INSTITUTE FOR QUANTUM MATTER

A collaboration between
JOHNS HOPKINS UNIVERSITY
and PRINCETON UNIVERSITY

Neutron Scattering at High Magnetic Fields

Collin Broholm
Johns Hopkins University

JOHNS HOPKINS UNIVERSITY

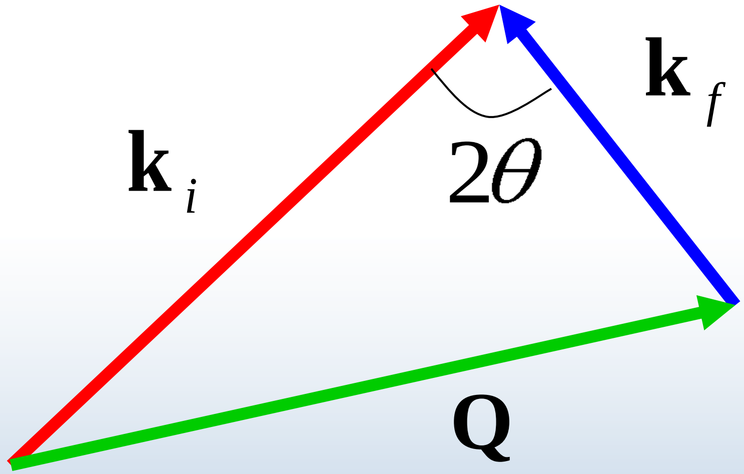
Overview

- **Neutron Scattering and high fields**
  - Probing atomic scale structure and dynamics
  - Controlling atomic scale properties
- **Past and present**
  - Experimental capabilities
  - Scientific impact
- **Recent developments & the future**
  - Magnet technology
  - Experimental capabilities
  - Scientific opportunities
- **Conclusions**

High fields and neutron scattering

- **Neutron Scattering: Probe matter**
  - Well defined interaction with nuclei and electrons
  - Structural information on 0.1 Å to 10^4 Å length scale
  - Dynamic information 10^{-9} eV to 1 eV
- **High magnetic field: Control matter**
  - Controlled perturbation of magnetizable matter
  - Produces new correlated states of matter
  - Affects nuclear spin polarization and resonance
  - Produce conditions that facilitate analysis of structure and dynamics
- **Combine to control & probe**
  - Demonstrated strong scientific productivity 0-15 T
  - Technical developments now bring 30-40 T into view
  - Brighter neutron sources offer greater details

One Simple Experiment



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

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

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

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

One versatile probe of materials

- **Atomic Scale and Microscopic Structure**
  - Crystalline structure associated with light atoms
  - Non-periodic super-structures in solids
  - Magnetic Structure
  - Structure in solution on 10-1000 nm length scales
  - Flux line lattice structure
  - Magnetic surface structure
  - Structure at buried interfaces
- **Atomic Scale Dynamics**
  - Lattice vibrations (phonons) in crystalline solids
  - Librations and "Neutron-Raman" spectroscopy
  - Magnetic Excitations
- **Macroscopic Imaging**
  - Residual stress within the bulk of structural object
  - Neutron vision: imaging light atoms within heavy matrix
  - Neutron activation analysis for compositional analysis

