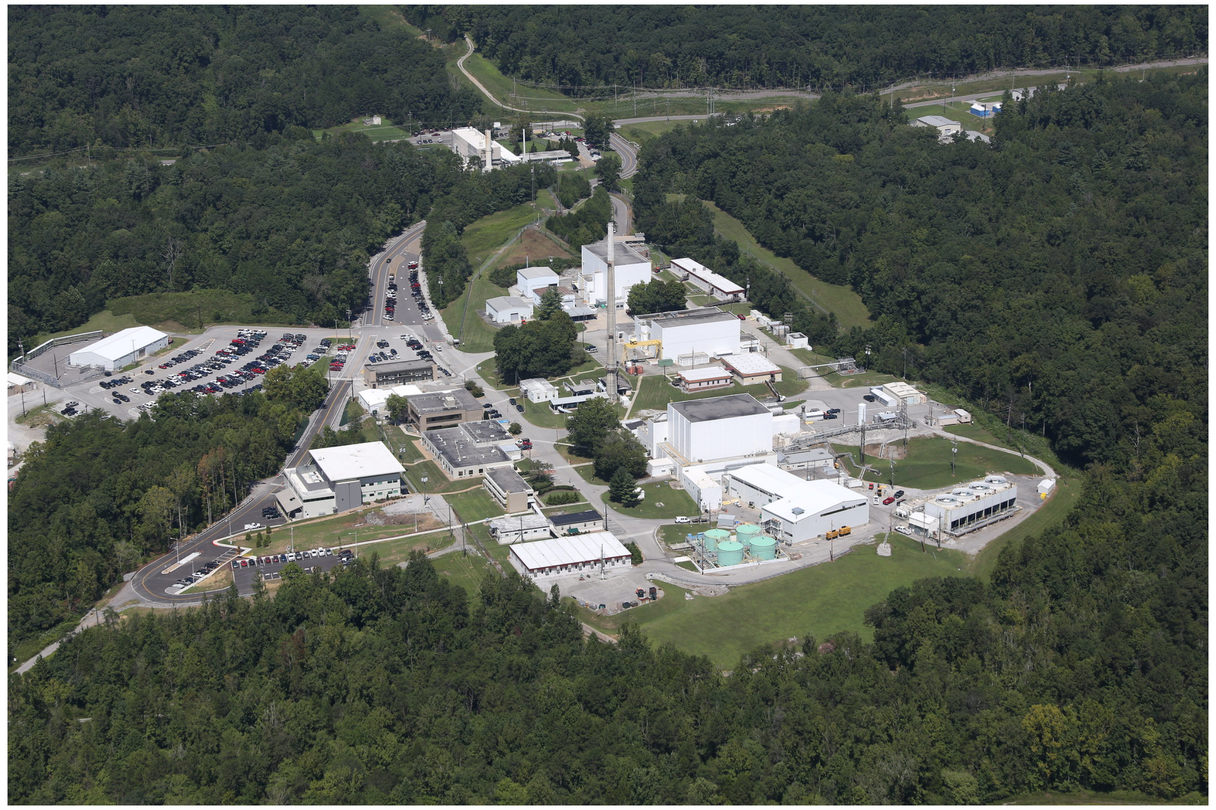
Outline

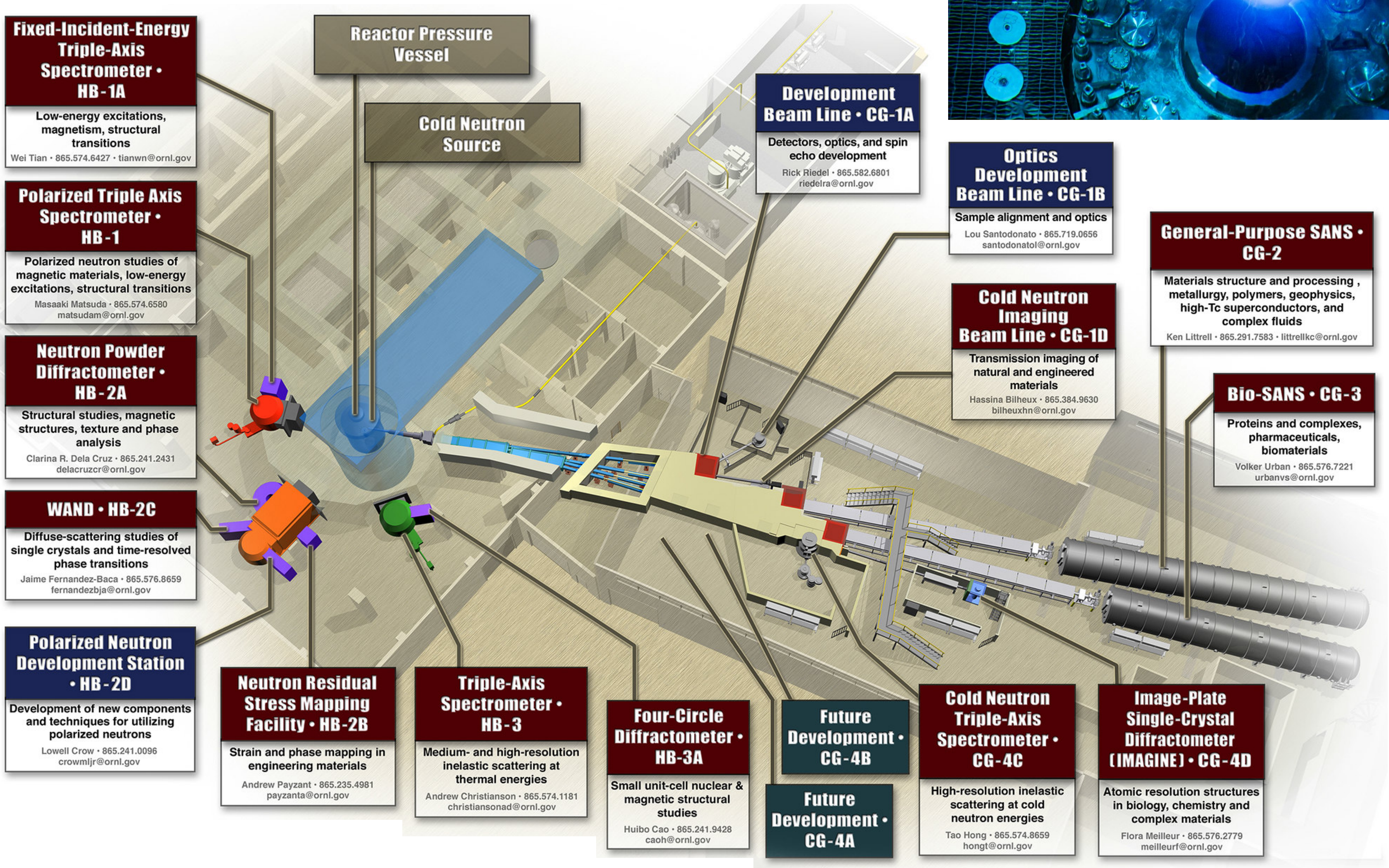
- Protein crystallography with spin polarization
- Proof-of-concept measurements
- Progress of DNP system development

High Flux Isotope Reactor (HFIR)



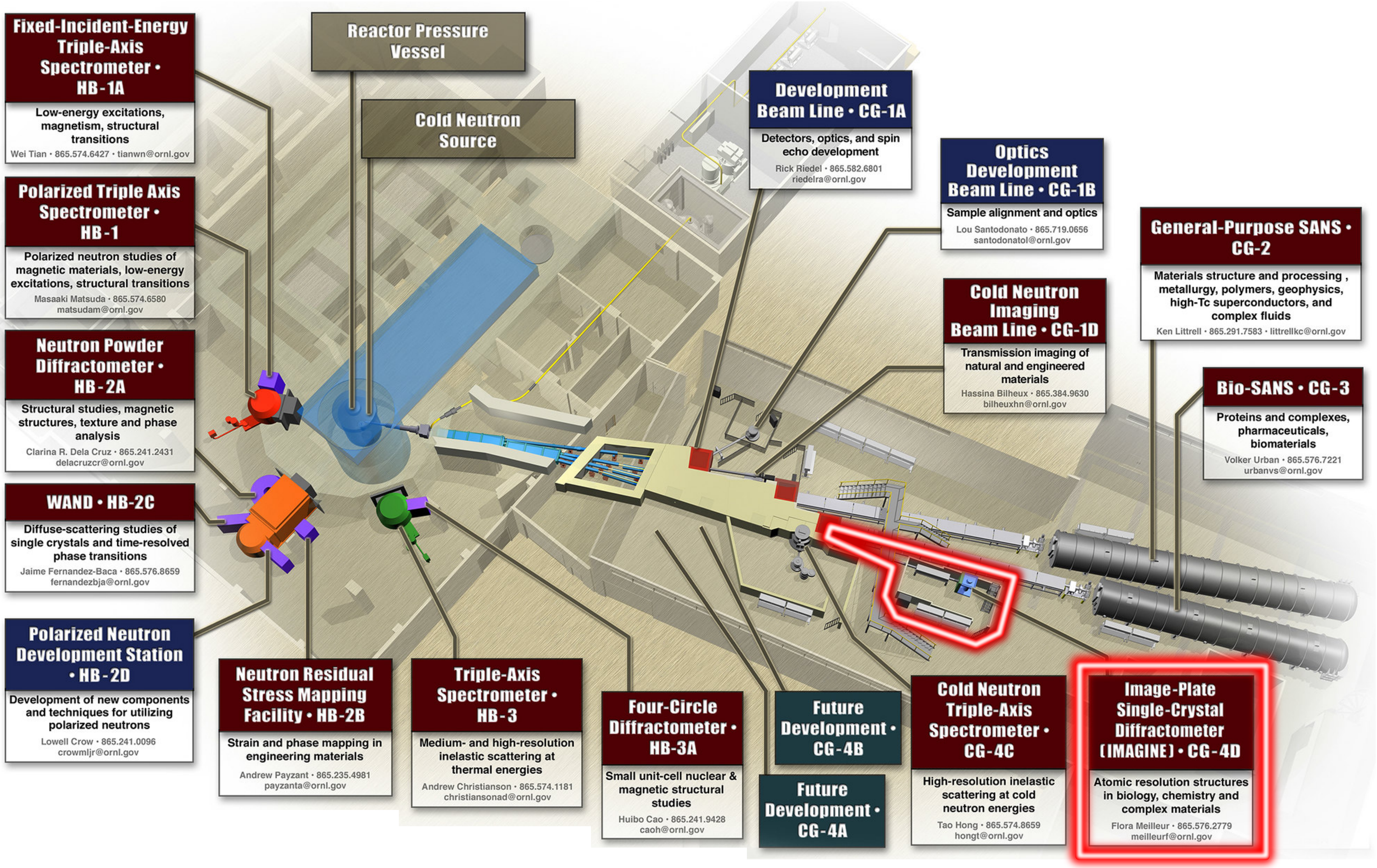
- HFIR, operating at 85 MW, is the highest flux reactor-based source of neutrons in US
- 4 beam ports with access to thermal and cold neutrons
- 13 instruments - diverse scientific program





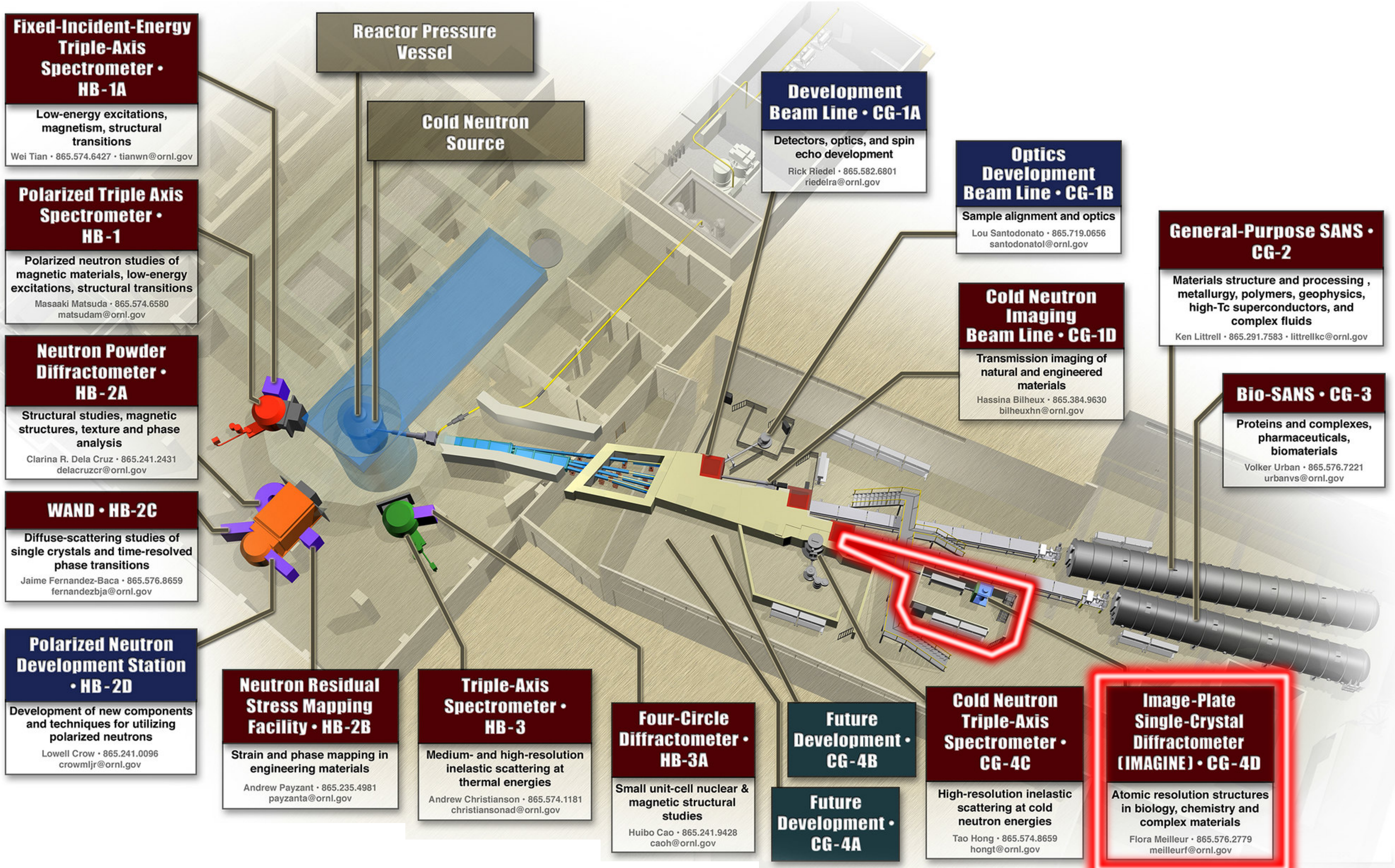
HFIR IMAGINE Beamline

- Studying biochemical processes at the atomic level, in particular proteins and other biomacromolecules
- Tunable beam optics for cold 2.0-4.5Å neutrons
- Quasi-Laue Diffractometer with neutron image plates



HFIR DNP-equipped IMAGINE-X Beamline

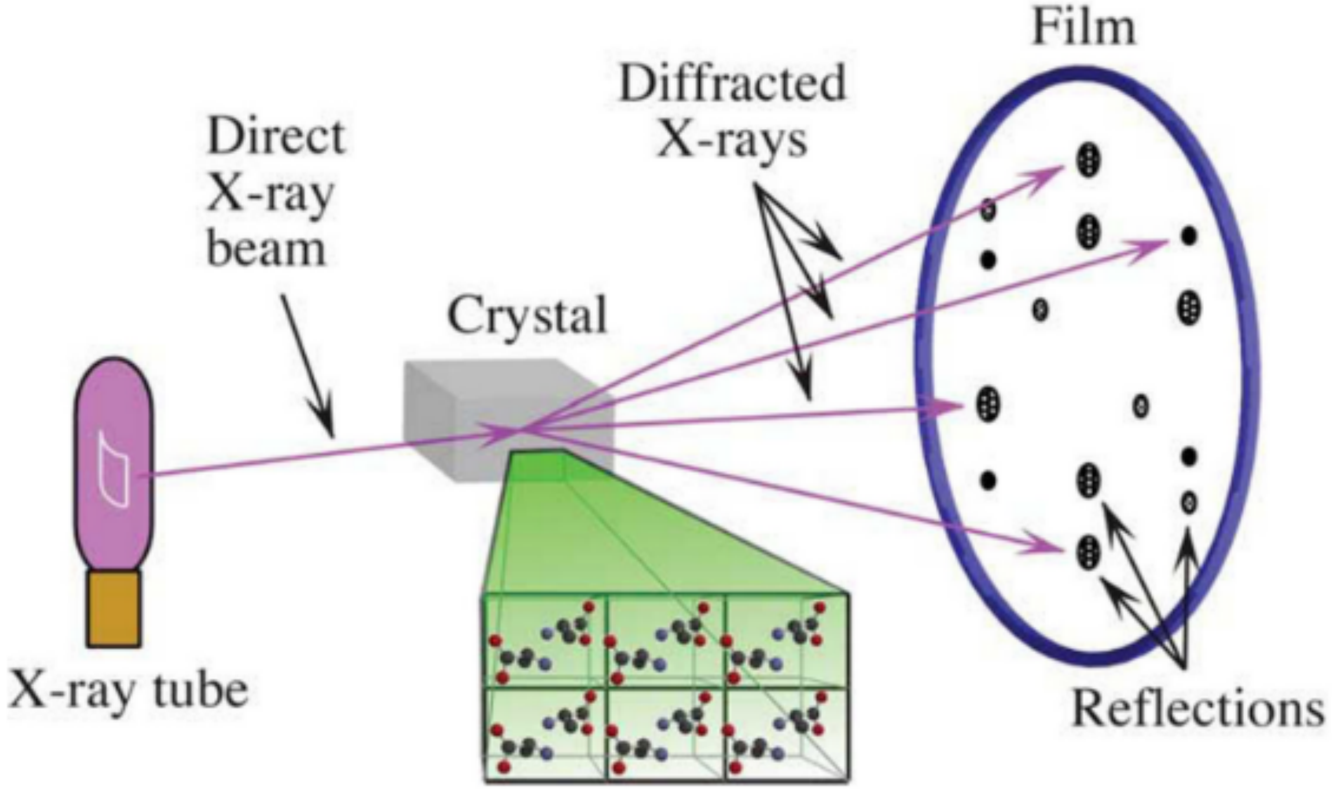
- Developing a new facility to amplify neutron diffraction from biological crystals
- Polarized neutron beam with spin flipper
- DNP system for sample polarization
- Two arrays of SiPM based Anger cameras to detect neutrons



X-Rays or Neutrons?

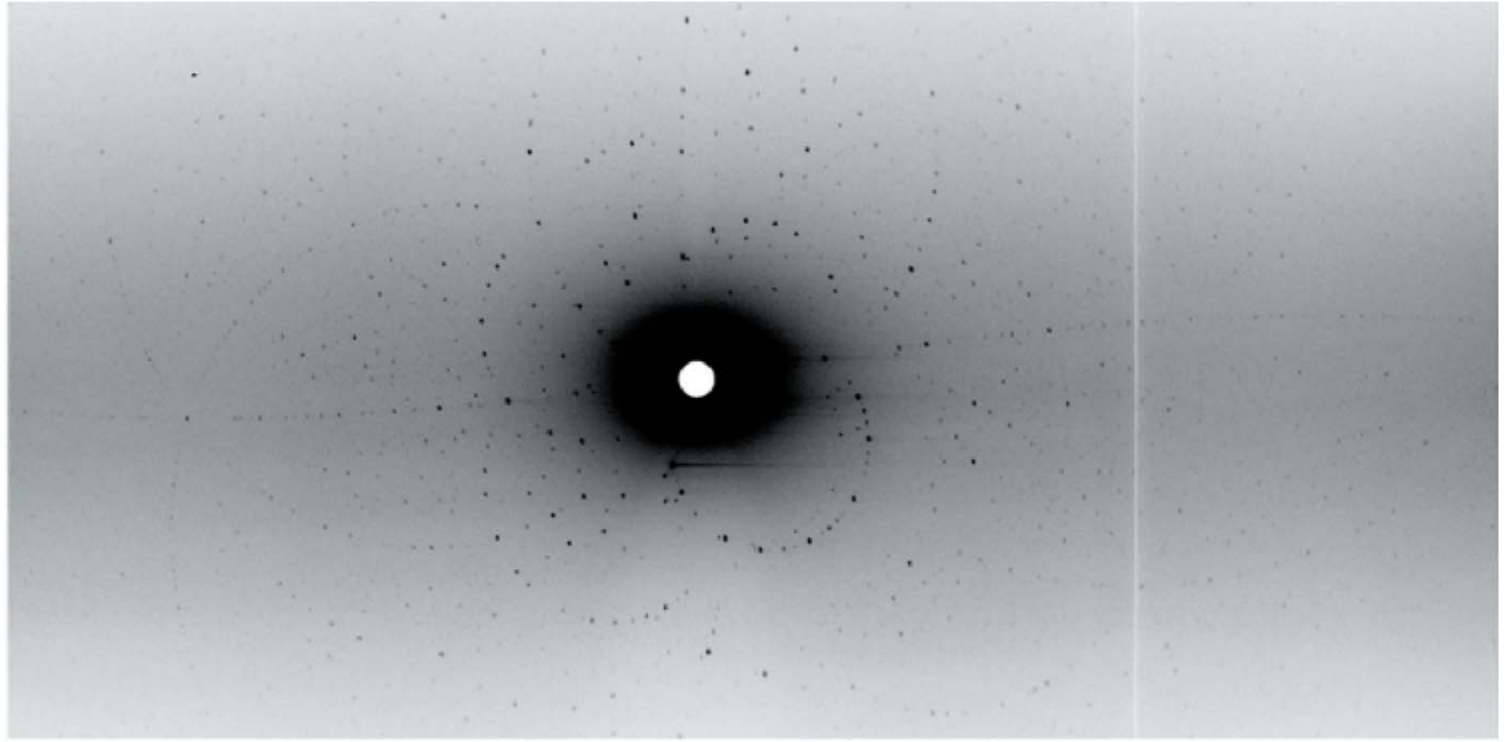
Neutron macromolecular crystallography - **NMC**