Bragg Diffraction

SANS

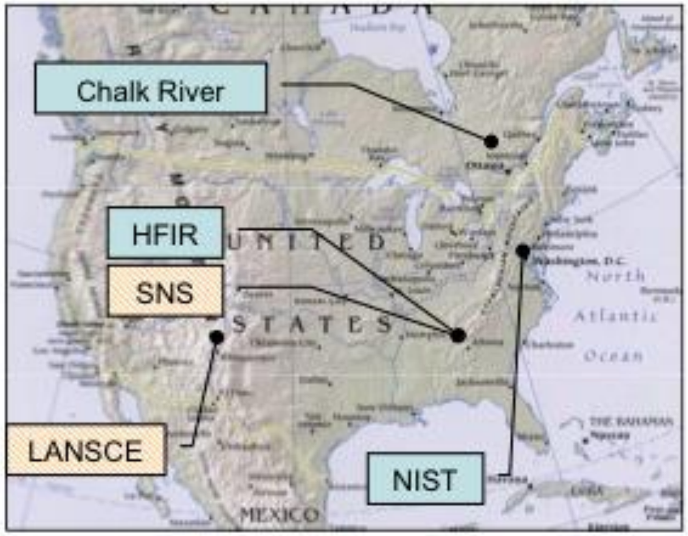
Reflectometry

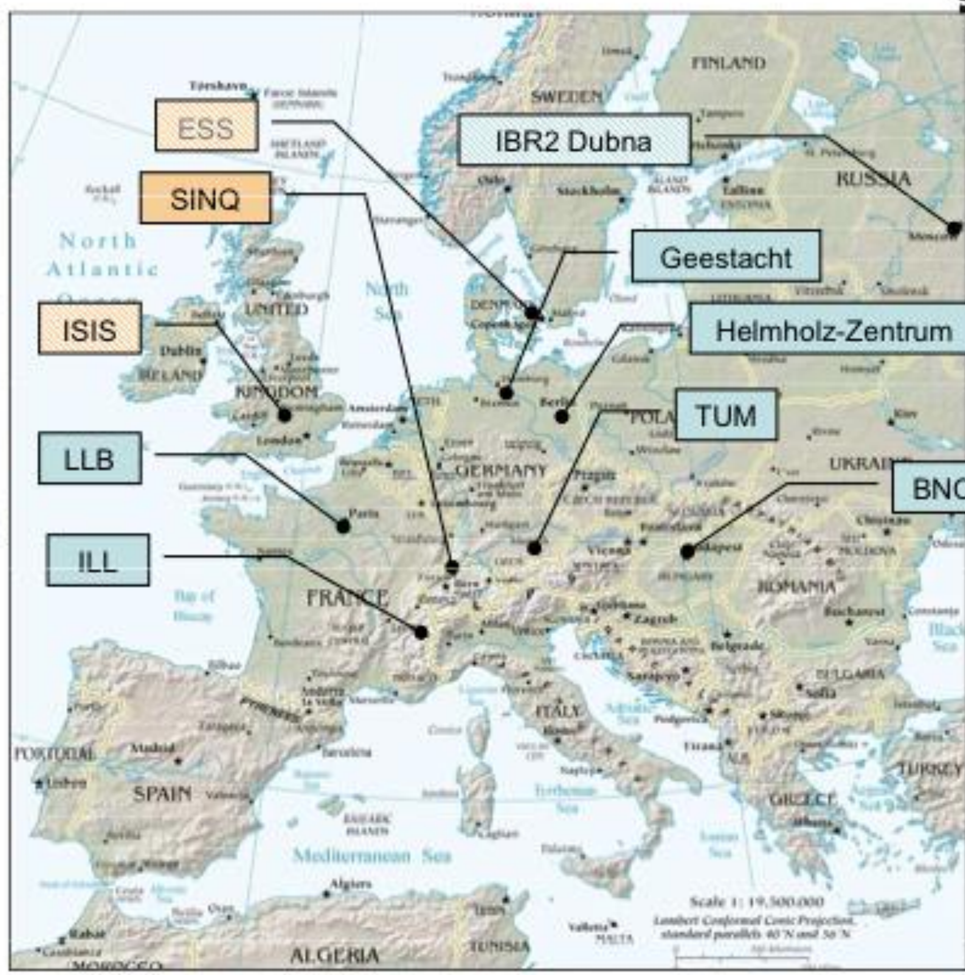
Spectroscopy

Diffraction shift

Radiography

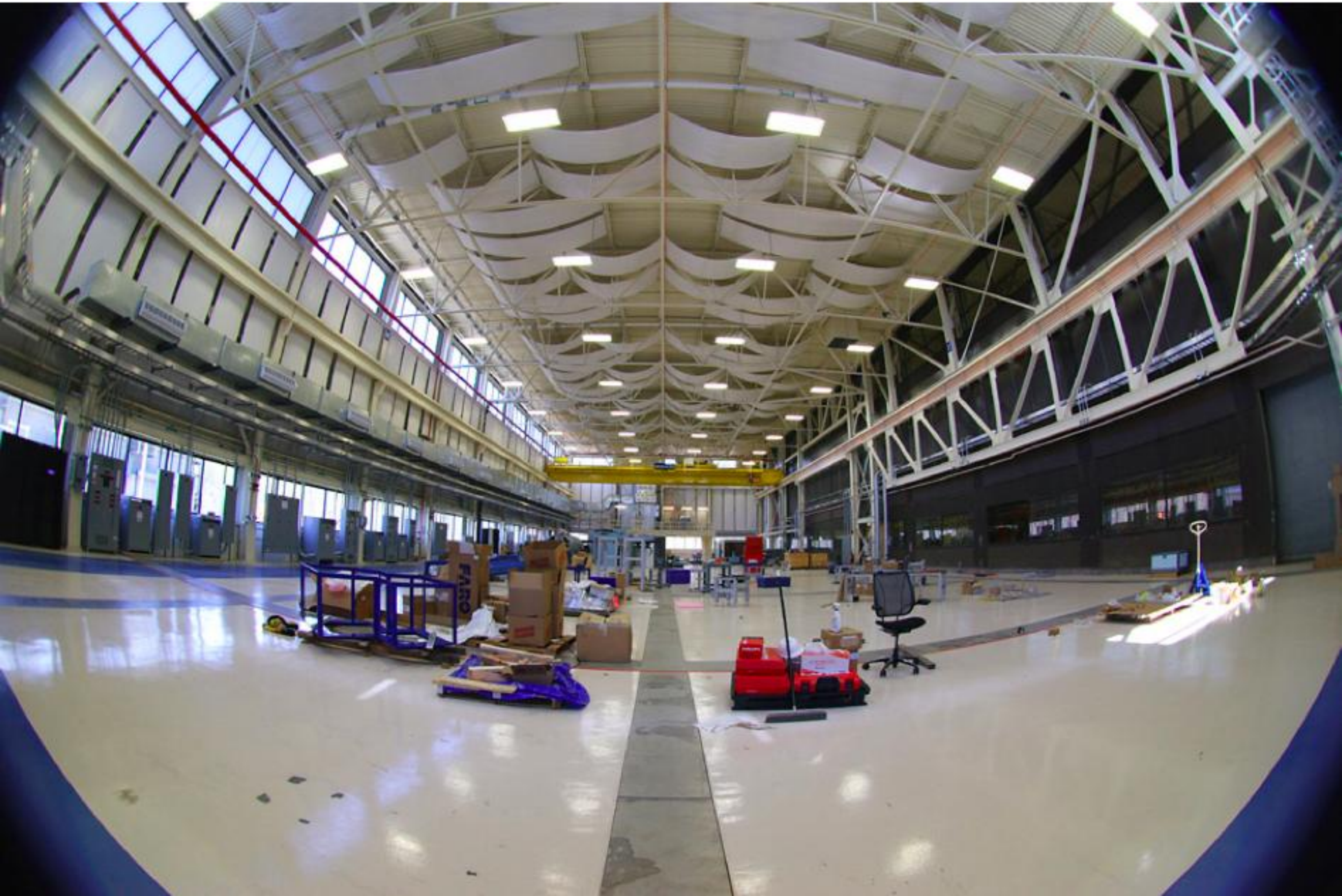
Famous operational neutron sources

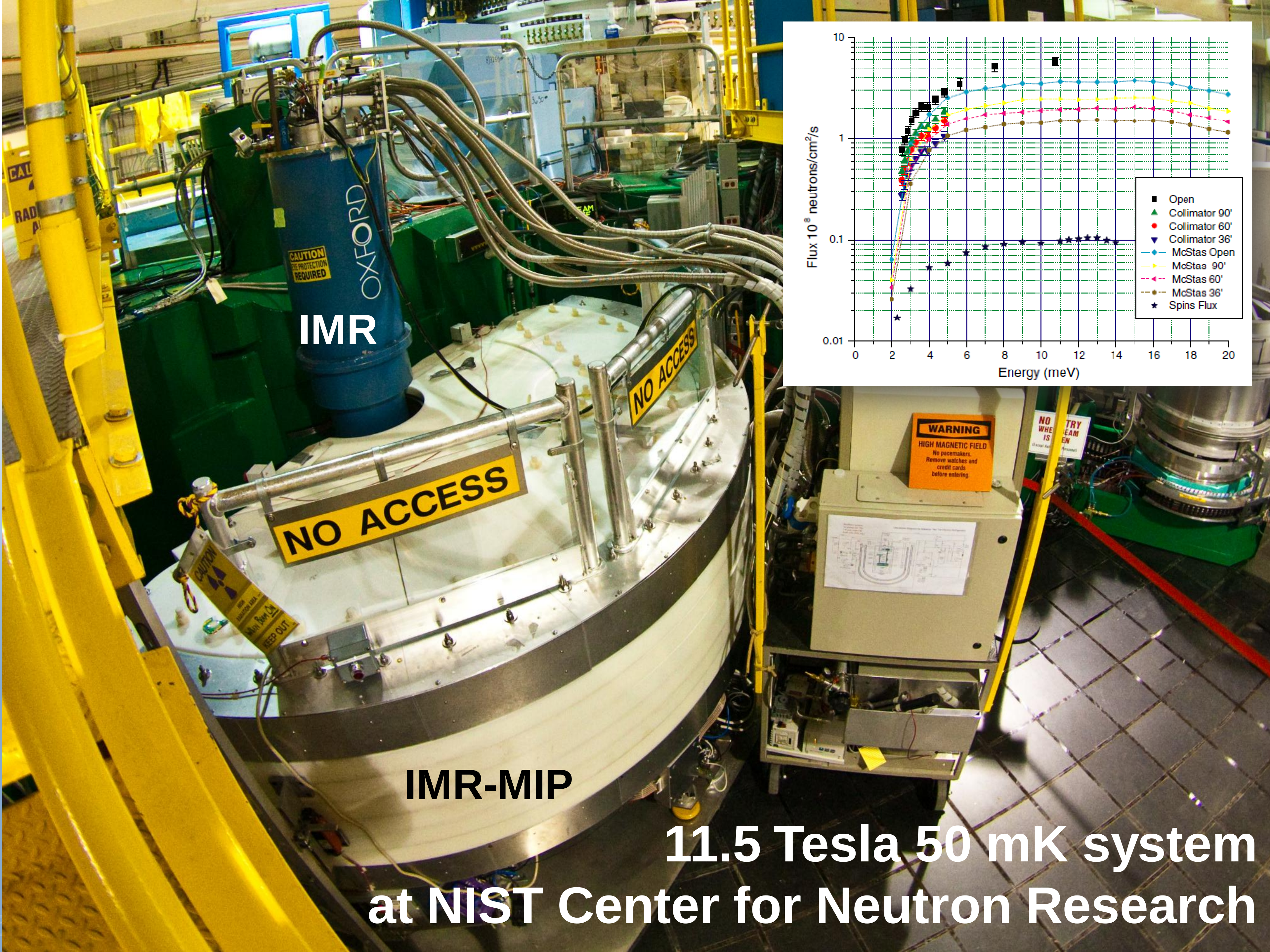




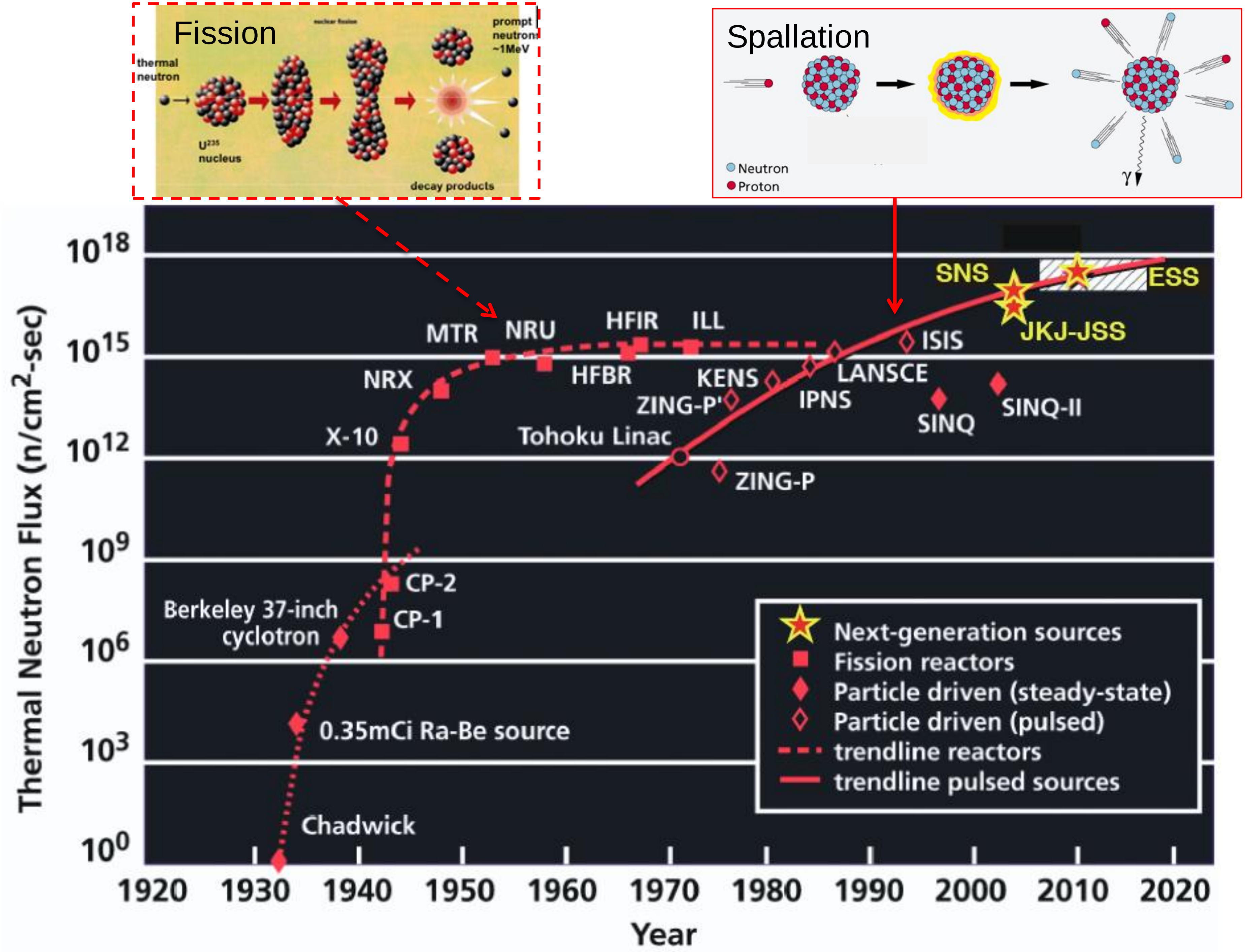
Neutron Scatterers Map of North America

New guide hall at NIST



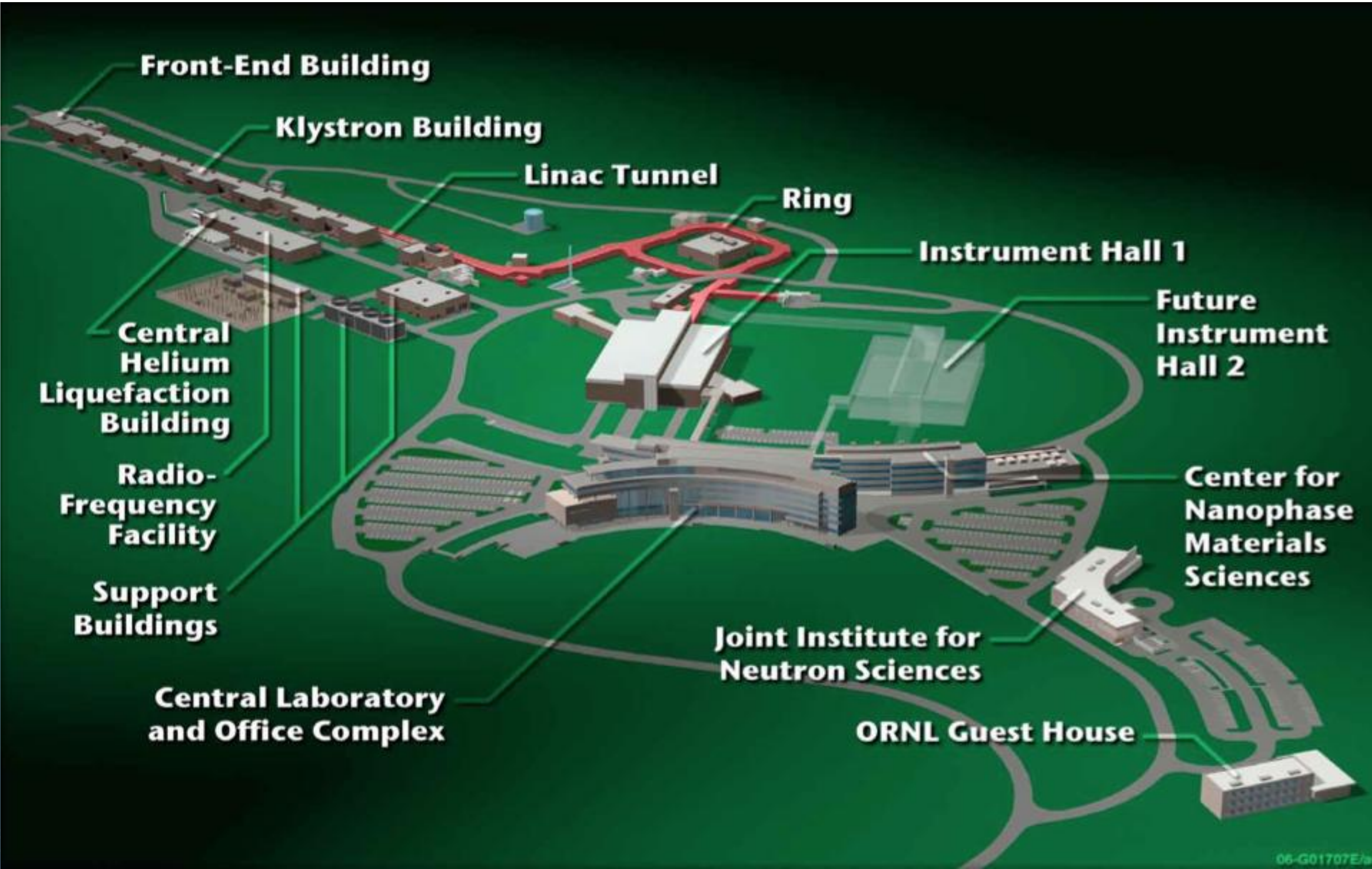


11.5 Tesla 50 mK system
at NIST Center for Neutron Research

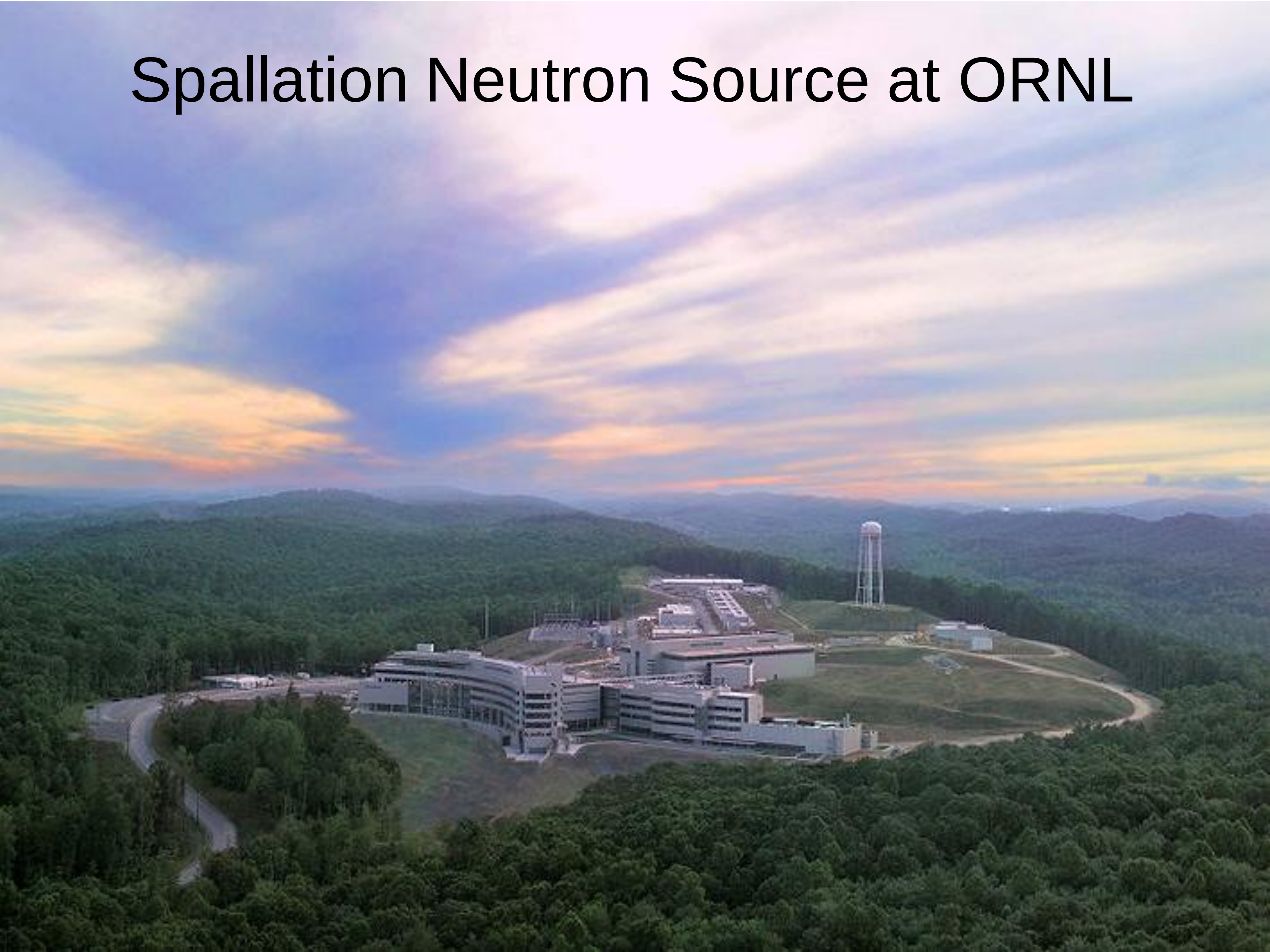


Fission
thermal neutron
U235 nucleus
prompt neutrons ~1MeV
decay products
Spallation
Neutron
Proton
γ
Thermal Neutron Flux (n/cm2-sec)
10^18
10^15
10^12
10^9
10^6
10^3
10^0
Year
1920
1930
1940
1950
1960
1970
1980
1990
2000
2010
2020
Chadwick
0.35mCi Ra-Be source
Berkeley 37-inch cyclotron
CP-1
CP-2
X-10
NRX
MTR
NRU
HFBR
HFIR
ILL
Tohoku Linac
ZING-P
ZING-P'
KENS
IPNS
LANSCE
ISIS
SINQ
SINQ-II
SNS
JKJ-JSS
ESS
Next-generation sources
Fission reactors
Particle driven (steady-state)
Particle driven (pulsed)
trendline reactors
trendline pulsed sources