- main objective: reconstruction of 3d molecular structure from a crystal
- importance: drug discovery for bio-preparedness, biochemistry at atomic scale



From “Crystallography Made Crystal Clear,” Gale Rhodes 2006

- Using X-rays scattering
  - modern X-rays facilities has very high beam flux
  - rapid data taking, small crystal size $< 0.001\mathrm{mm}^3$
- Using neutron scattering
  - Sensitivity to lighter elements (especially hydrogen)
  - Sensitivity to isotopes (deuteration is very powerful, where possible)

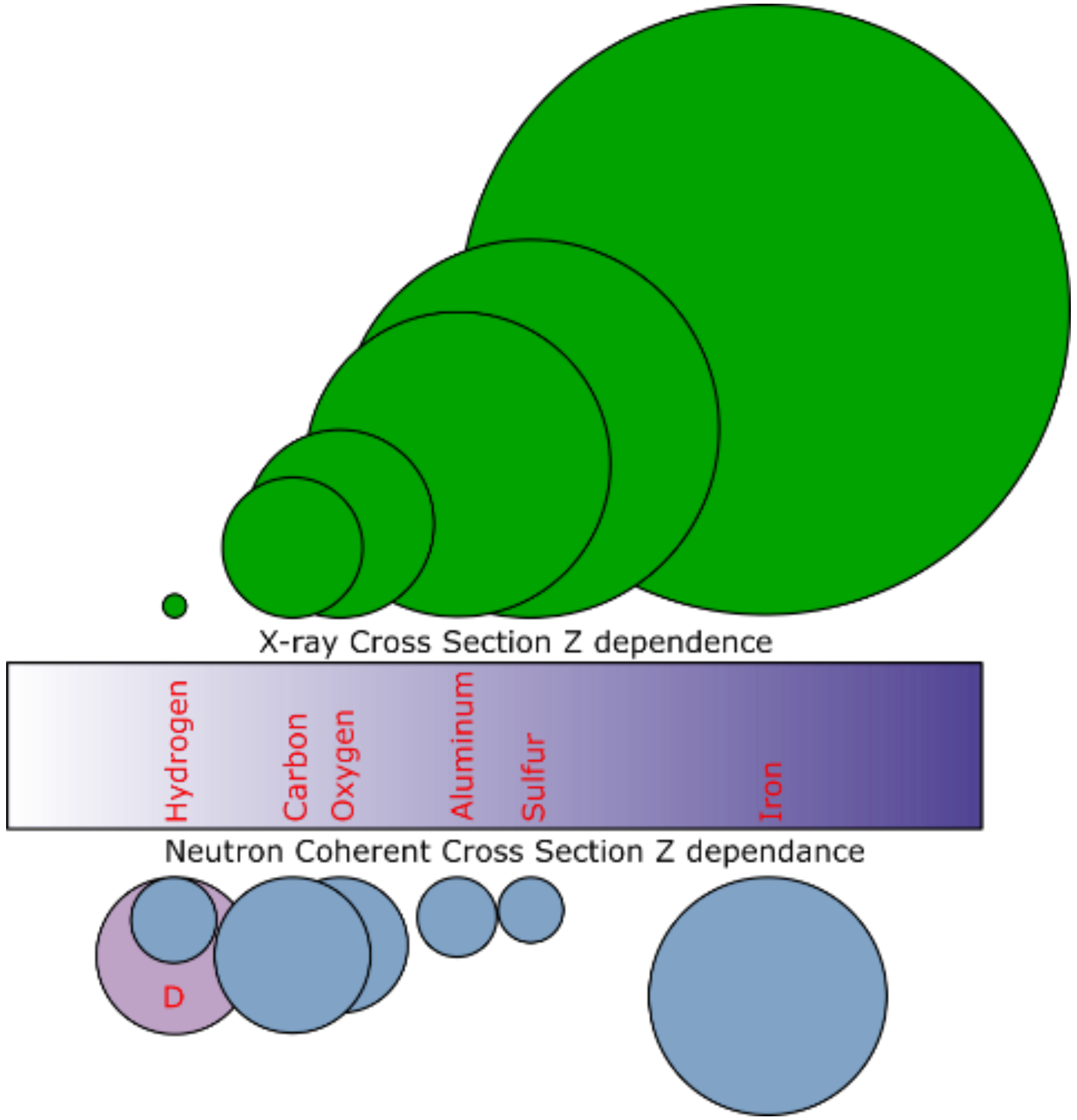


Laue diffraction image from IMAGINE

NMC or Polarized NMC?

Neutron macromolecular crystallography - **NMC**

- Using neutron scattering - challenges:
  - comparatively low flux: limitation on crystal size $>0.1\text{mm}^3$
  - for hydrogen: presence of the incoherent (background) contribution that dominates coherent (signal) component



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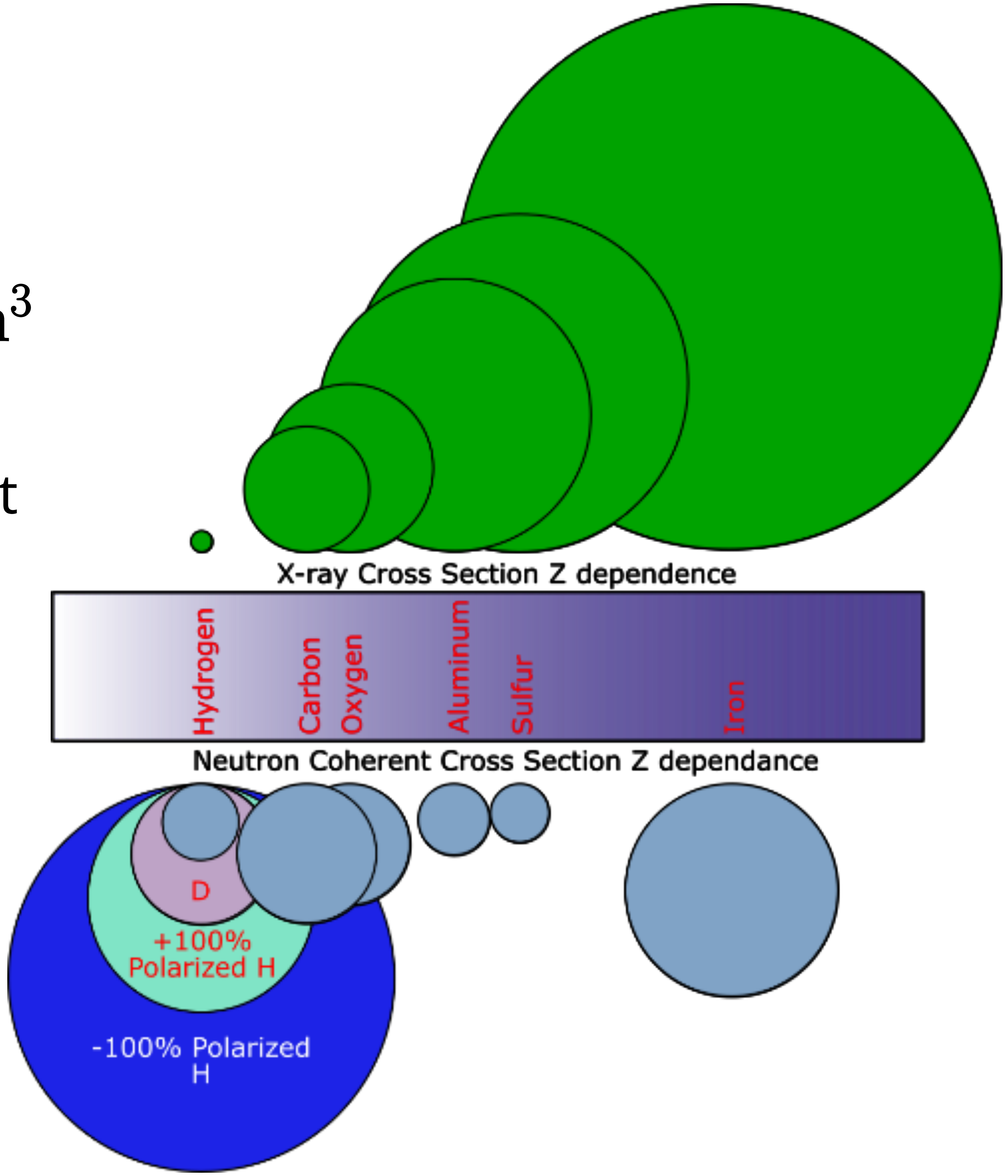
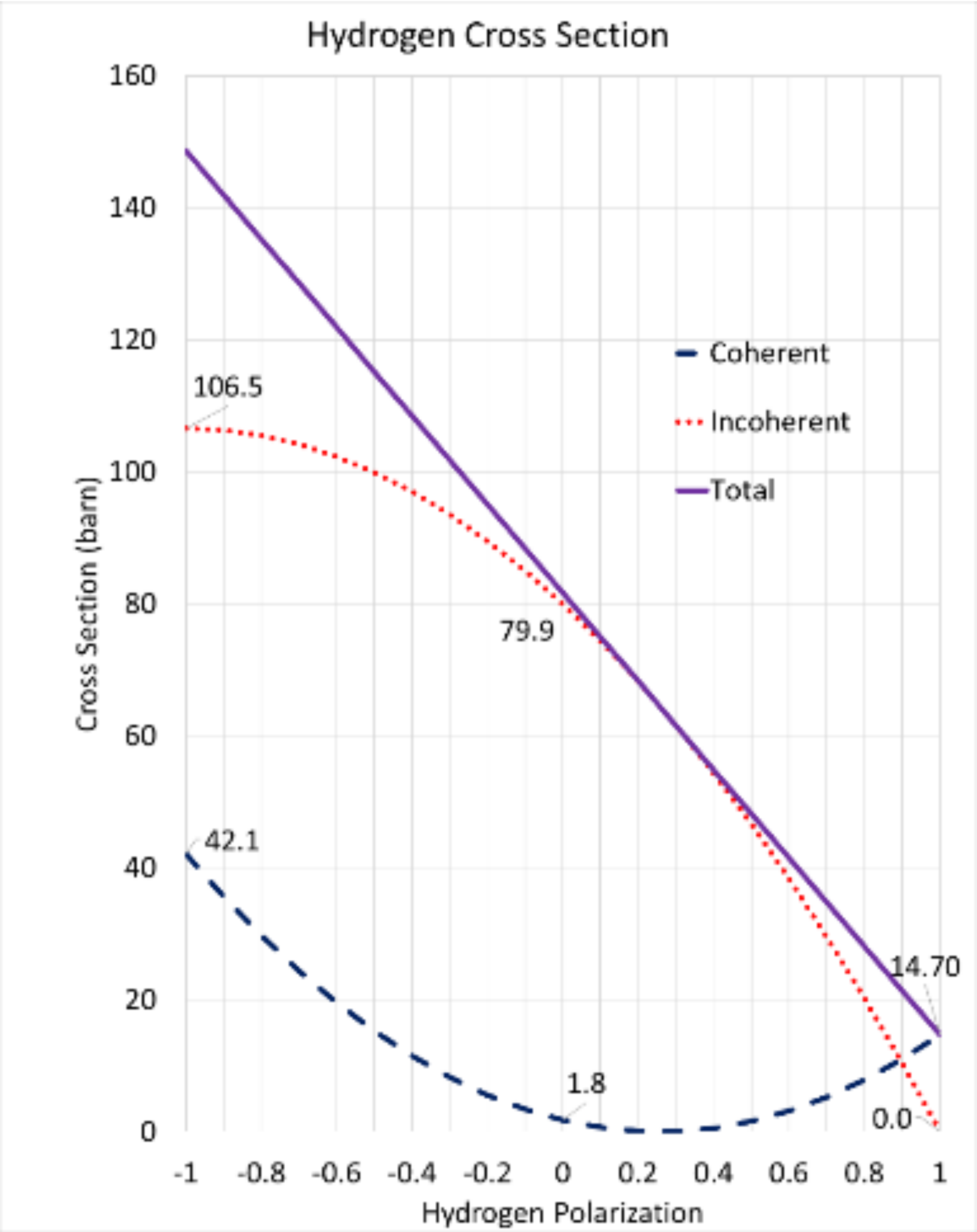
- Using spin alignment between the neutron and the struck nucleus
  - spin dependence particularly strong for hydrogen
  - affects both coherent and incoherent cross section
  - neutron and nuclear polarization can increase coherent scattering by a factor of 7 (or 20)



NMC or Polarized NMC?