The Spallation Neutron Source @ ORNL

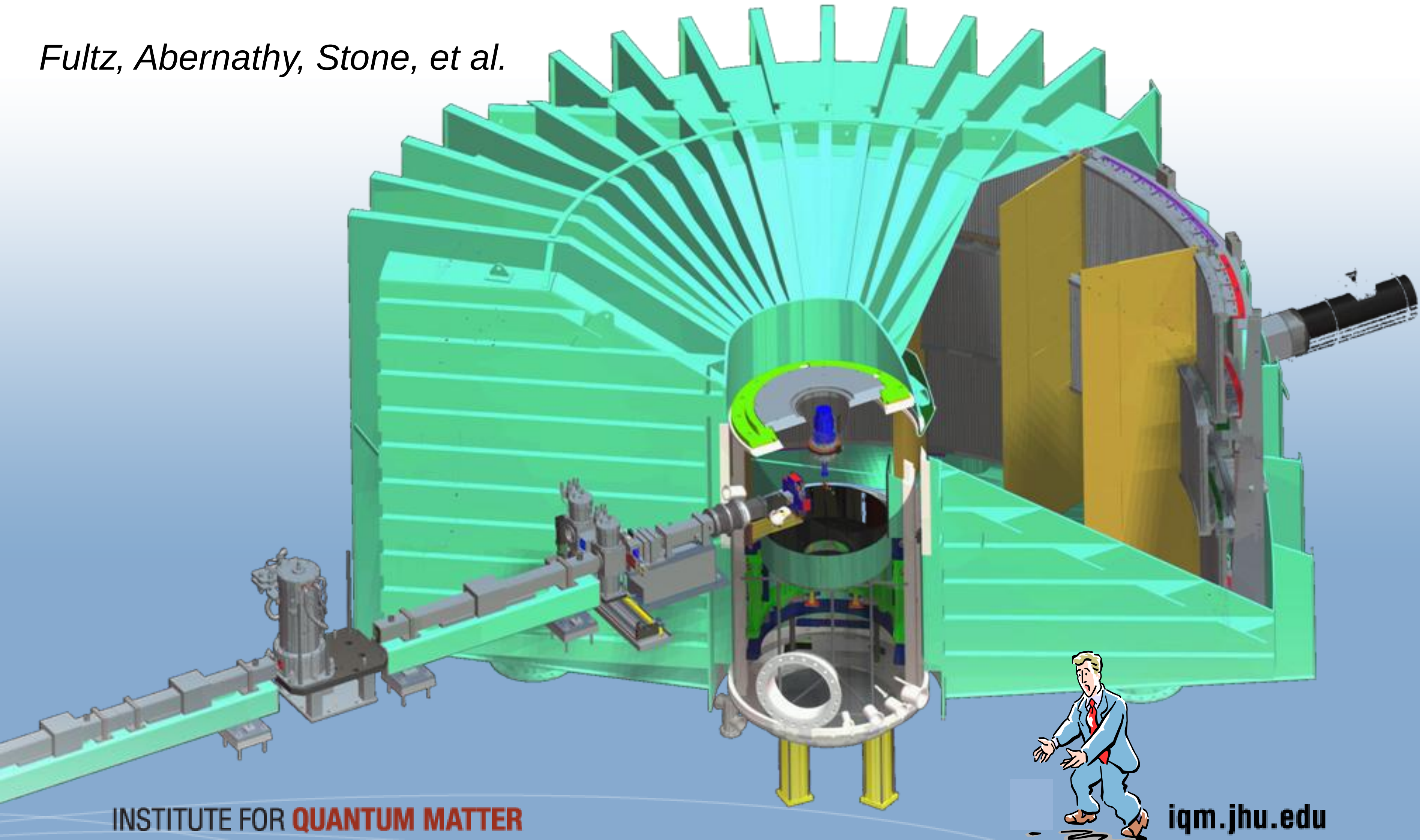


Spallation Neutron Source at ORNL

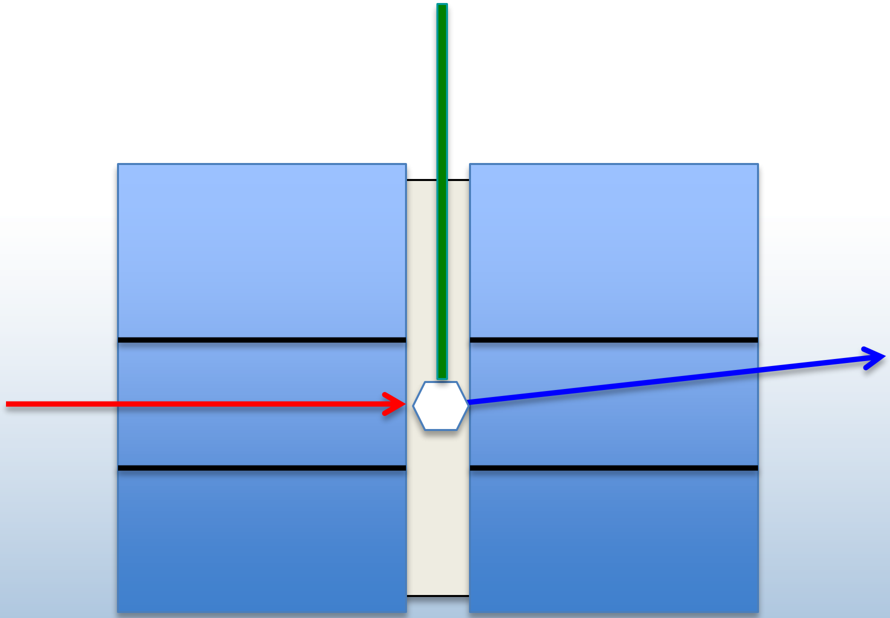


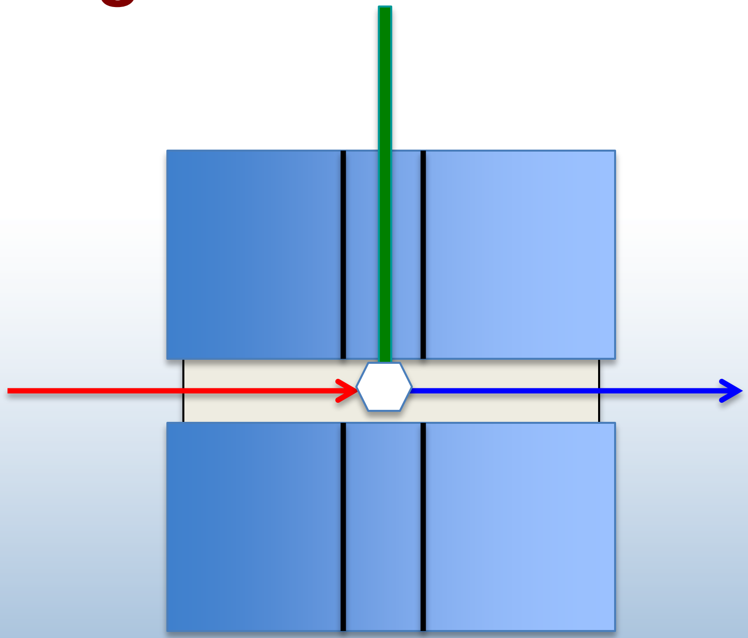
ARCS Spectrometer at SNS

Fultz, Abernathy, Stone, et al.



Geometries of Superconducting magnets for neutron scattering





Horizontal field geometry

- Up to 17 Tesla
- Sample typically introduced through split
- Low background for SANS
- Limited angular access

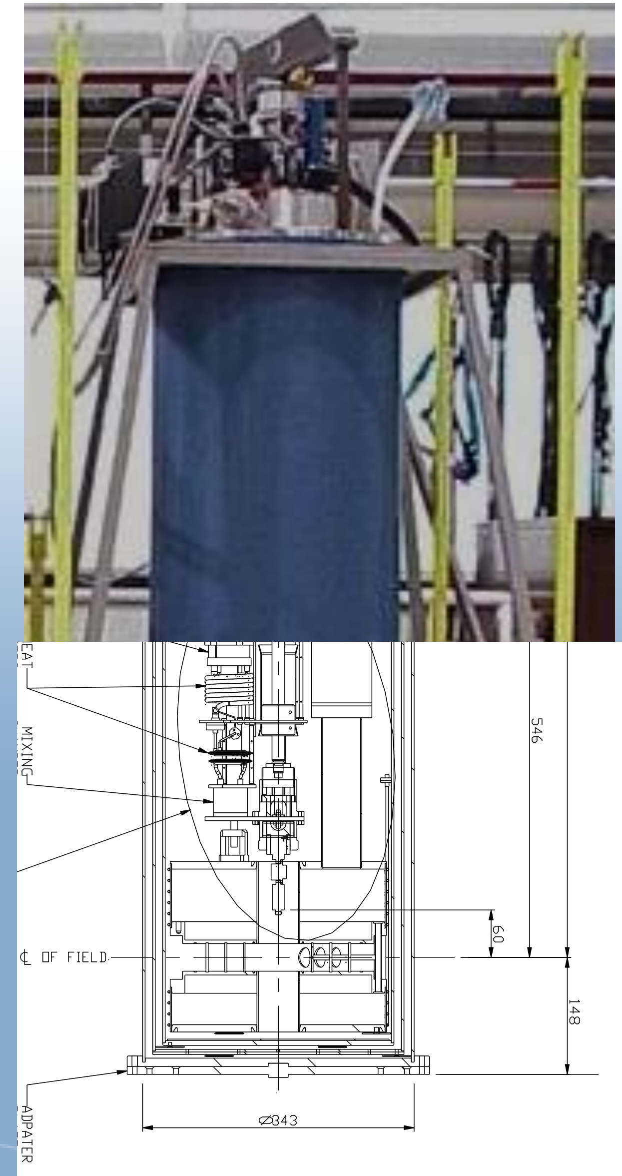
Vertical field geometry

- Up to 16 Tesla (18 T with poles)
- Sample introduced through bore
- Beam passes through structural rings
- 360 degree angular access
- Limited vertical beam divergence

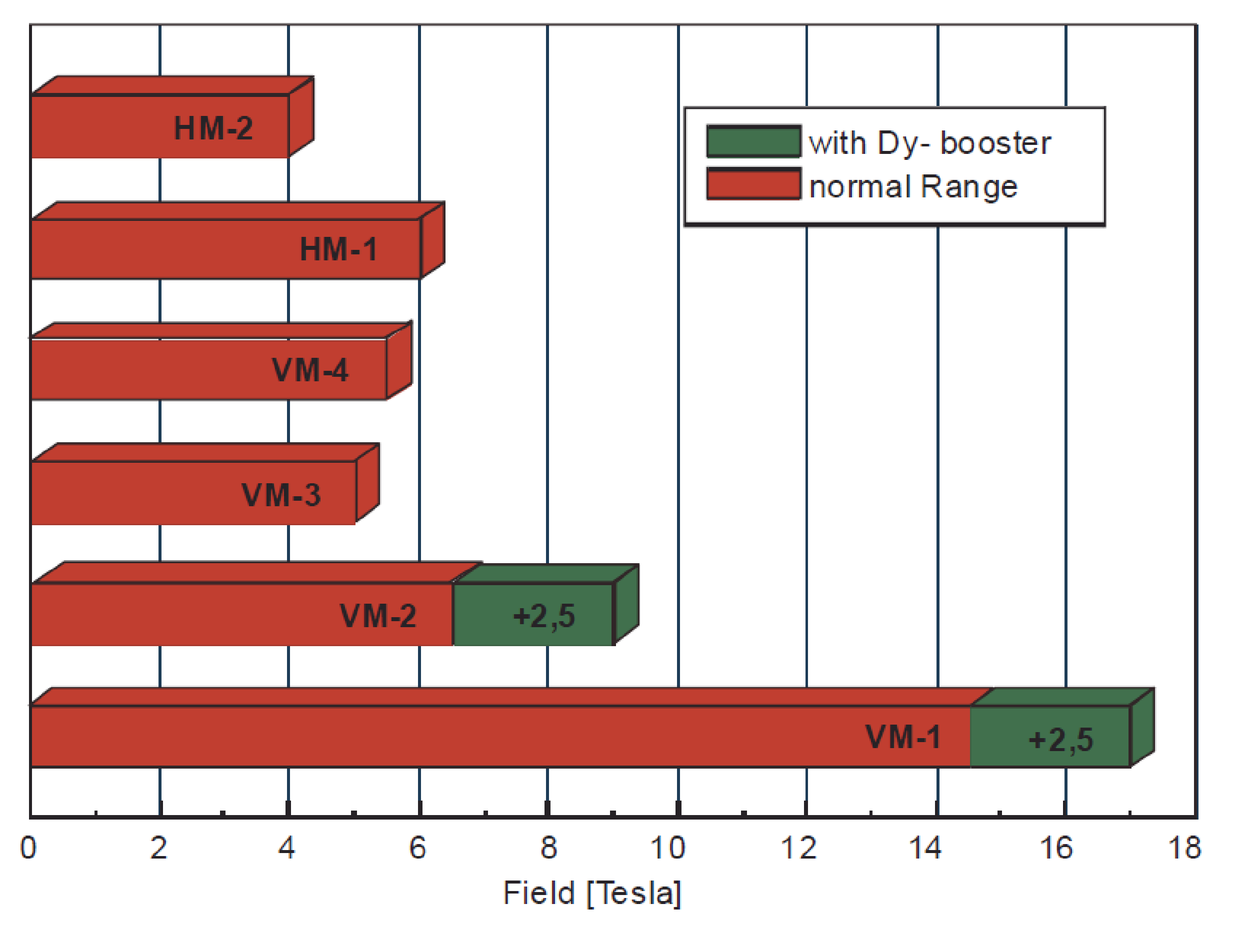
Split coil superconducting magnets for neutron scattering

17 Tesla horizontal field Birmingham Univ.

11.5 Tesla vertical field @ NIST



High field magnets at HZB, Berlin

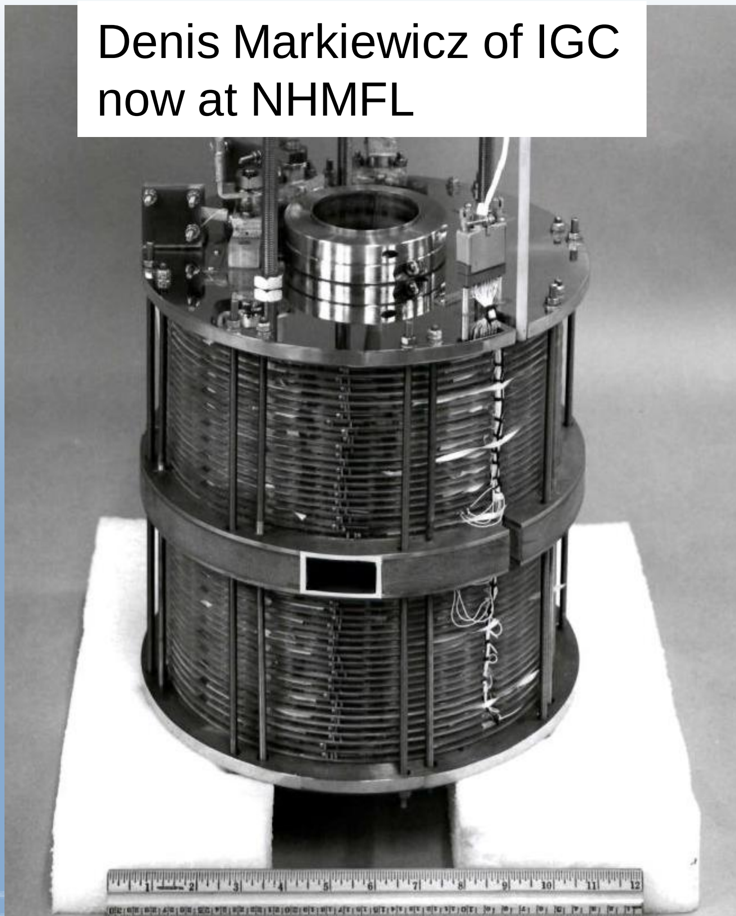


The Helmholtz Zentrum Berlin has strengthened it's neutron scattering program by specializing in extreme sample environments