Neutron macromolecular crystallography - **NMC**

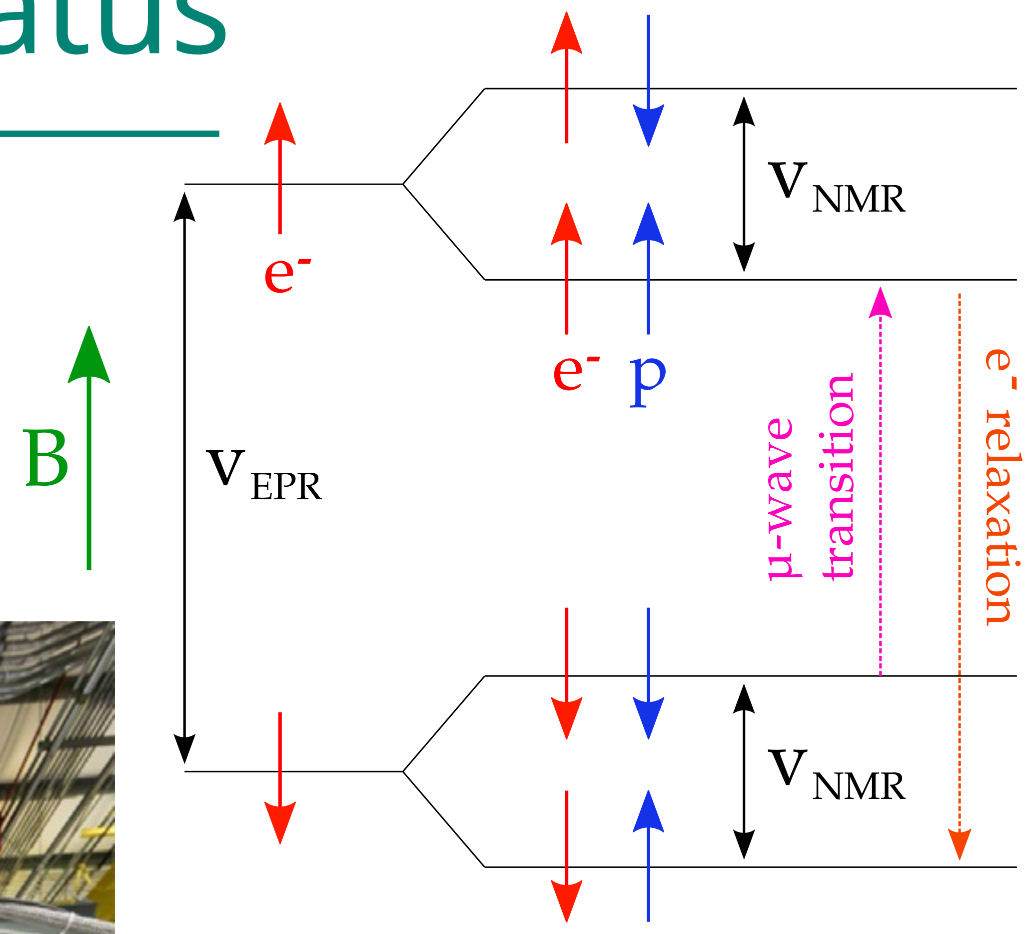
- Coherent/incoherent dependence
  - Nuclear incoherent scattering can be removed entirely (true for any nucleus)
- Polarizing neutrons
  - supermirror polarizers
  - 3He filters
- Polarizing hydrogen nucleus
  - dynamic nuclear polarization (DNP)



Coherent, incoherent and total scattering cross section of hydrogen as a function of the proton polarization for fully polarized neutrons.

Proof-of-concept DNP System - Apparatus

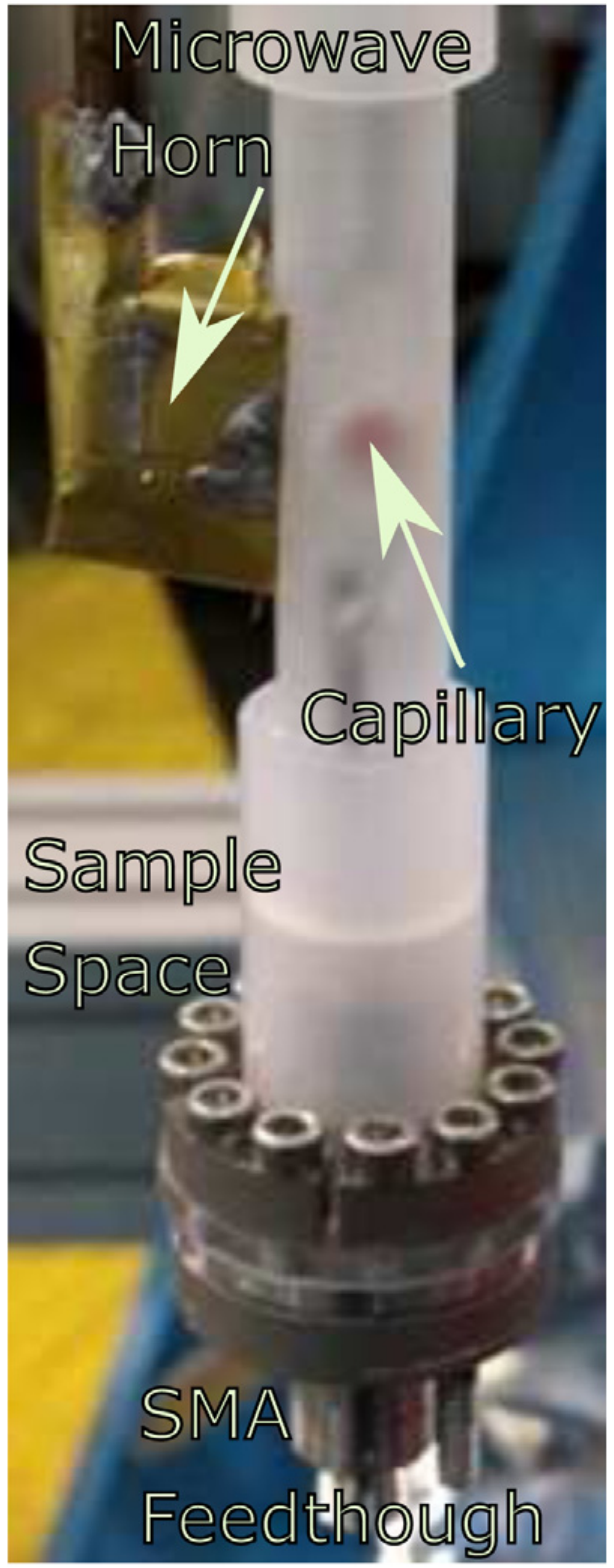
- use DNP process to get nucleon polarized
- use CW-NMR to get polarization measured



- Magnet (Cryomagnetics Inc.)
  - 5T, 100mm clear warm bore, cryogen-free
  - 10^4 uniformity in 2 cm DSV
  - custom designed lift table
- Dilution refrigerator (Bluefors Oy)
  - cryogen-free
  - custom tail and sample interface
  - cooling power of
    400 μW at 100 mK;
    12 mW at 1 K

Proof-of-concept DNP System - Apparatus

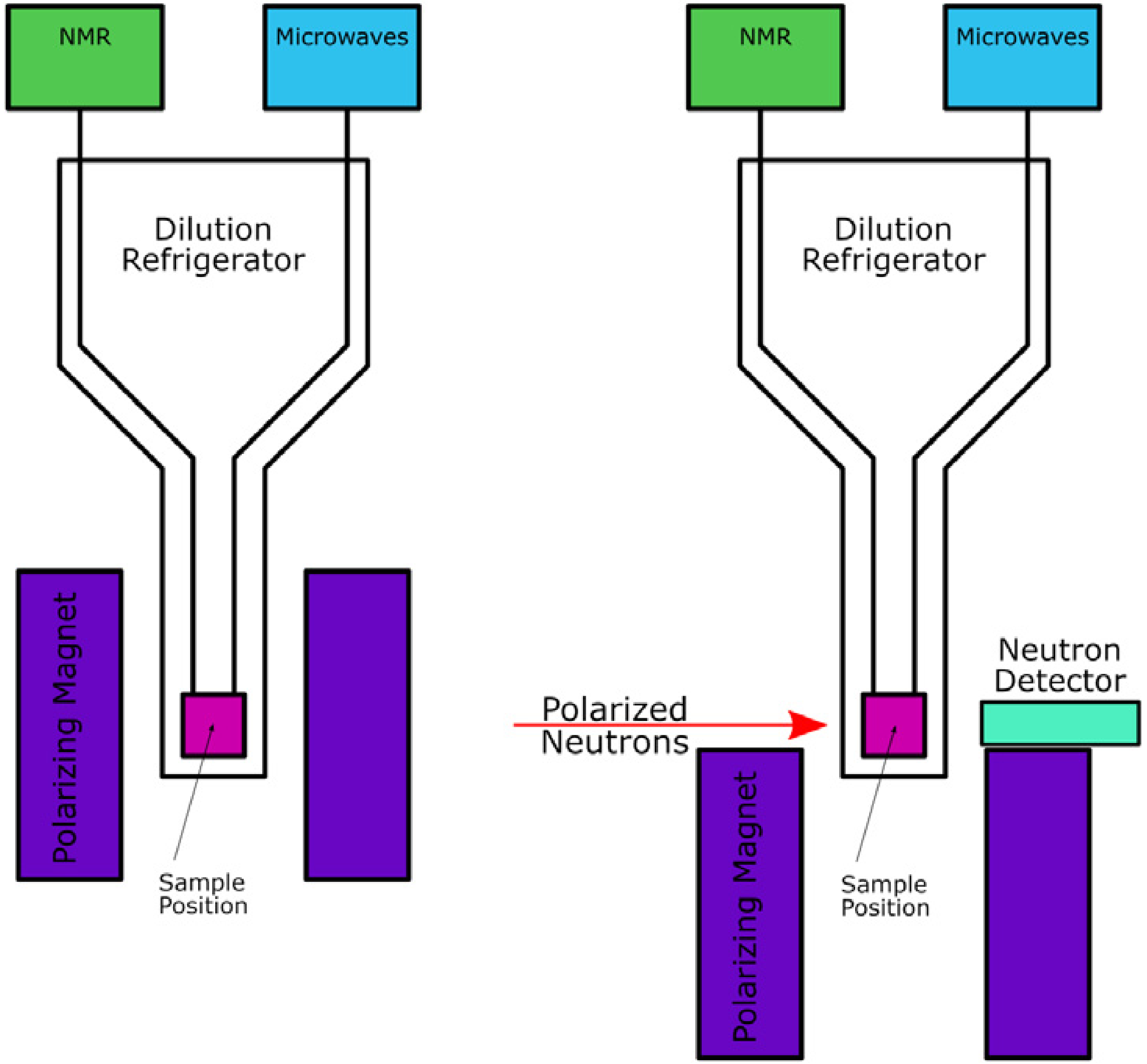
- Microwaves source (Virginia Diodes, Inc.)
  - 300 mW at 140 GHz from solid state source
  - enough power for small sample $< 1\mathrm{mm}^3$
  - attenuator, power meter, custom waveguide
- NMR system (Liverpool Q-meter)
  - additional pre-amplifier card with a gain of 500
  - extremely small sample - hard to observe TE signal
  - monitor the relative decay of the polarization
- Sample preparation
  - T4 Lysozyme crystal (0.9mm on edge) doped with TEMPO
  - soaked for 30 to 60 min in a solution and flash frozen in LN2