16 T compensated magnet at SNS

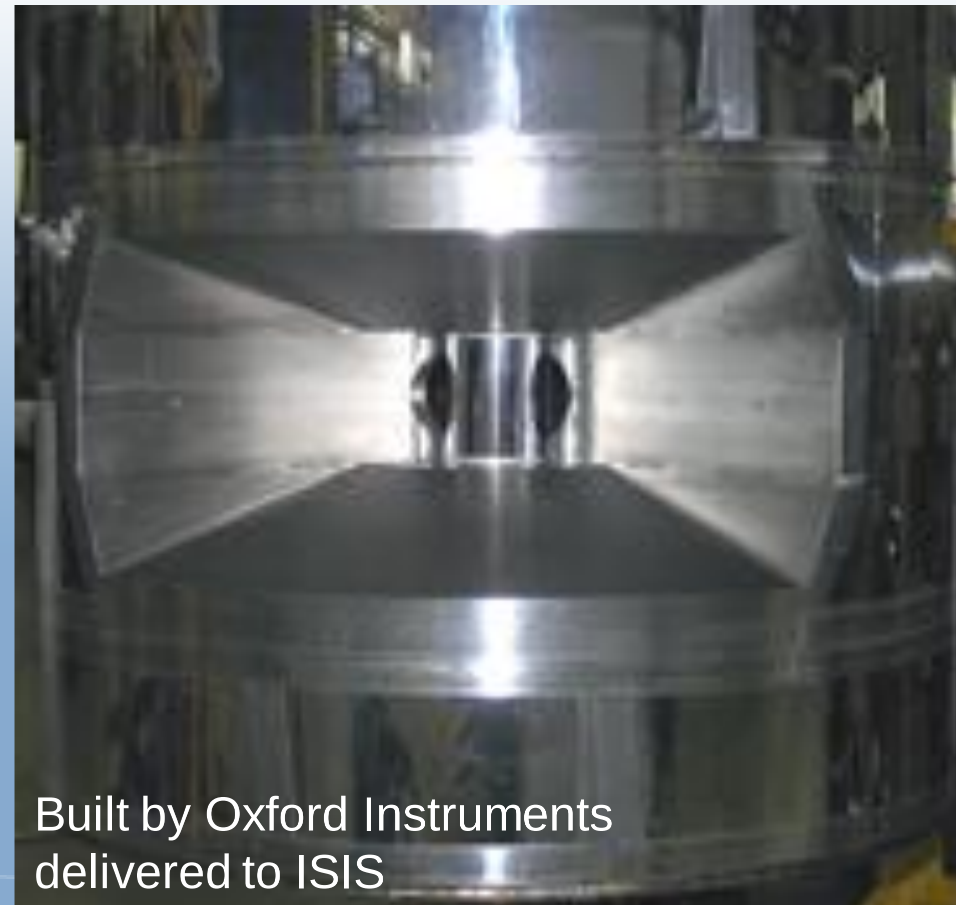
Improved access for scattering

1978:
13 Tesla, 15 mm split



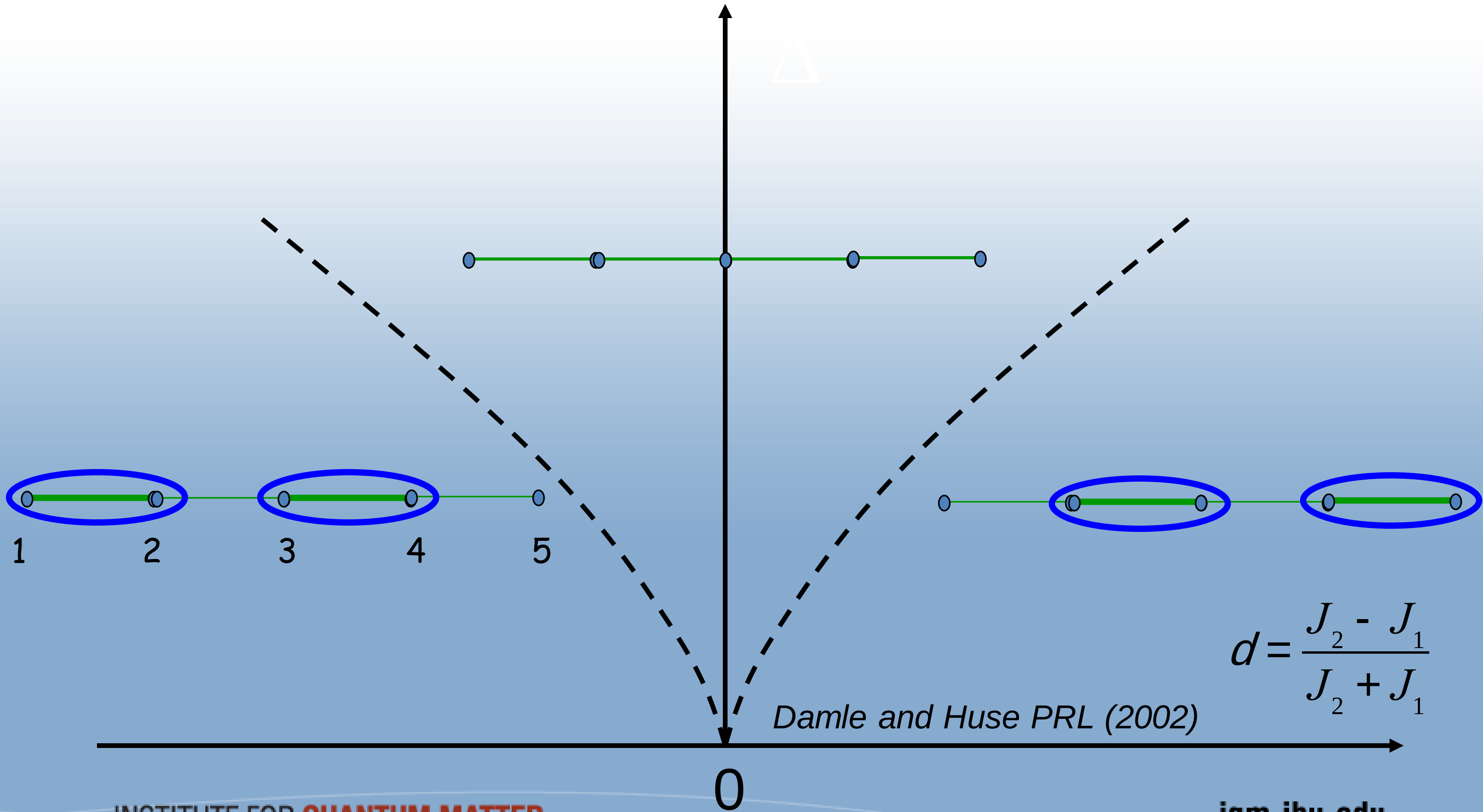
Denis Markiewicz of IGC now at NHMFL

2009:
9 Tesla, ±15 degrees access



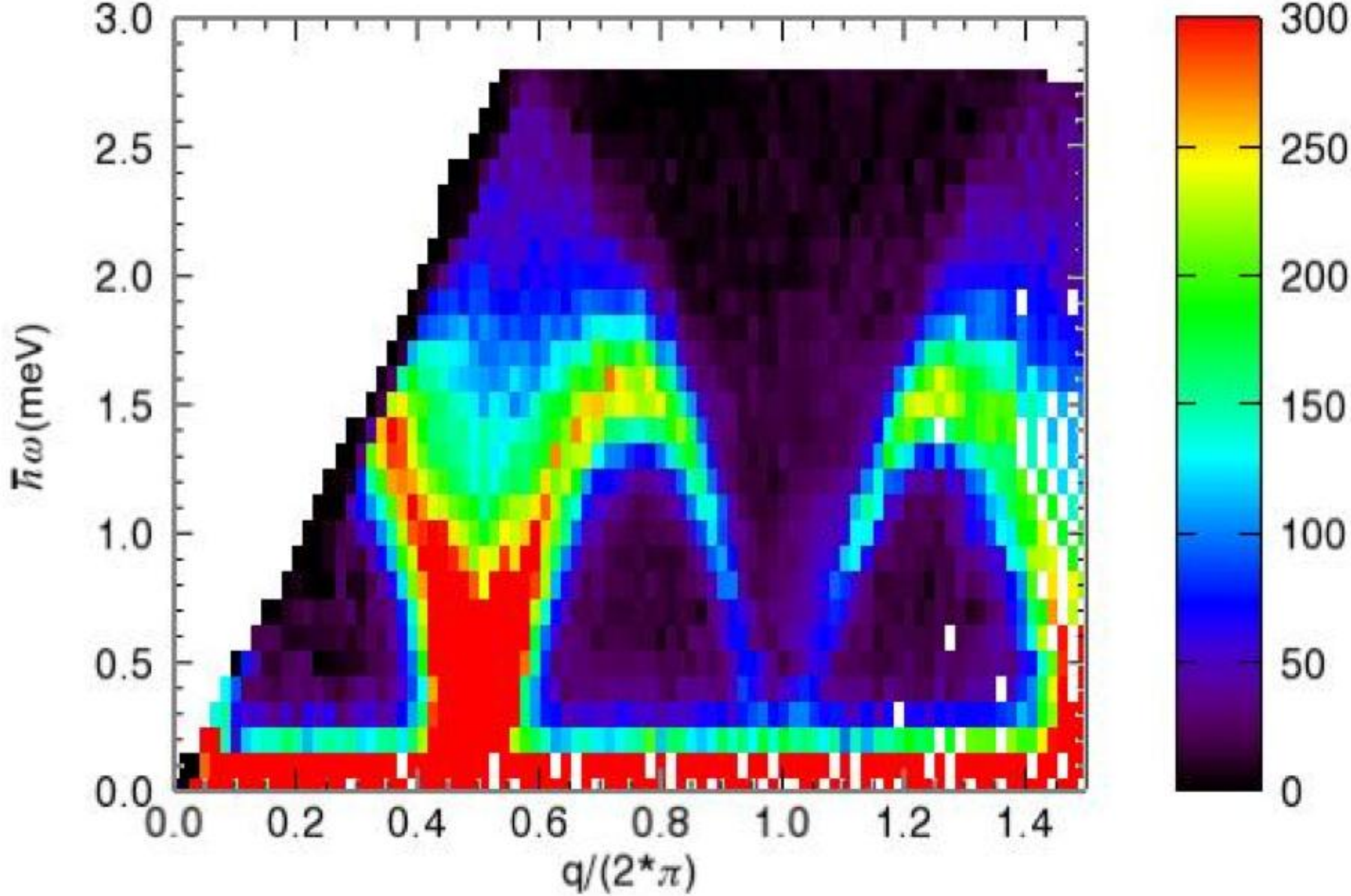
Built by Oxford Instruments delivered to ISIS

Quantum Critical Spin-1/2 chain



$$d = \frac{J_2 - J_1}{J_2 + J_1}$$

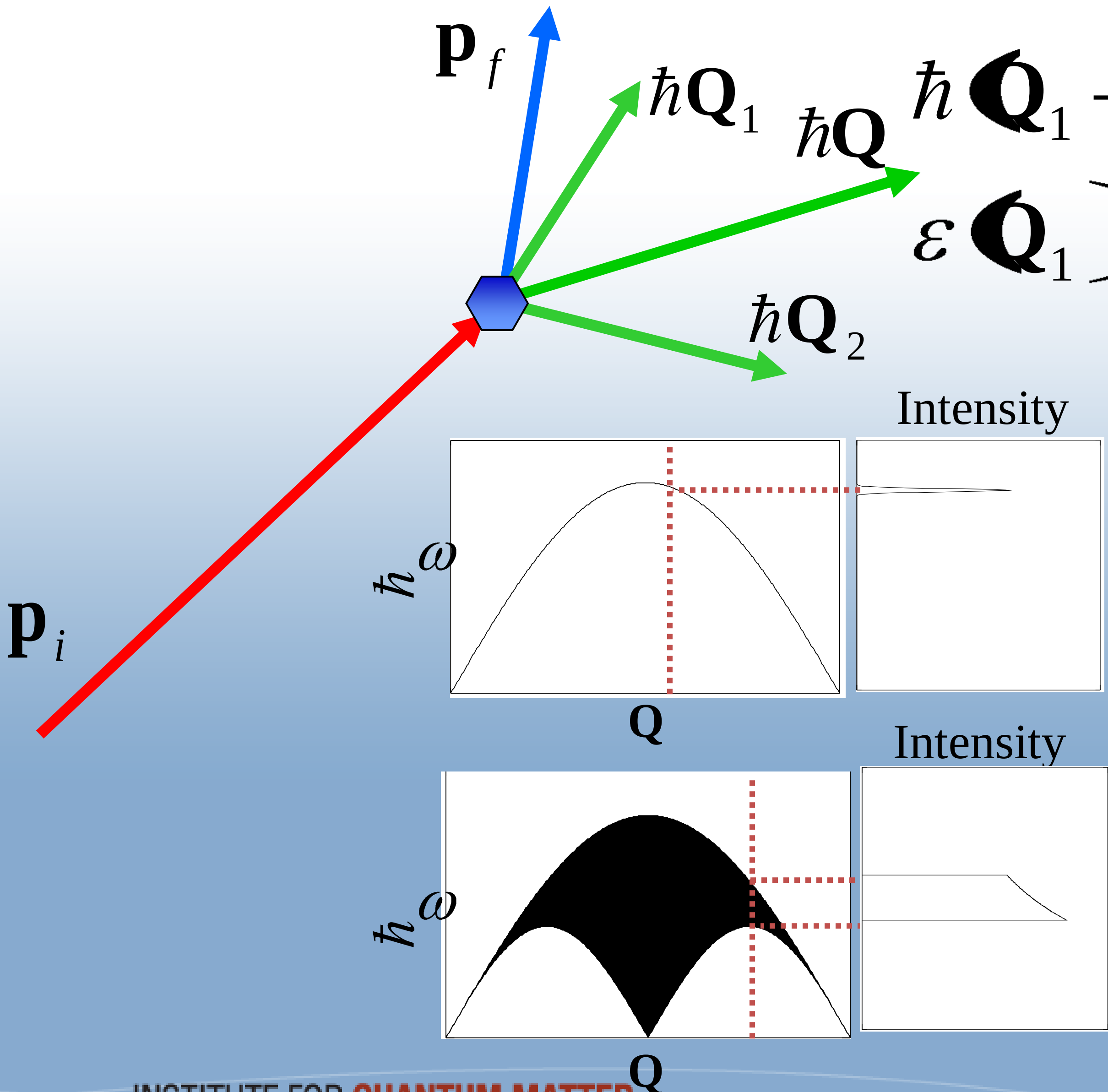
Copper Pyrazine dinitrate H_z=0



Probing matter through scattering

$$\hbar\left(\mathbf{Q}_1+\mathbf{Q}_2\right)=\mathbf{p}_i-\mathbf{p}_f \qquad \hbar\mathbf{Q}=\mathbf{p}_i-\mathbf{p}_f$$

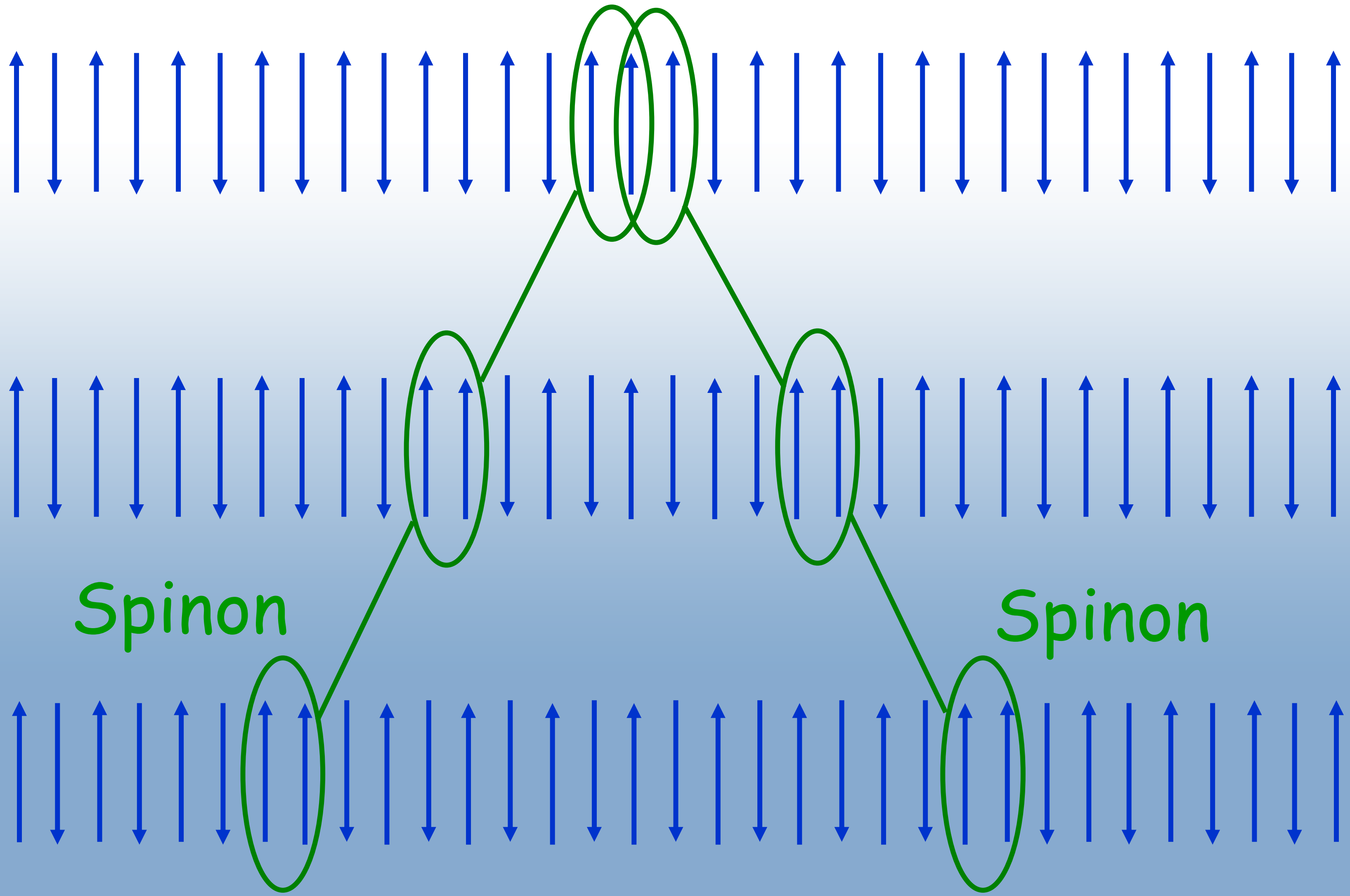
$$\varepsilon\left(\mathbf{Q}_1\right)+\varepsilon\left(\mathbf{Q}_2\right)=E_i-E_f \qquad \varepsilon\left(\mathbf{Q}\right)=E_i-E_f$$



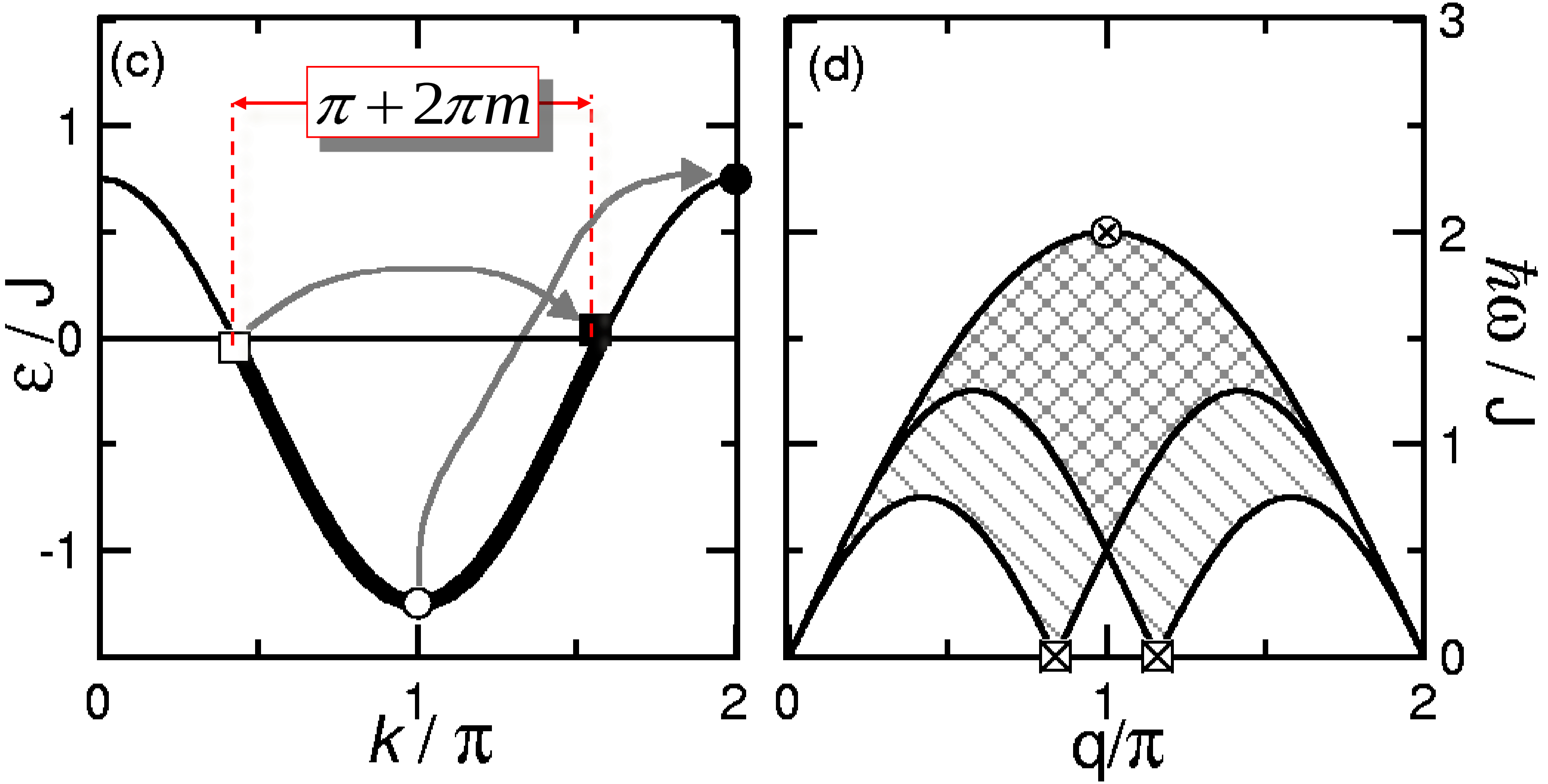
Single Particle Process

Multi Particle Process

Disintegration of a spin flip

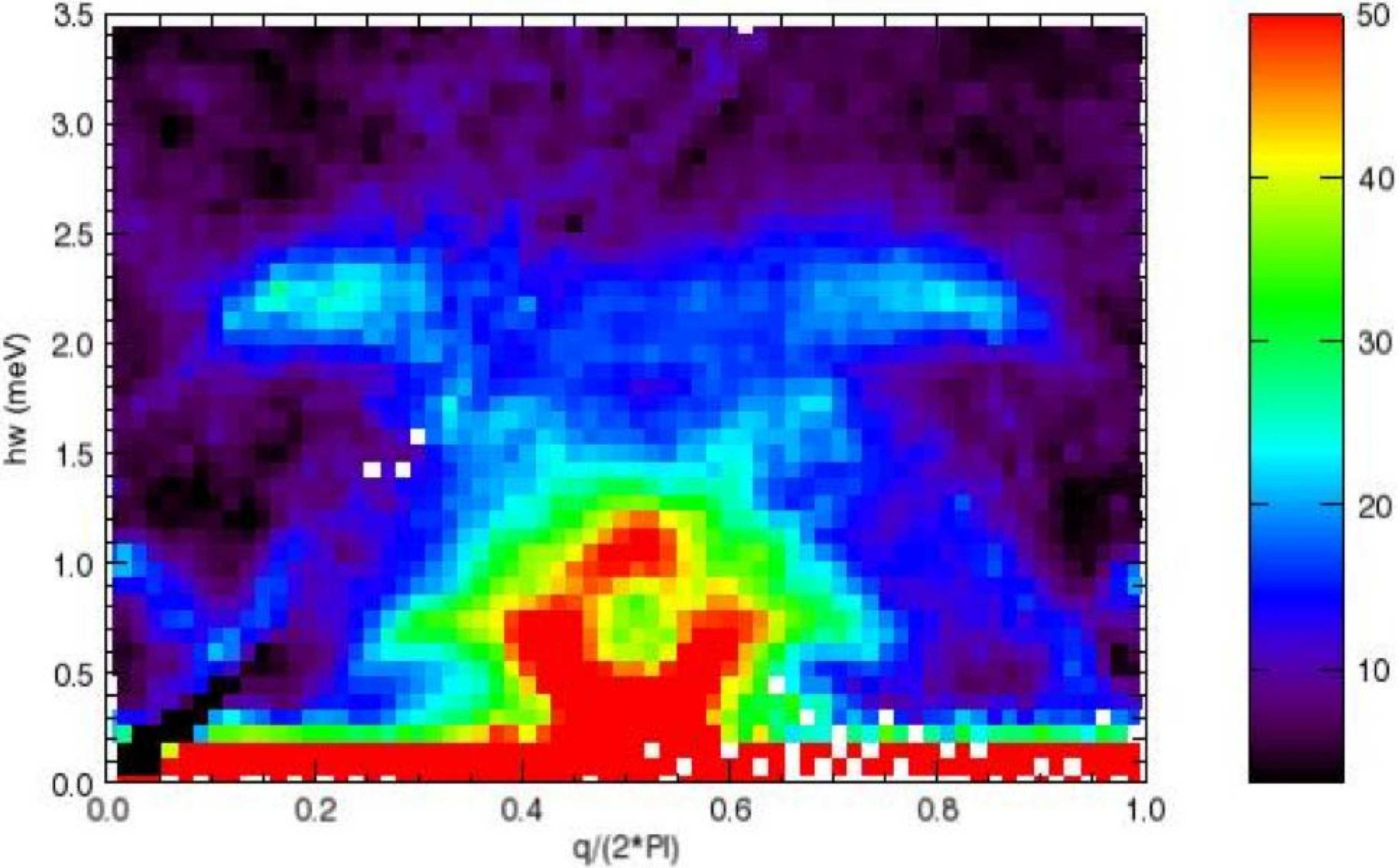


Spinons in magnetized spin-½ chain



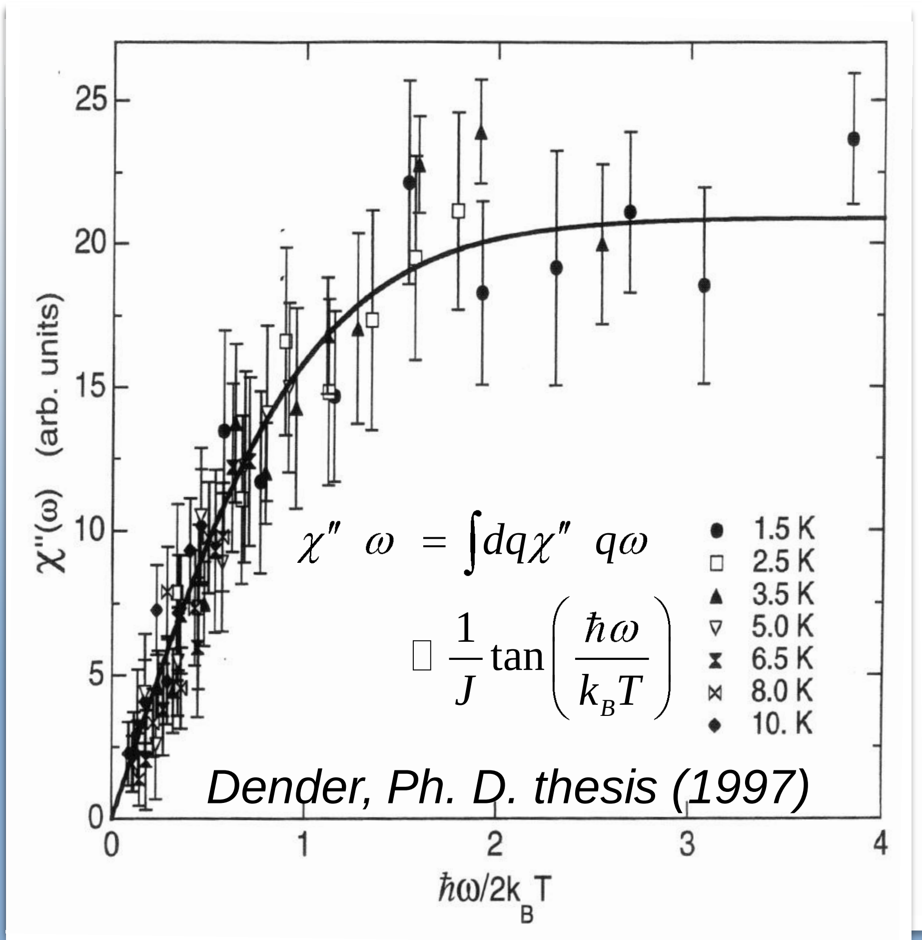
(XY case for simplicity)

Copper Pyrazine dinitrate $\mu_0 H_z$=8.5 T



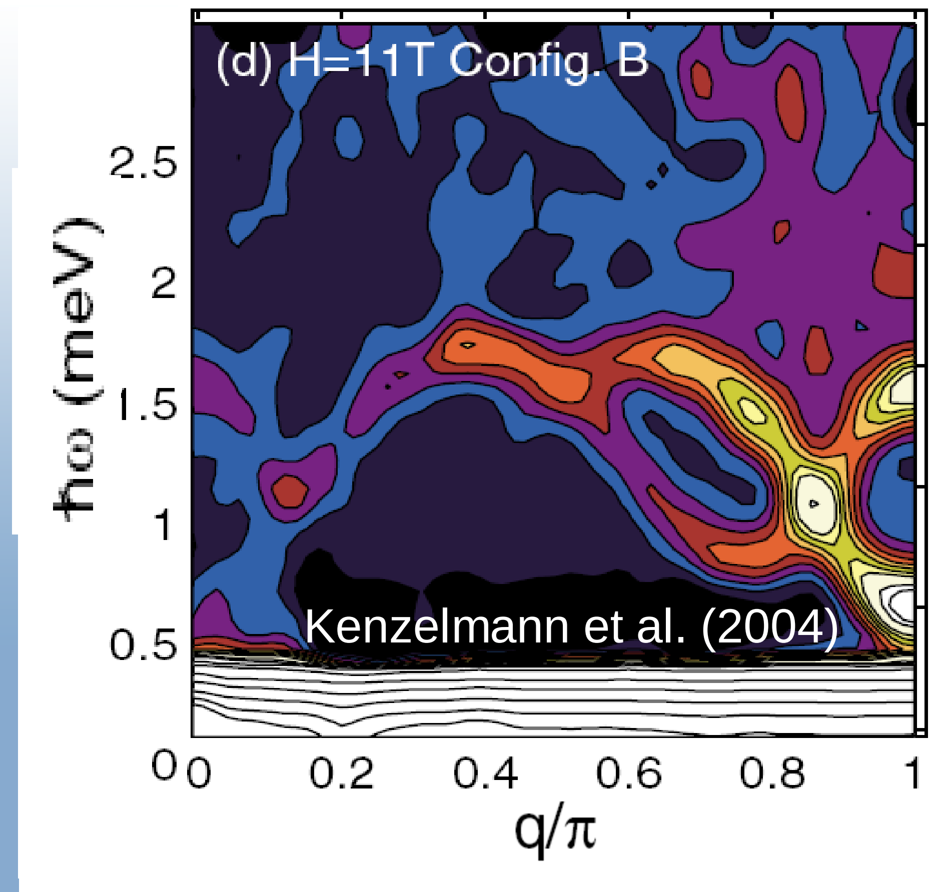
Quantum Criticality in spin-1/2 chain

Scaling



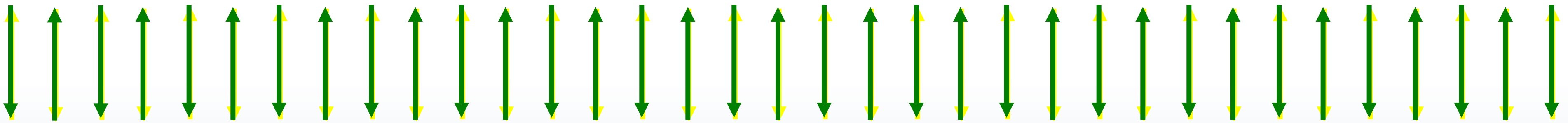
$$\chi''(\omega) = \int dq\, \chi''(q\omega) \sim \frac{1}{J}\tan\left(\frac{\hbar\omega}{k_B T}\right)$$

Sensitivity to perturbations:
Staggered field

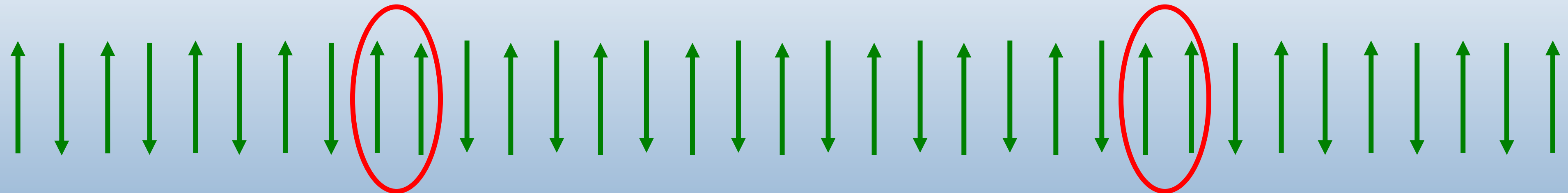


Why staggered field yields bound states

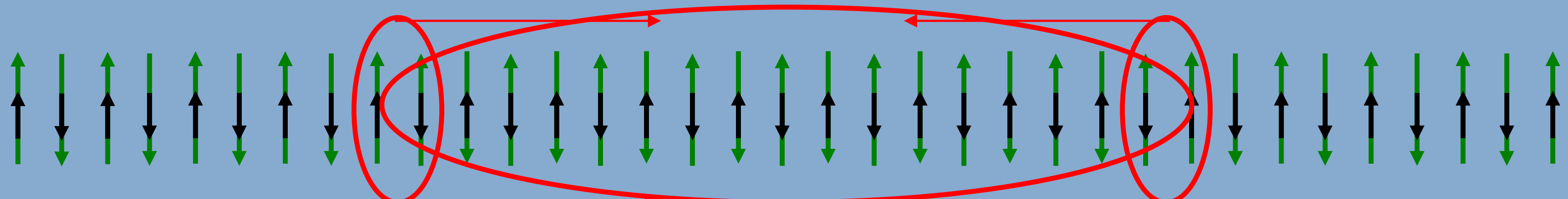
Zero field state quasi-long range AFM order