Photograph of the PCTFE section of the sample space

Proof-of-concept DNP System - Operation

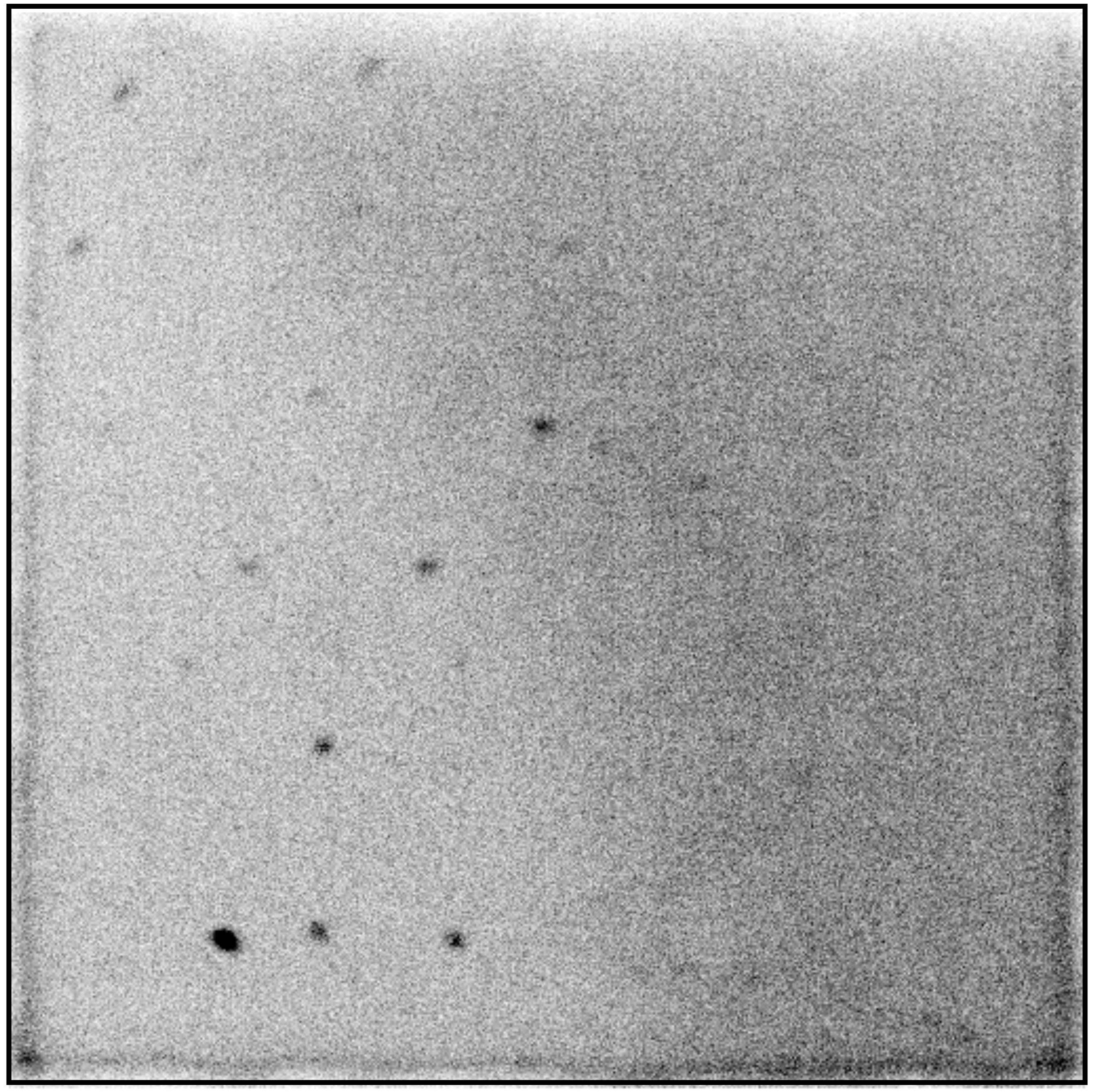
- No beam compatible magnet
  - two modes of operation: polarizing and frozen spin
  - at 1 K in a 5 T hydrogen is polarized via DNP (>8hrs)
  - at 200 mK in a 0.5 T data taking (T_1 ~120-240min)
- Beamline and detector
  - neutron beam was polarized using a V-cavity supermirror polarizer
  - diffraction image was reconstructed in a single arm prototype Anger camera
- Measured diffraction pattern change
  - recorded both spin aligned and anti-aligned configurations by changing the frequency of MW



Proof-of-concept DNP System - Results

- Average neutron polarization of >96.5%
- Maximum sample polarization was estimated to be ~50%
- Preliminary results
  - an overall 3-fold average spin-dependent amplification of background subtracted diffraction peaks between polarized and unpolarized states
  - almost double the number of diffraction peaks in anti-aligned configuration

Unpolarized



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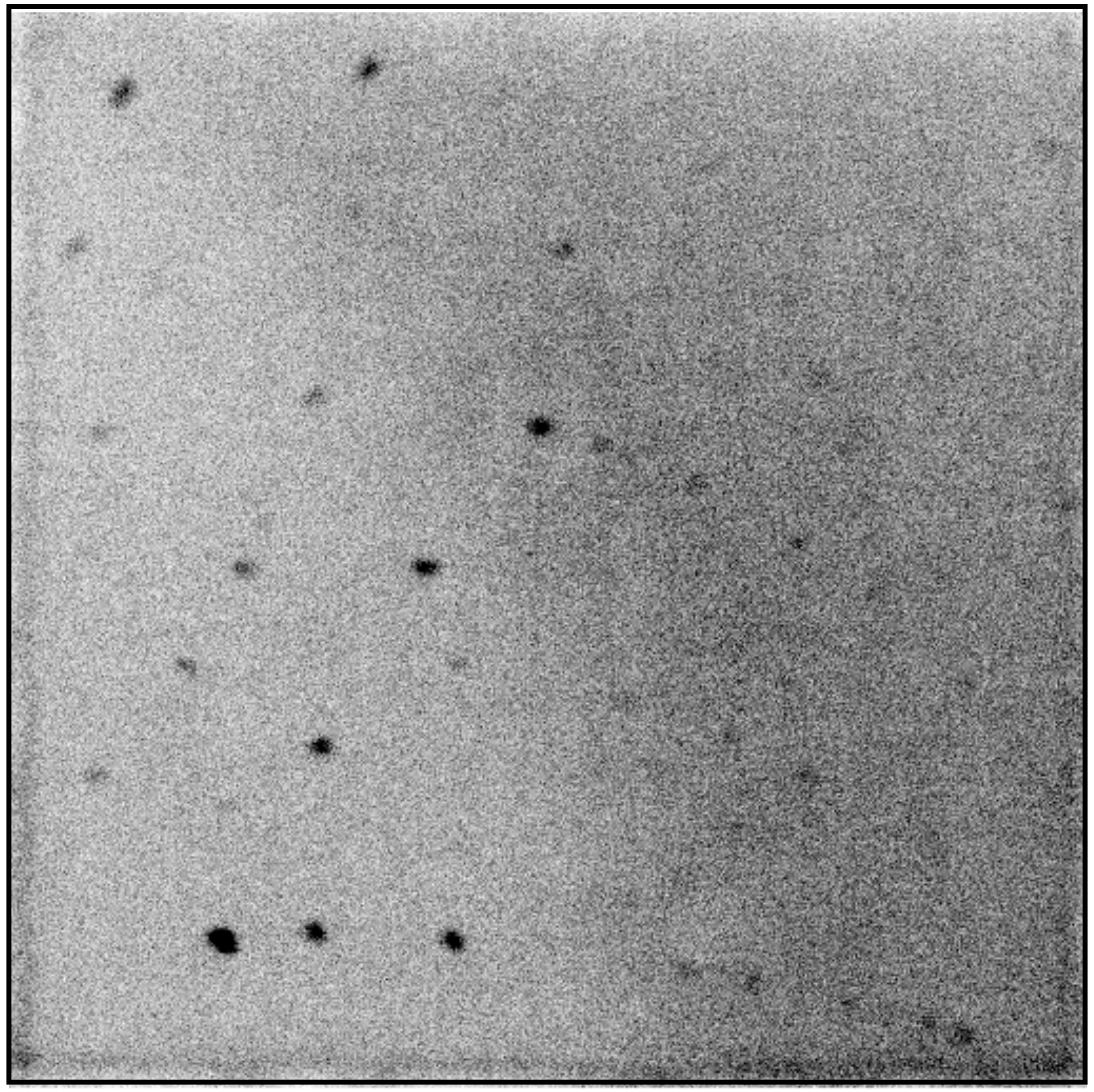
Polarized anti-aligned



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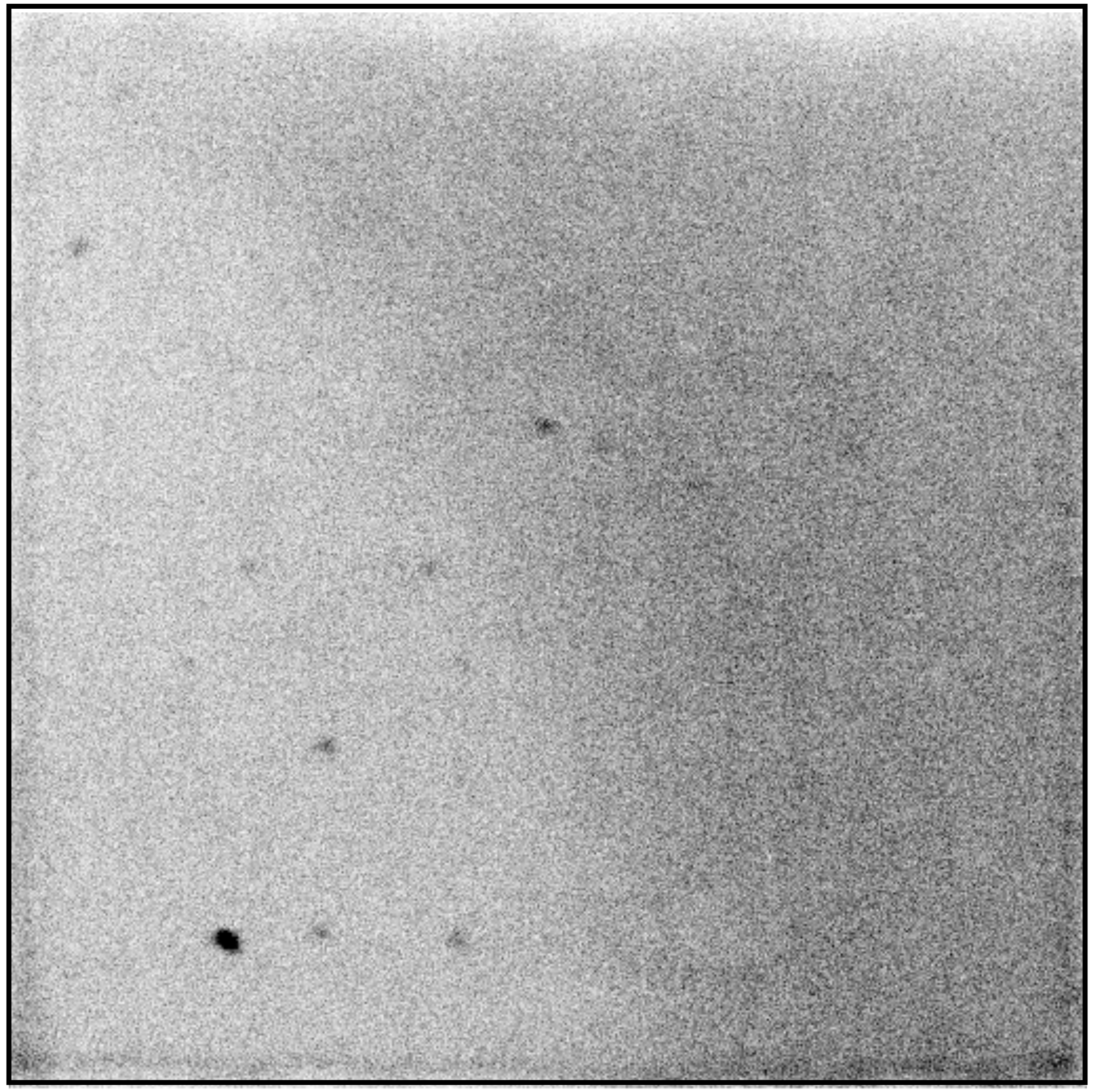
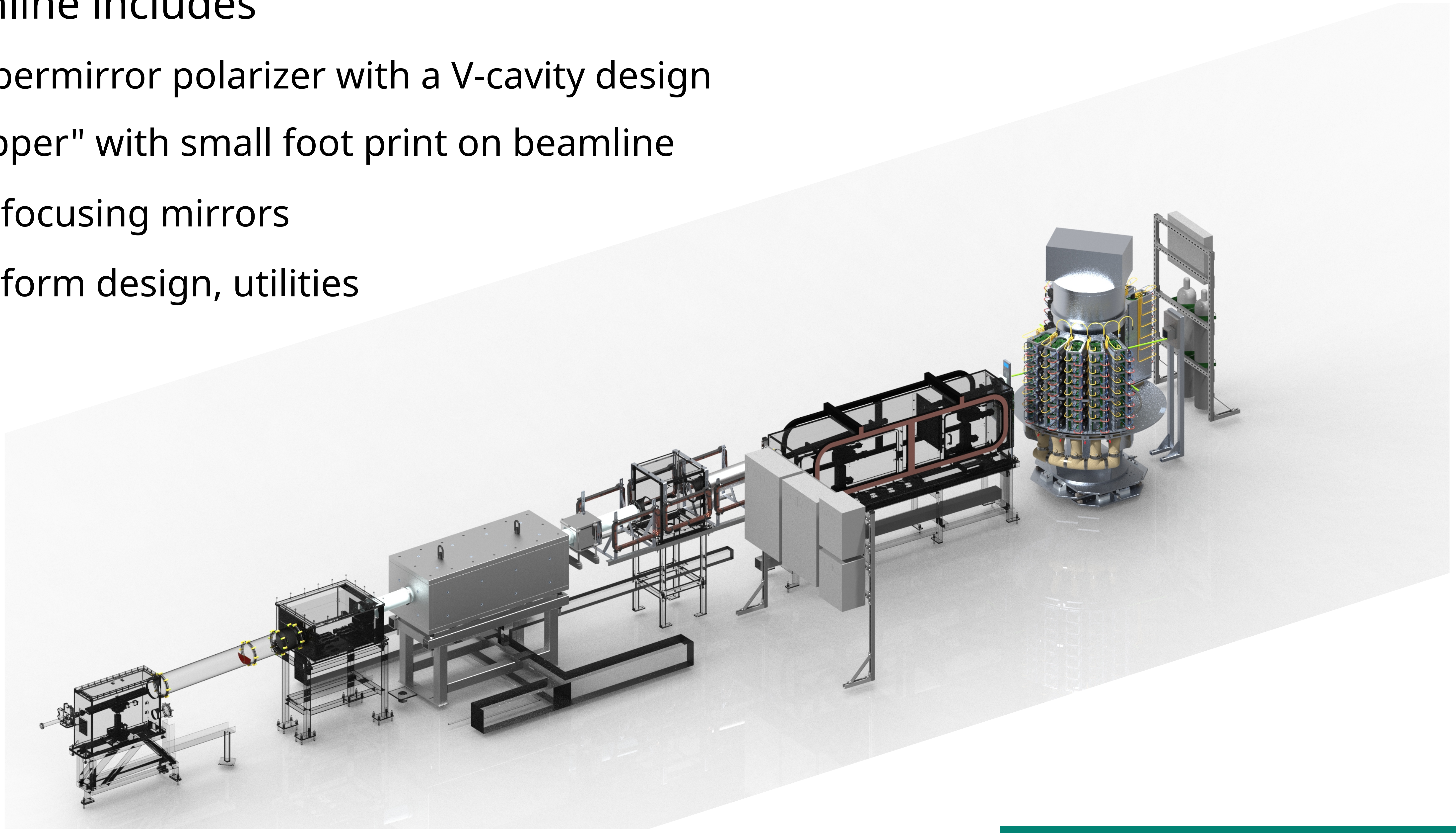
Polarized aligned



New Facility: IMAGINE-X for DNP-NMC

- New beamline includes
  - multi-supermirror polarizer with a V-cavity design
  - "Cryo-flipper" with small foot print on beamline
  - elliptical focusing mirrors
  - cryo-platform design, utilities



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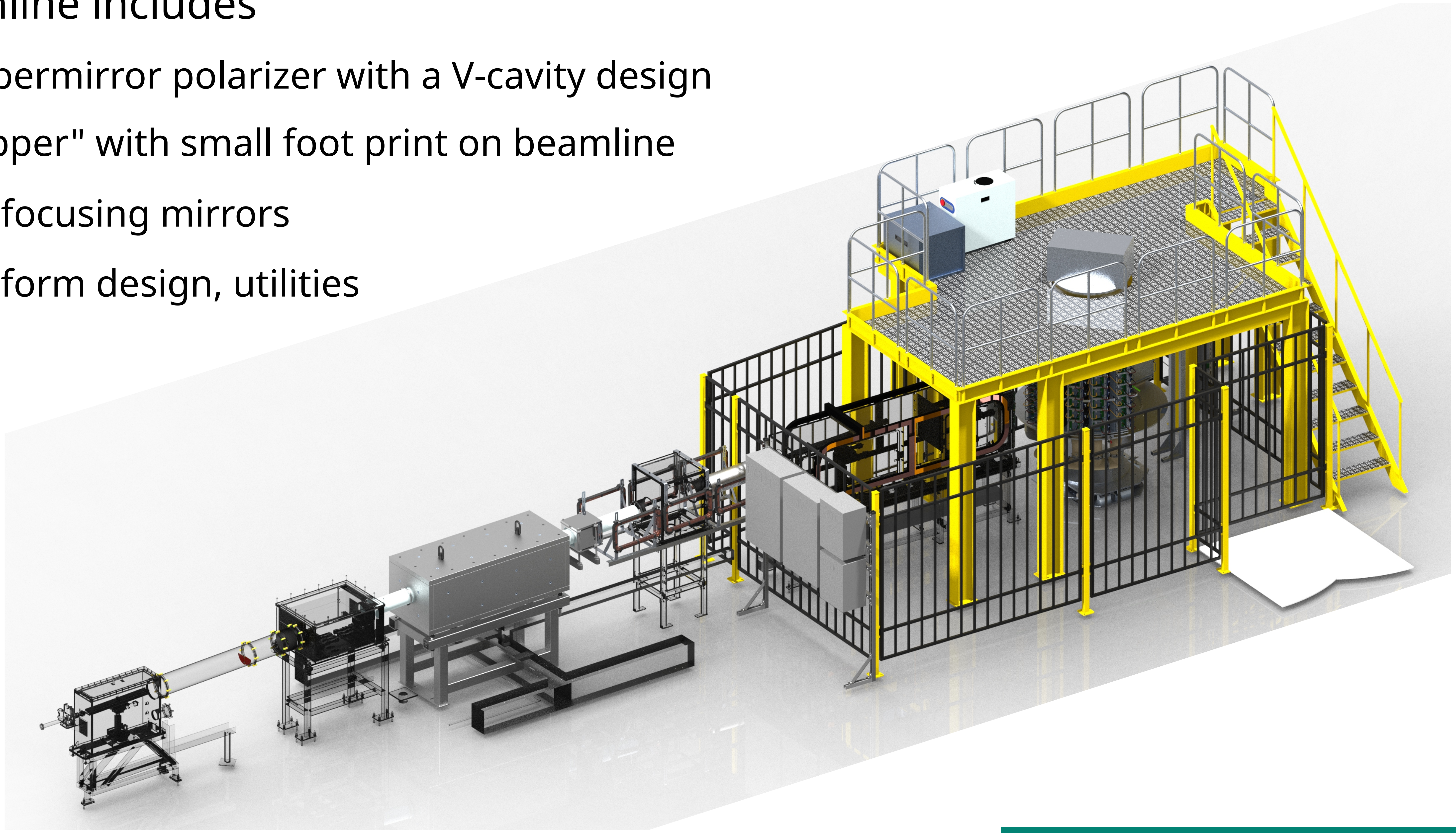