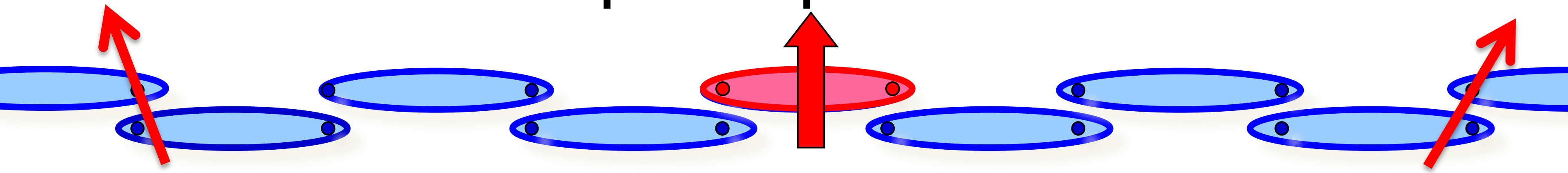
Without staggered field distant spinons don't interact

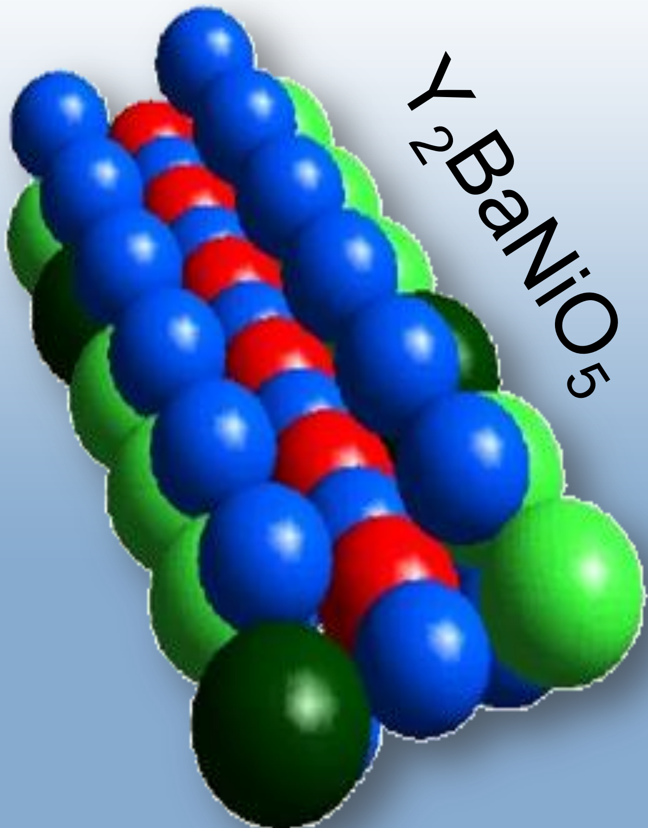


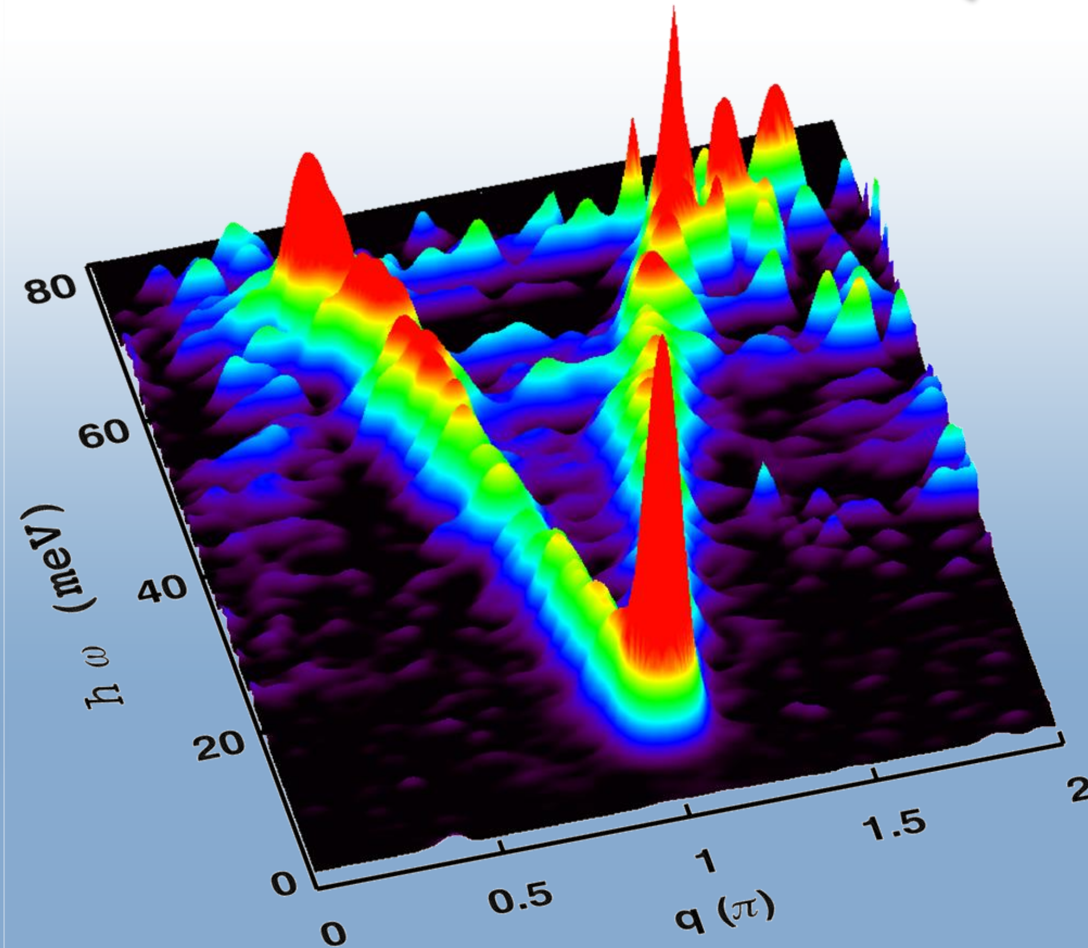
With staggered field solitons separate "good" from "bad" domains, which leads to interactions and "soliton" bound state



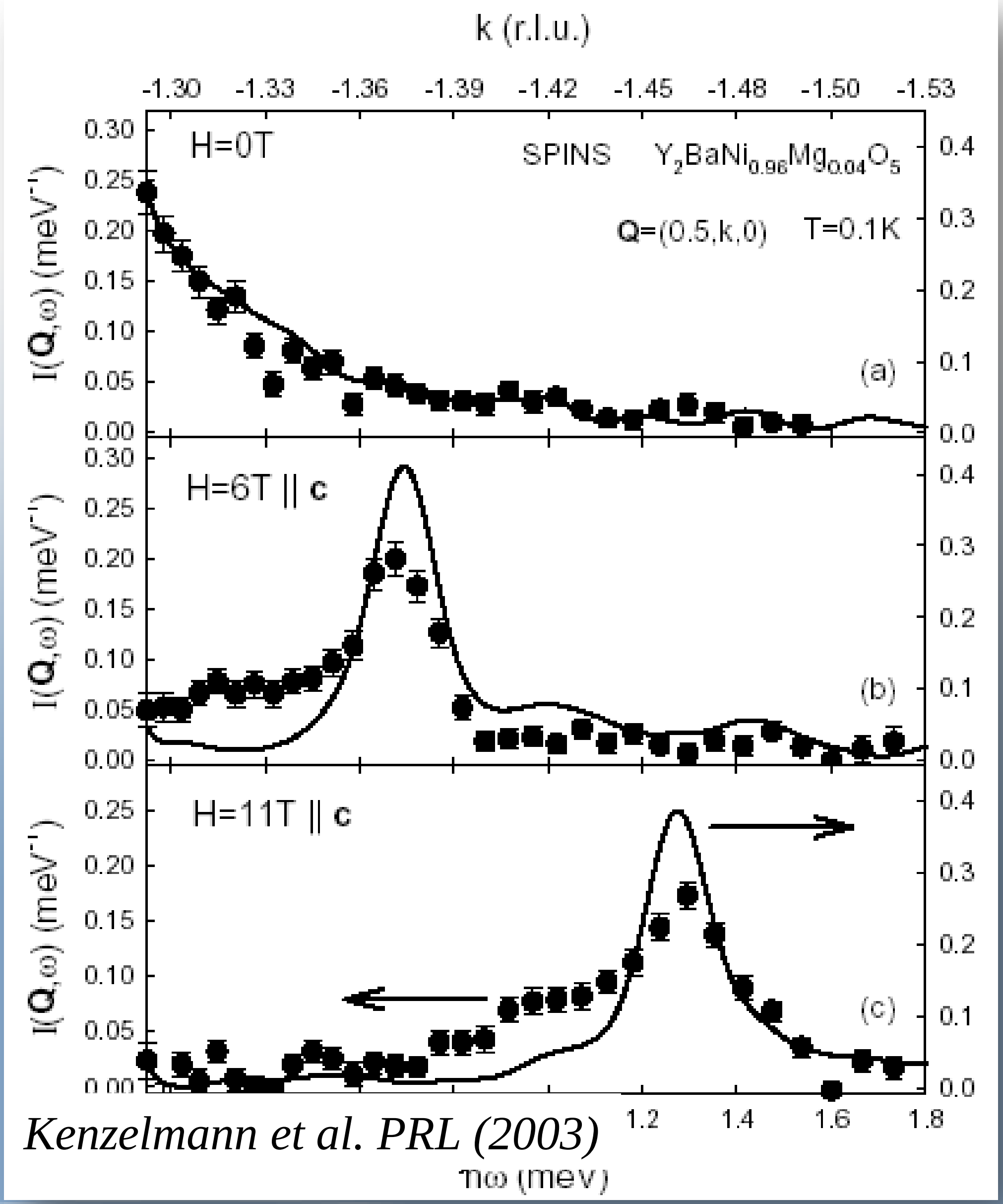
Collective Gap in Spin-1 AFM Chain





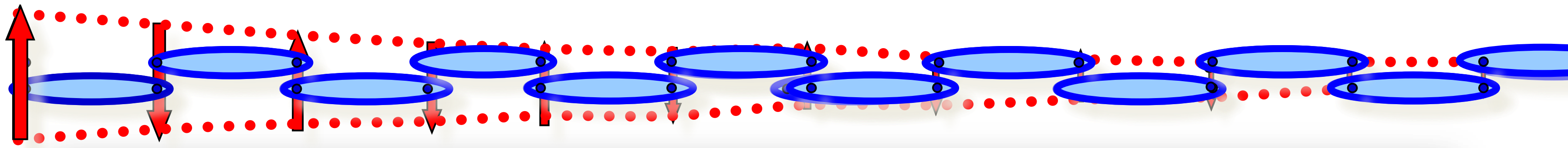


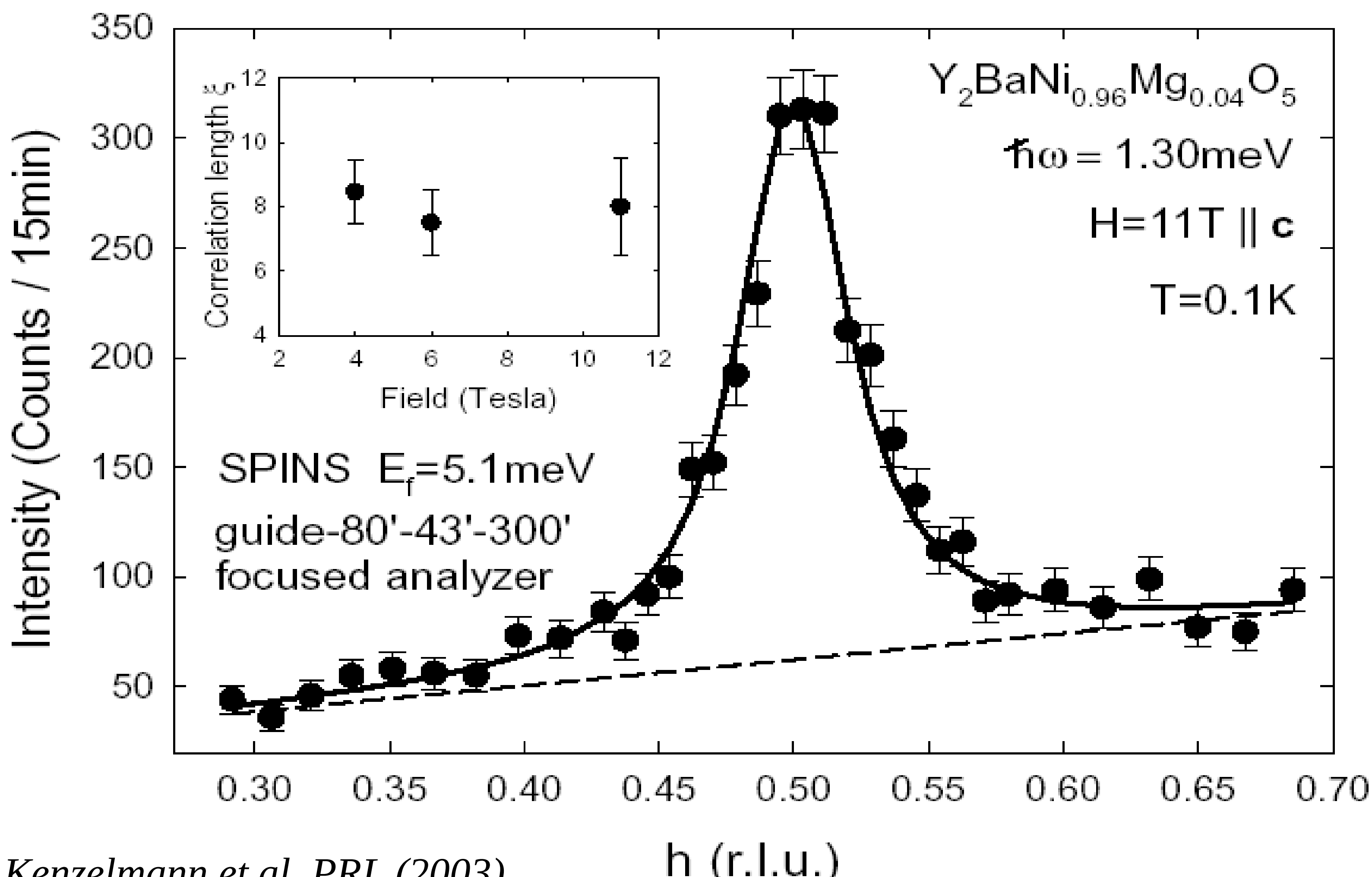
ESR with neutrons



Kenzelmann et al. PRL (2003)

Form factor for chain-end spin

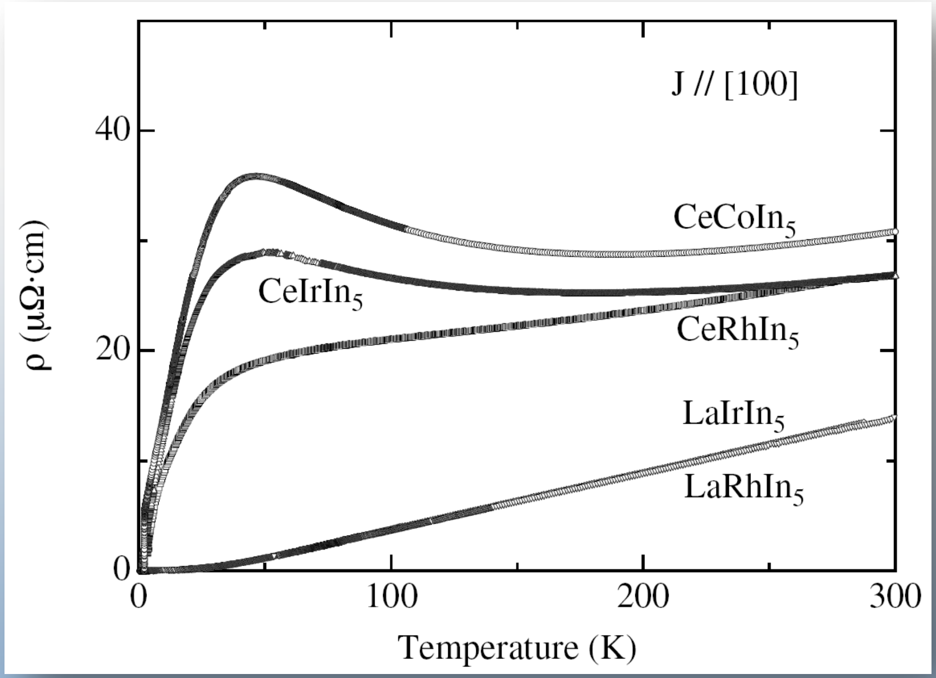


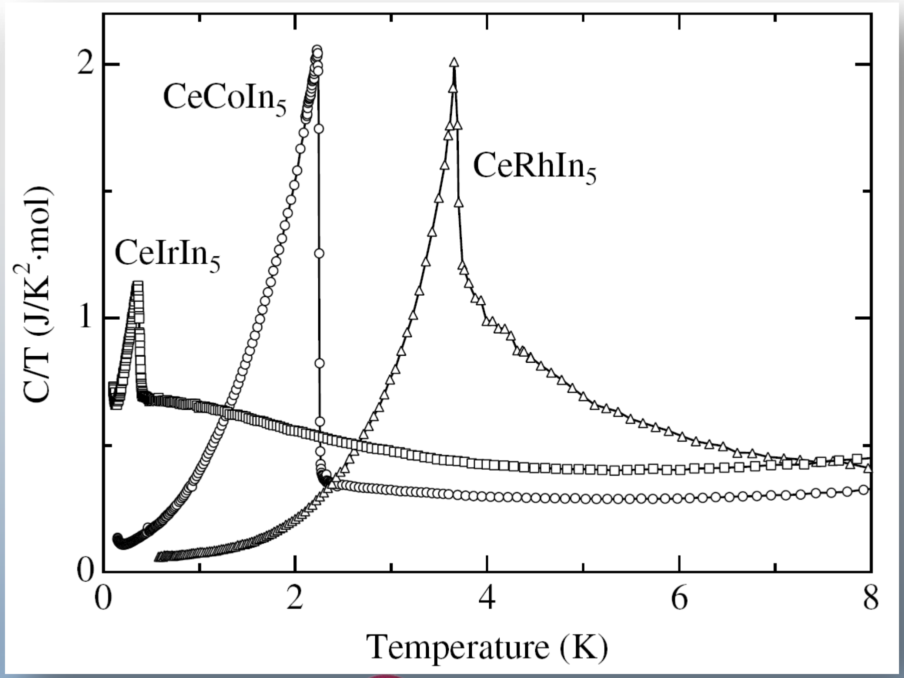


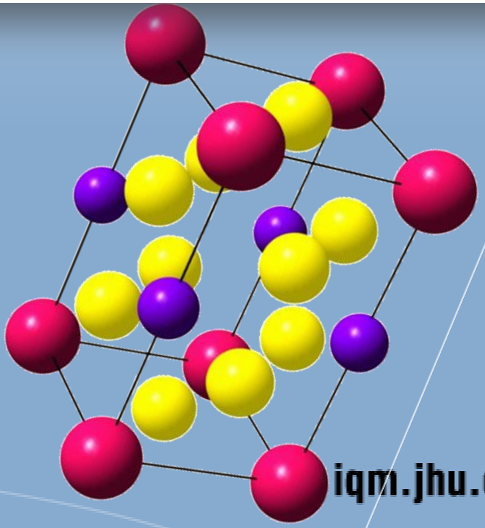
Kenzelmann et al. PRL (2003)

A family of correlated metals

Fisk, Thompson, Petrovic,…





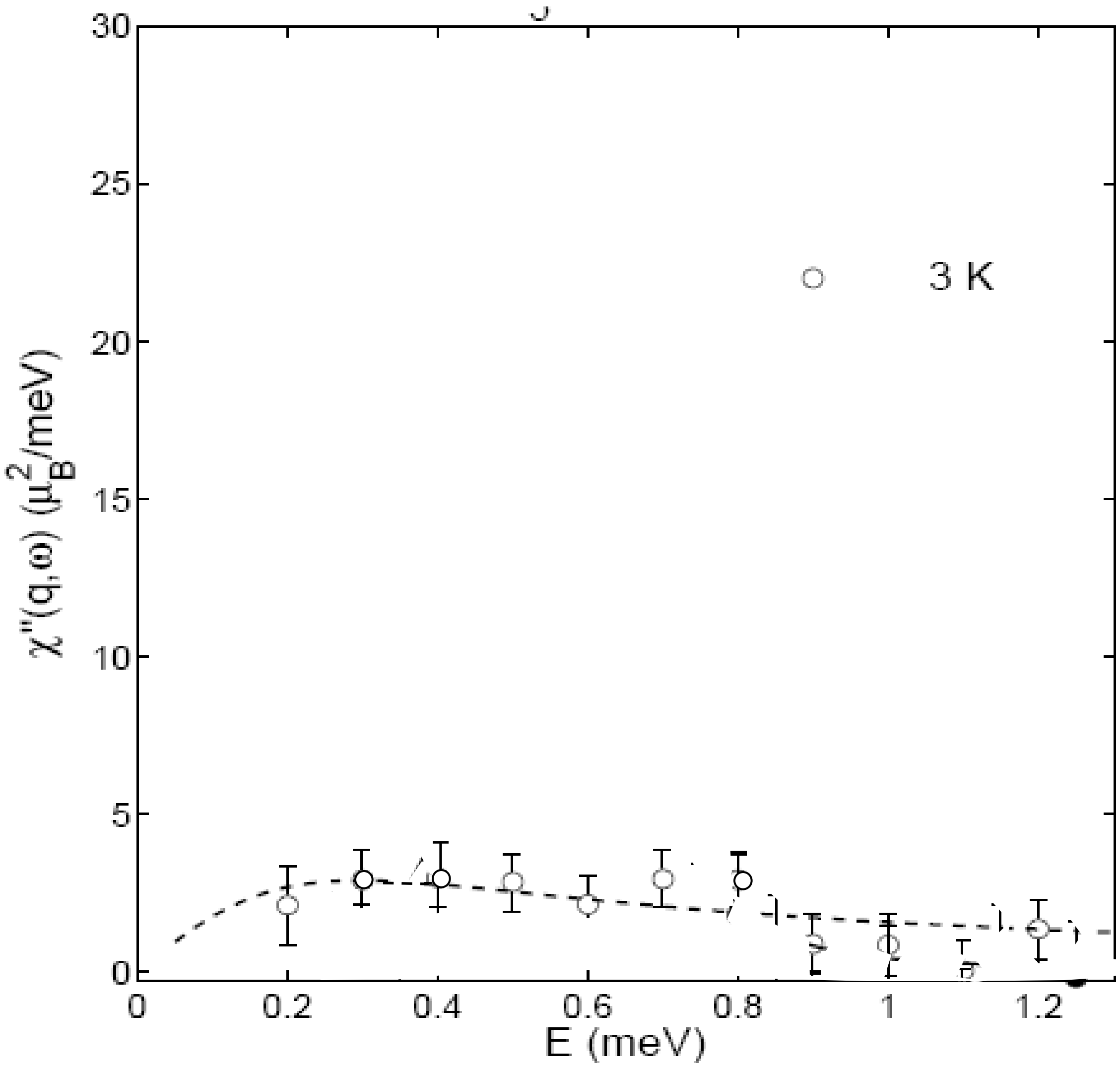


$CeCoIn_5$: HF superconductor T_c=2.3 K

$CeIrIn_5$: HF superconductor T_c=0.38 K

$CeRhIn_5$: HF Antiferromagnet T_N=3.8 K

Excitation Spectrum at Q_c



Spectral function:

Normal State: relaxation

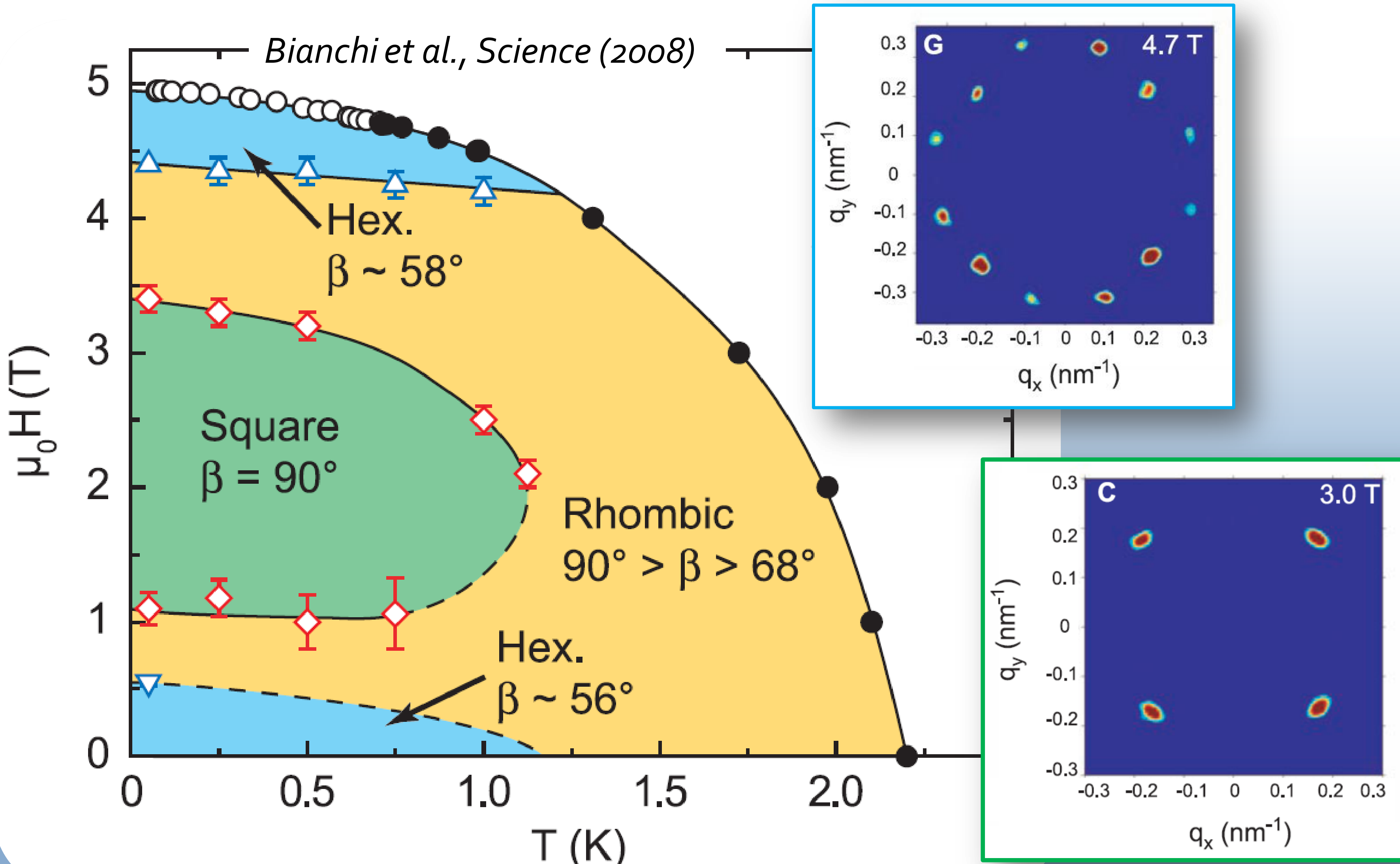
$$\hbar\Gamma = 0.30 \pm 0.15\,\mathrm{meV}$$

Superconductor: Resonance

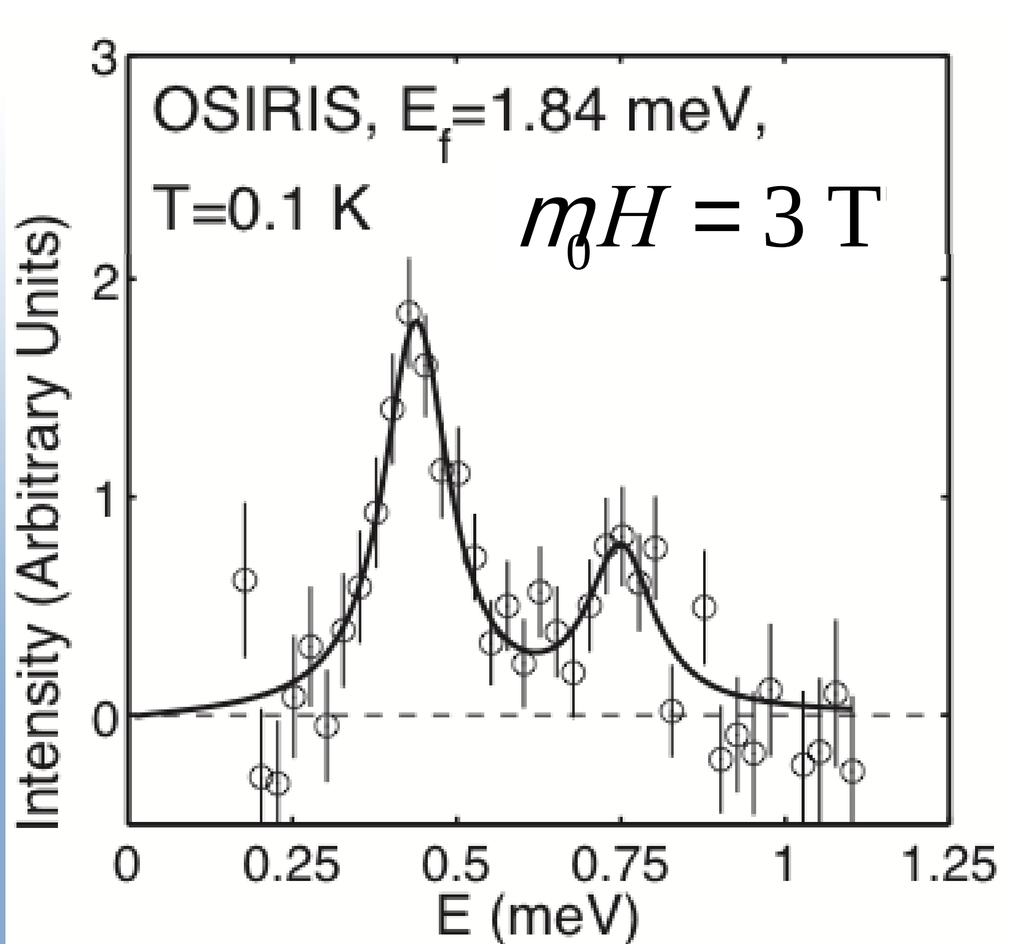
$$\hbar\omega_0 = 0.60 \pm 0.03\,\mathrm{meV}$$