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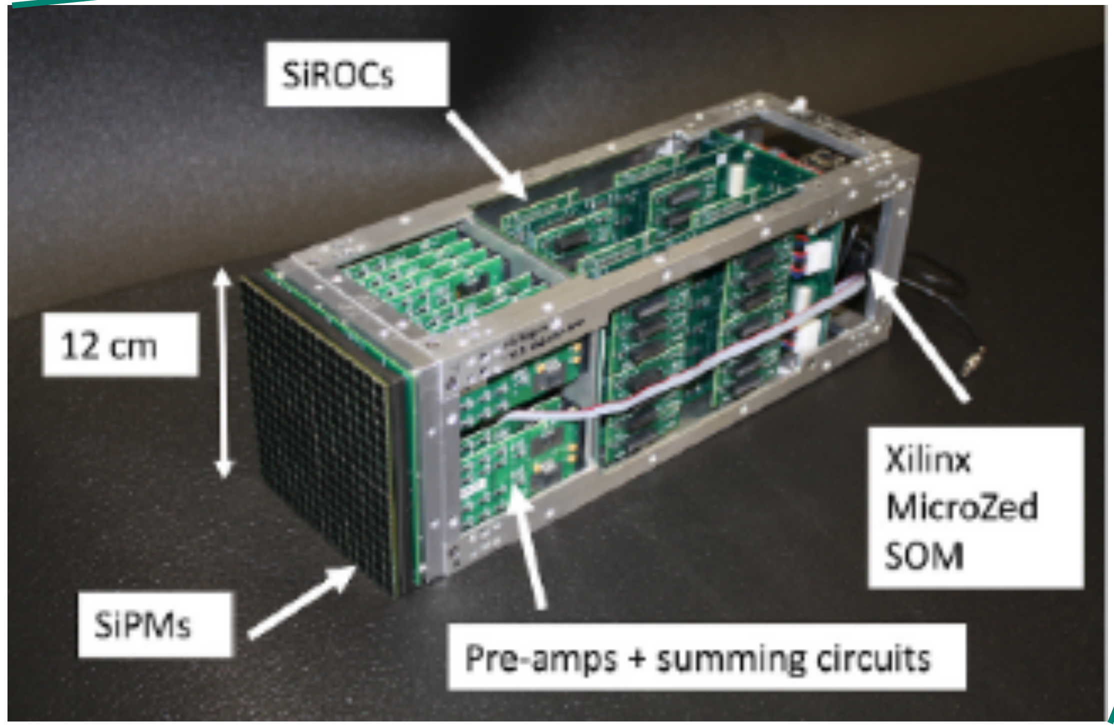
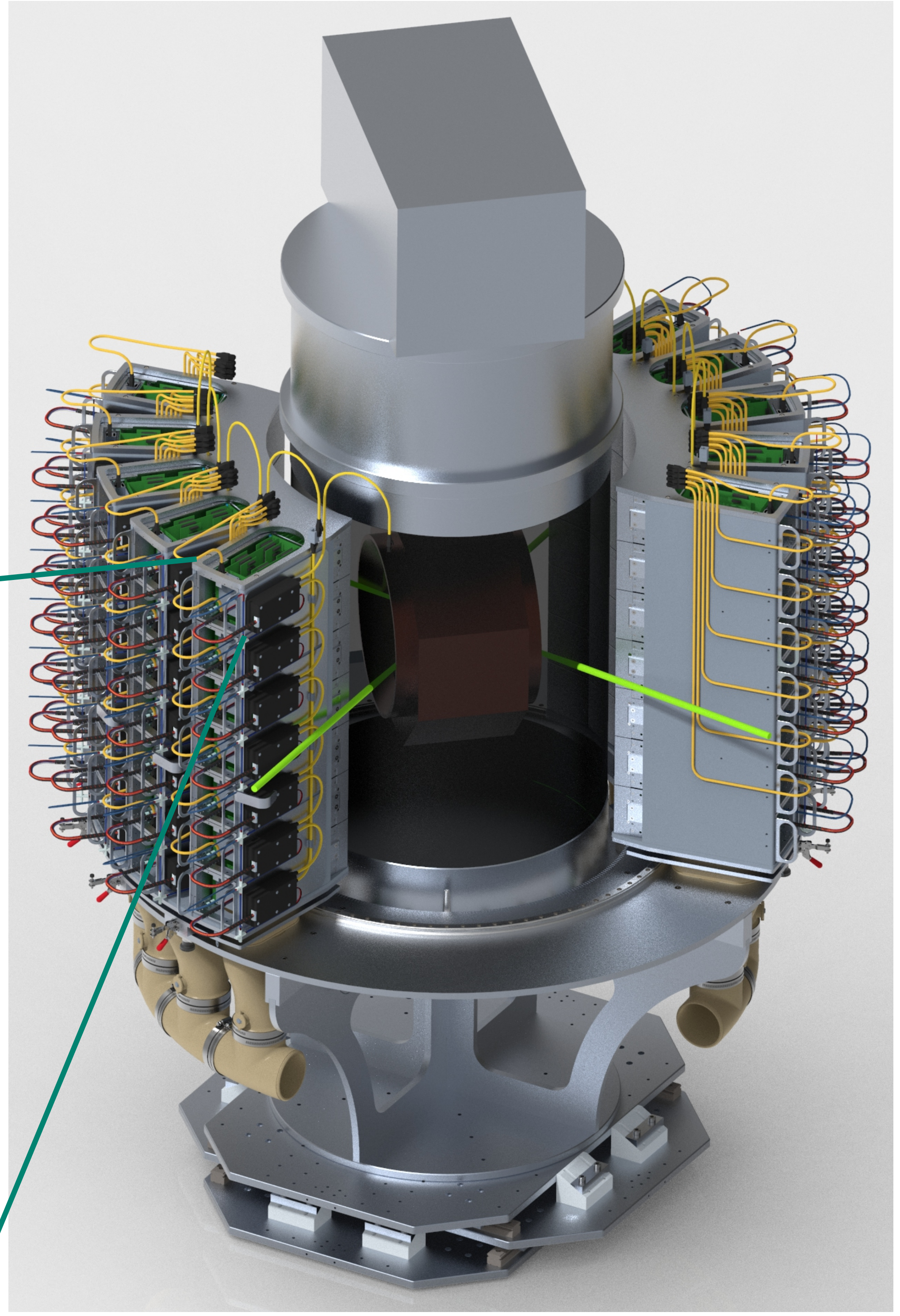
  - multi-supermirror polarizer with a V-cavity design
  - "Cryo-flipper" with small foot print on beamline
  - elliptical focusing mirrors
  - cryo-platform design, utilities
- New detector array
  - SiPM based Anger cameras
  - two arrays of 50 cameras
  - 0.45mm resolution, insensitive to magnetic fields

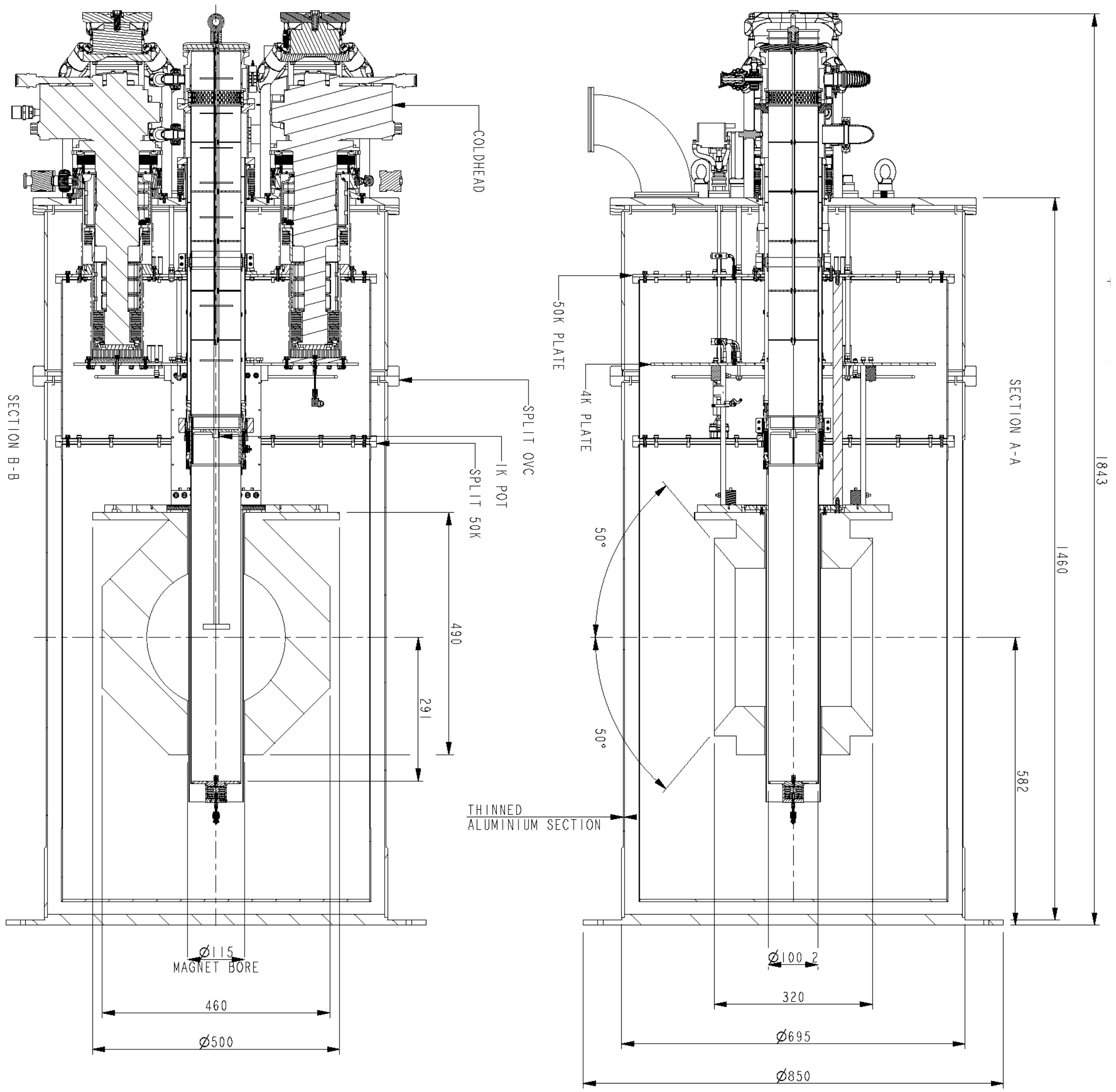




Loyd, M., Khaplanov, A., Sedov, V., Beal, J., Visscher, T., Donahue, C., ... & Diawara, Y. (2023).

New Facility: IMAGINE-X for DNP-NMC

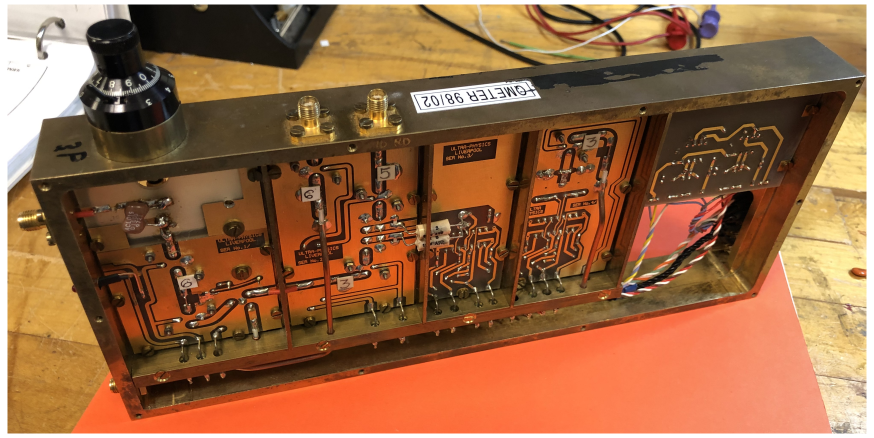
- DNP magnet + cryostat (ICEoxford)
  - Cryogen-free, superconducting 5T Helmholtz Coil
  - Cryogen-free 1K recirculating 4He refrigerator
  - continuously pumped DNP to maximize and maintain high, steady state levels of polarization
  - 54 deg opening coil angle:
    ~2π acceptance for scattered neutrons
  - ~200mW at ~1K cooling power
  - large sample space to accommodate goniometer

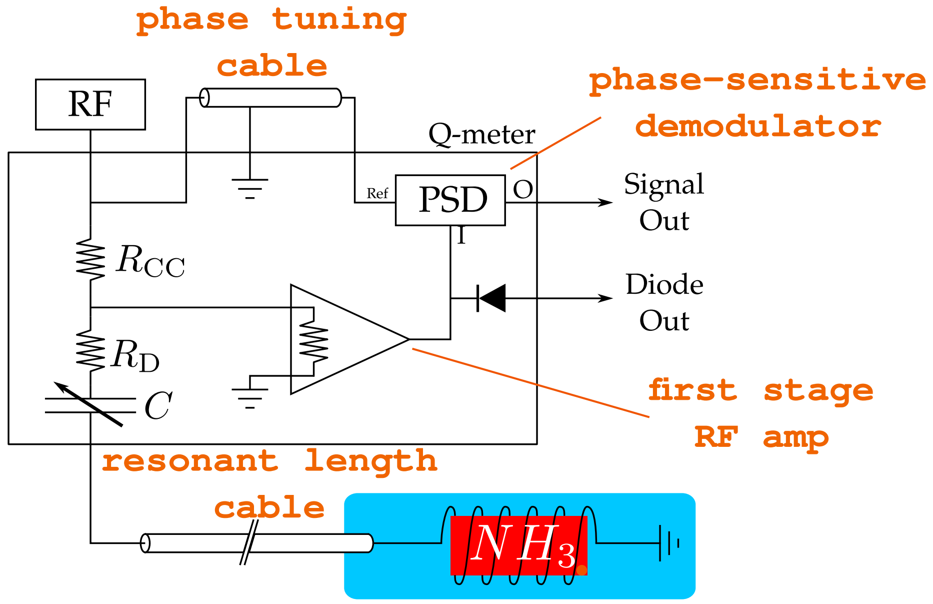


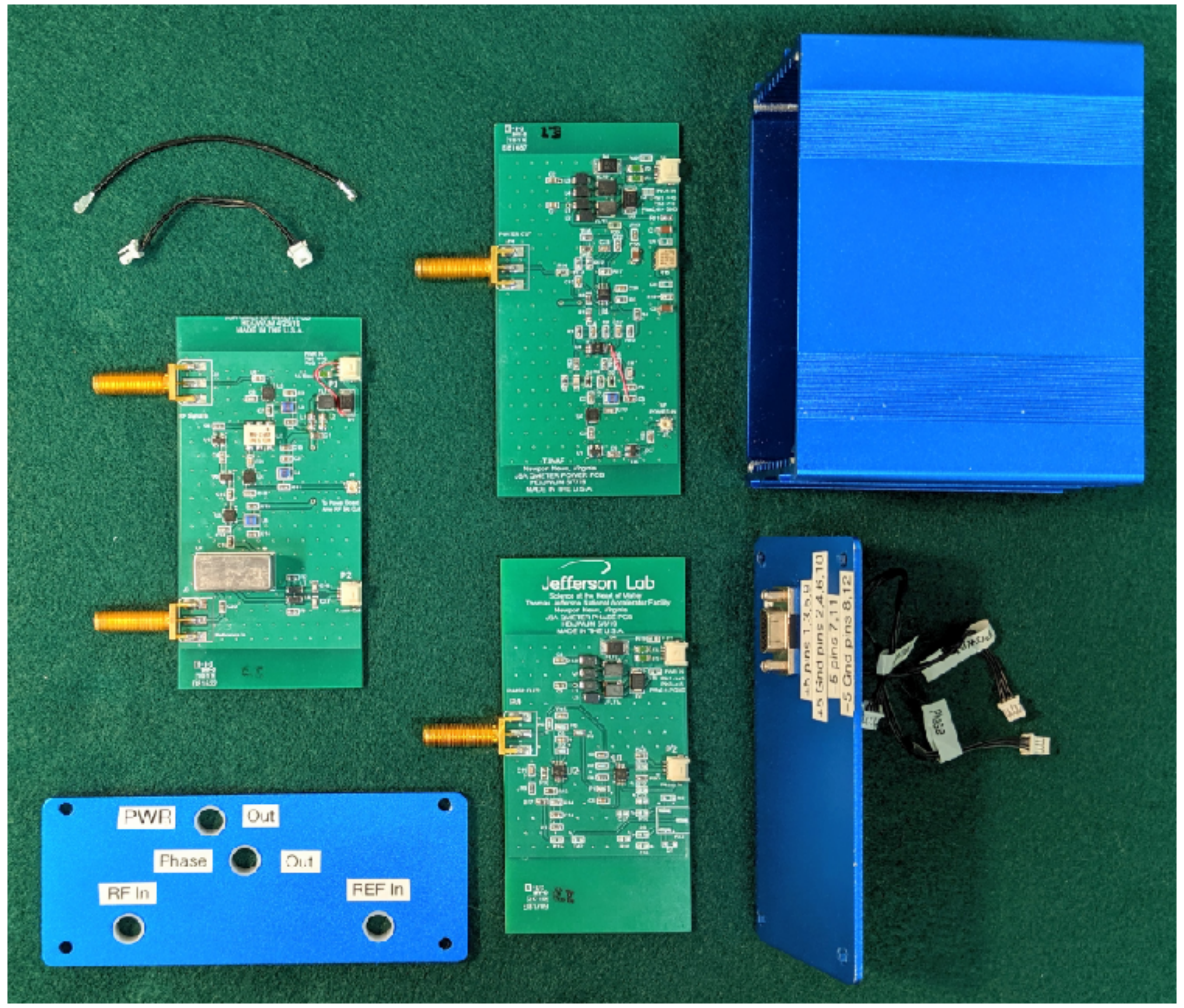
New Facility: IMAGINE-X for DNP-NMC

- NMR TE on extremely small samples is challenging
  - ORNL collaborates with JLab Target group to adopt and produce JLab Q-meters
  - 3-board design, cold-NMR ready
  - remote LCR tuning, remote phase tuning
- Potential improvements
  - variable RF amplification
  - cold first stage RF amplifier

Liverpool Q-meter ⟶ JLab Q-meter



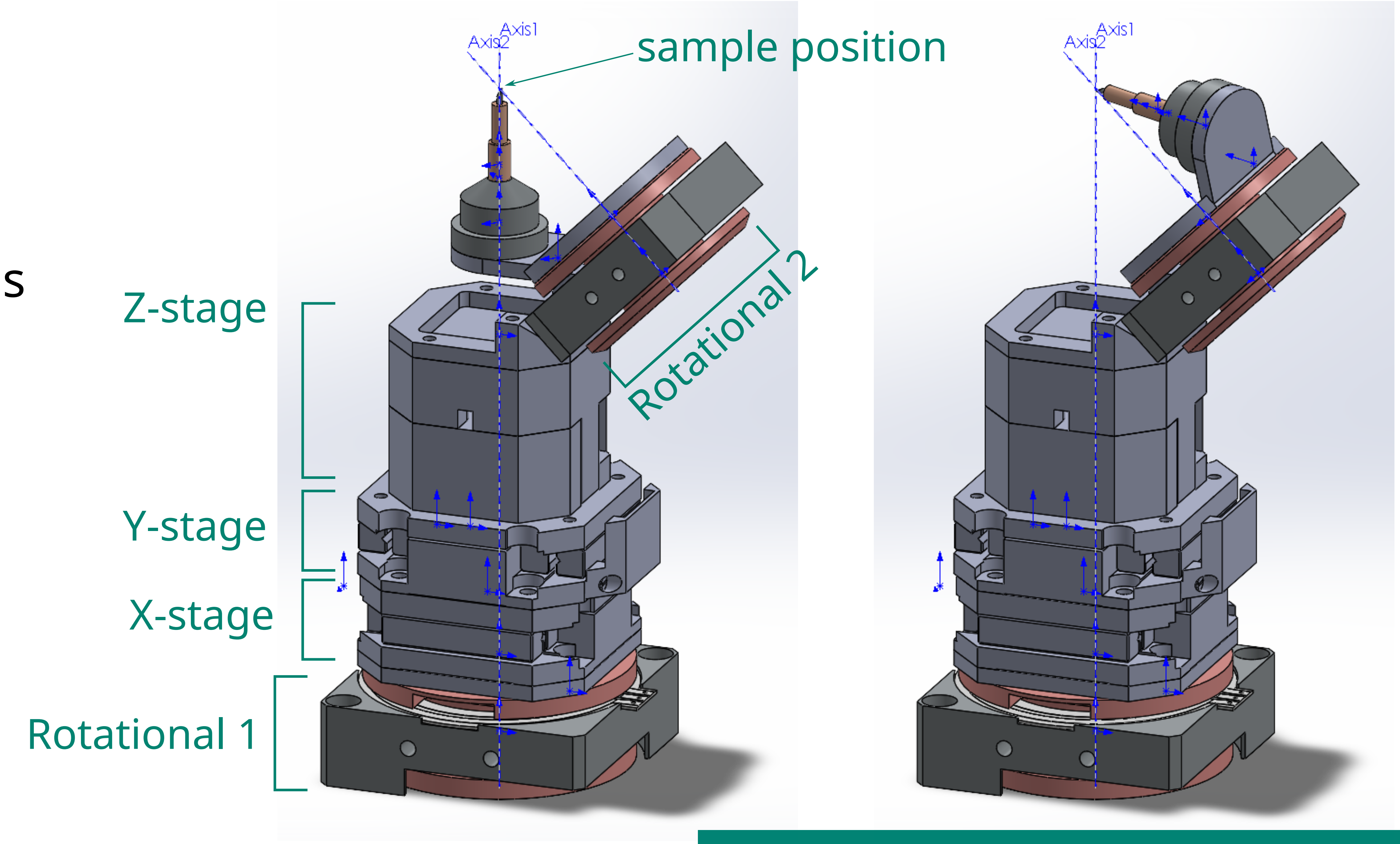




J. Maxwell, PSTP2019

New Facility: IMAGINE-X for DNP-NMC

- Multiple crystal orientations are required to reconstruct its structure
- Goniometry
  - ideally 3 translational and 3 rotational degrees of freedom
  - piezo-electrically activated stages from Attocube
  - preliminary design includes 5 stages



Outlook

- DNP-NMC has potential to dramatically improve signal-to-noise ratio of the conventional NMC and is powerful complementary tool to X-ray crystallographic studies
- Proof-of-concept measurements are very promising
  - sufficient sample polarization
  - 200-250mK temperature limited data collection time in frozen spin mode
  - factor of 2-3 increase of polarized diffraction pattern integrated intensity
- New DNP-NMC facility at HFIR - aggressive schedule
  - ongoing beamline + detector design and construction
  - expecting delivery of the magnet + cryostat in Aug. 2025