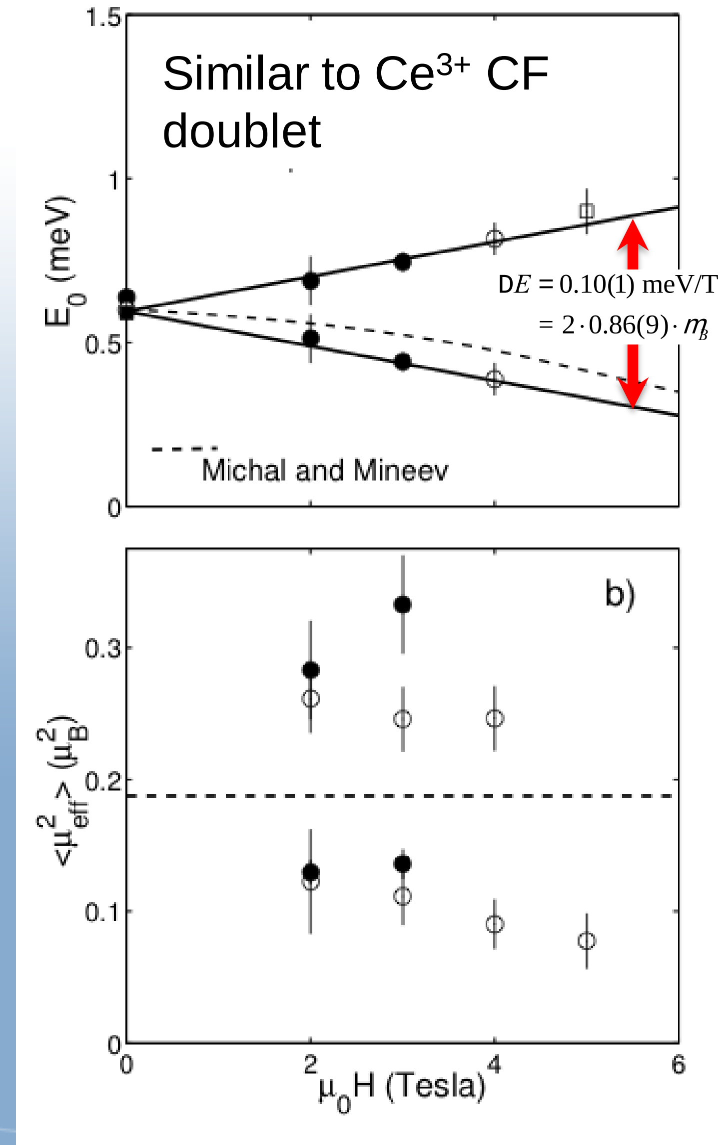
$$\hbar\Gamma < 0.05\,\mathrm{meV}$$

Structure of vortex matter: $CeCoIn_5$



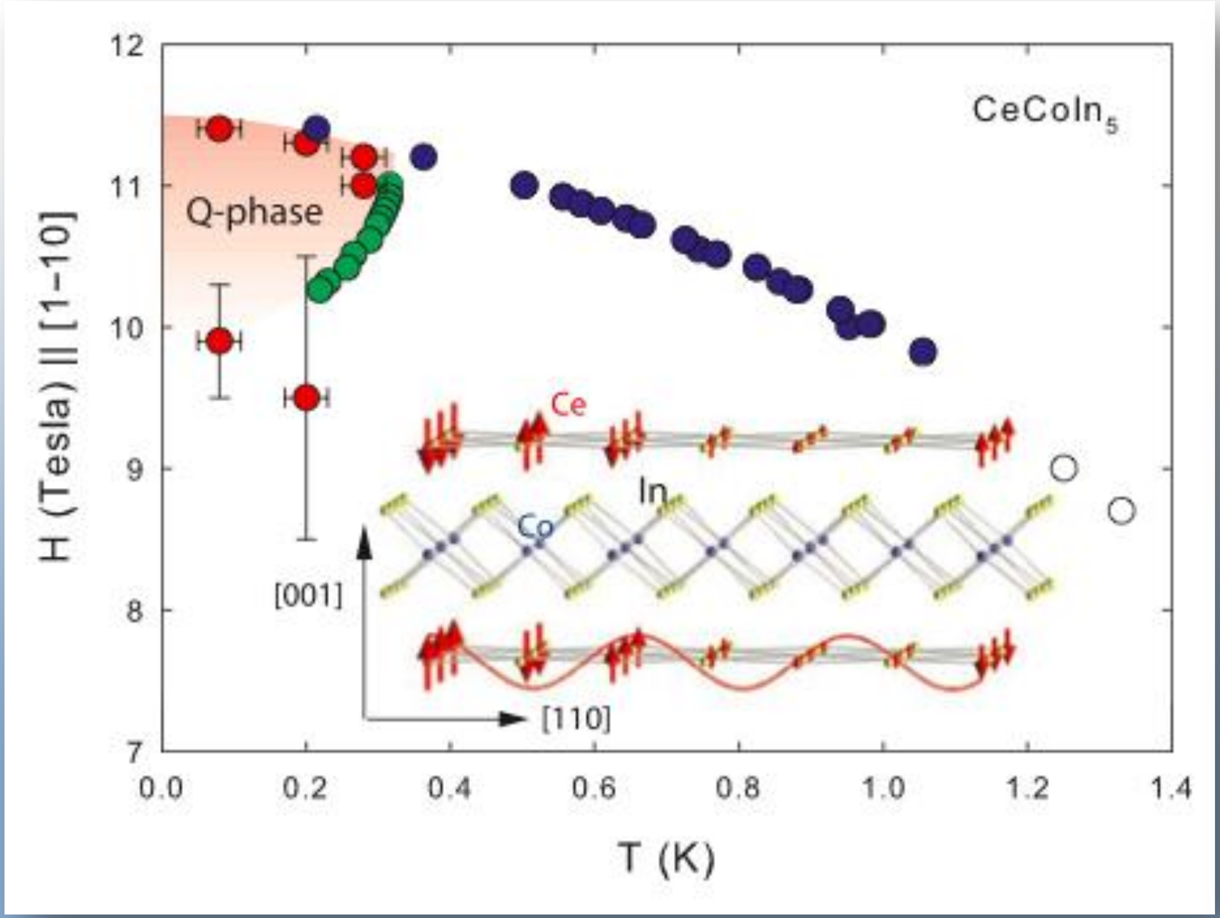
Higher Resolution data H||(110)

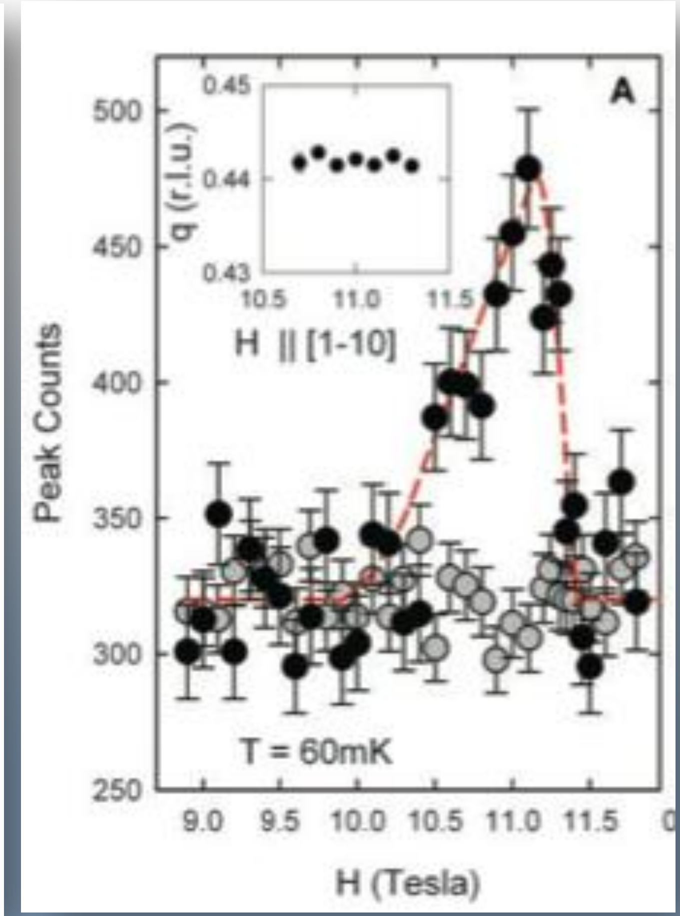




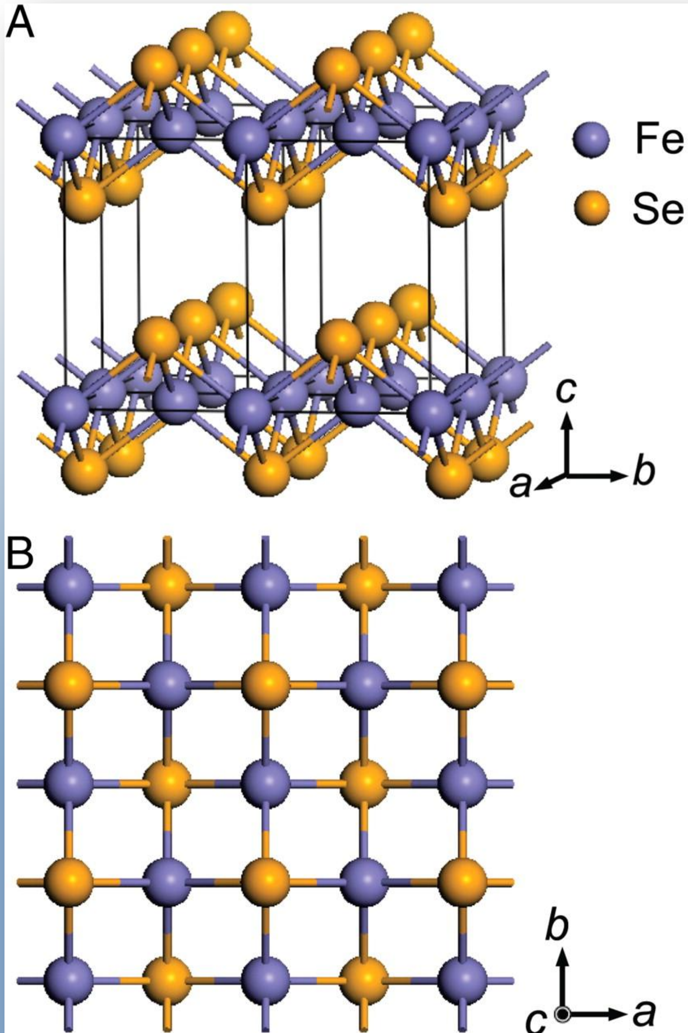
Spin Density Wave & Superconductivity

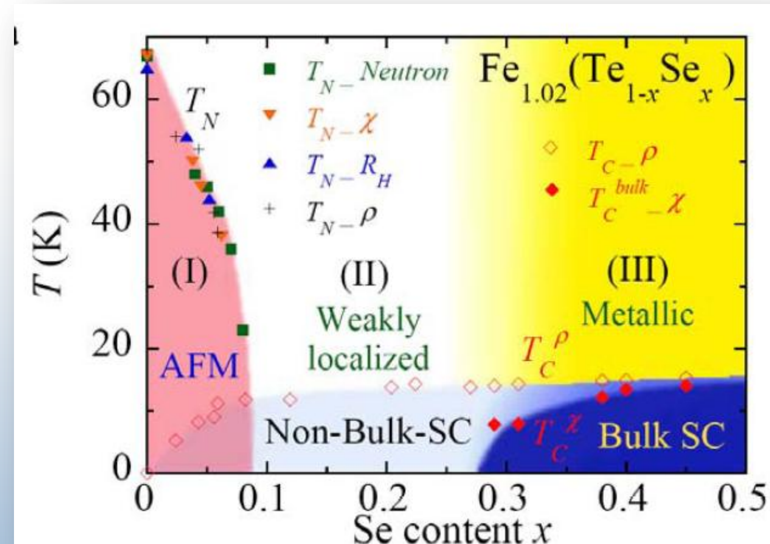
M. Kenzelmann et al. Science (2008)





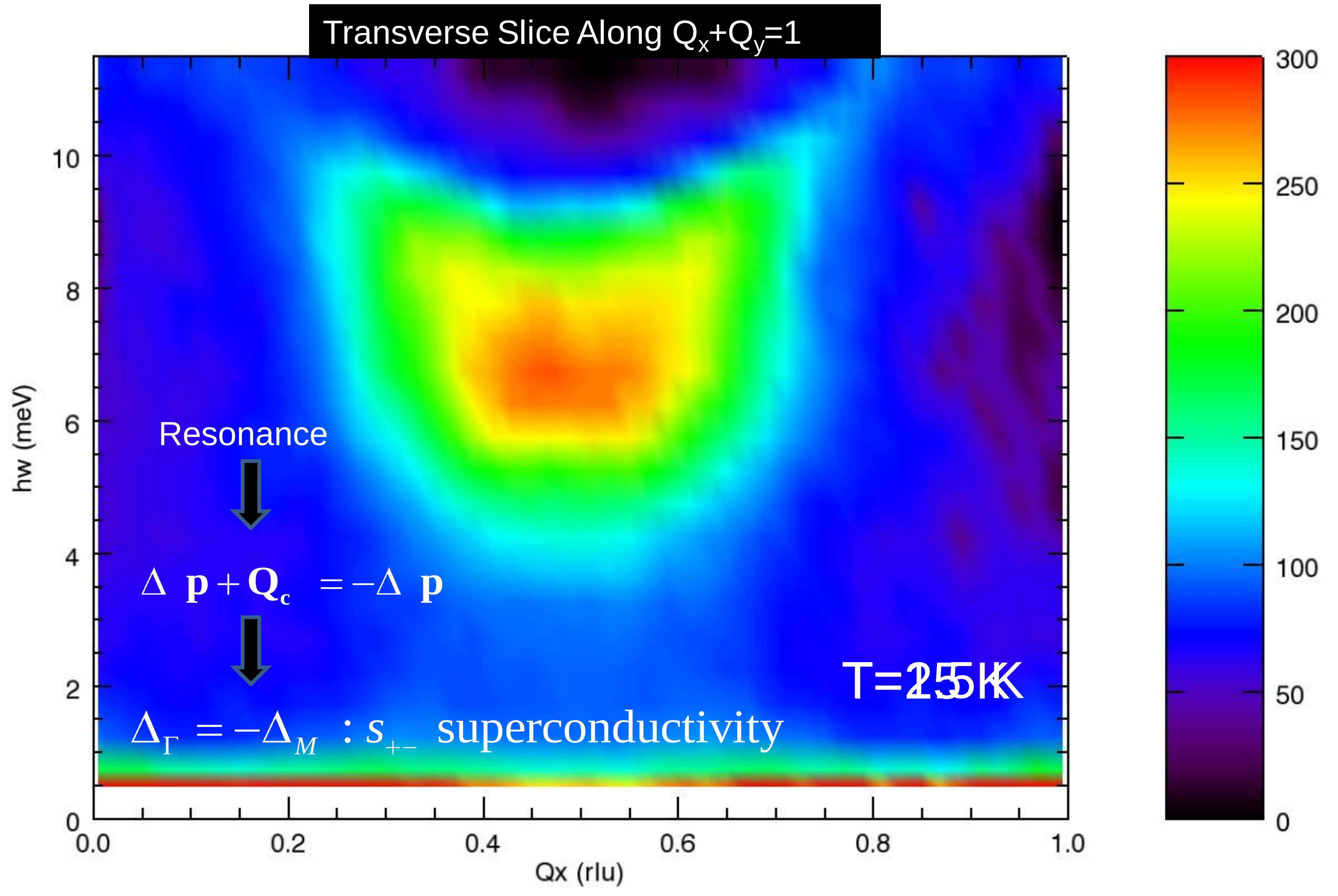
Superconductivity in $FeSe_{0.4}Te_{0.6}$



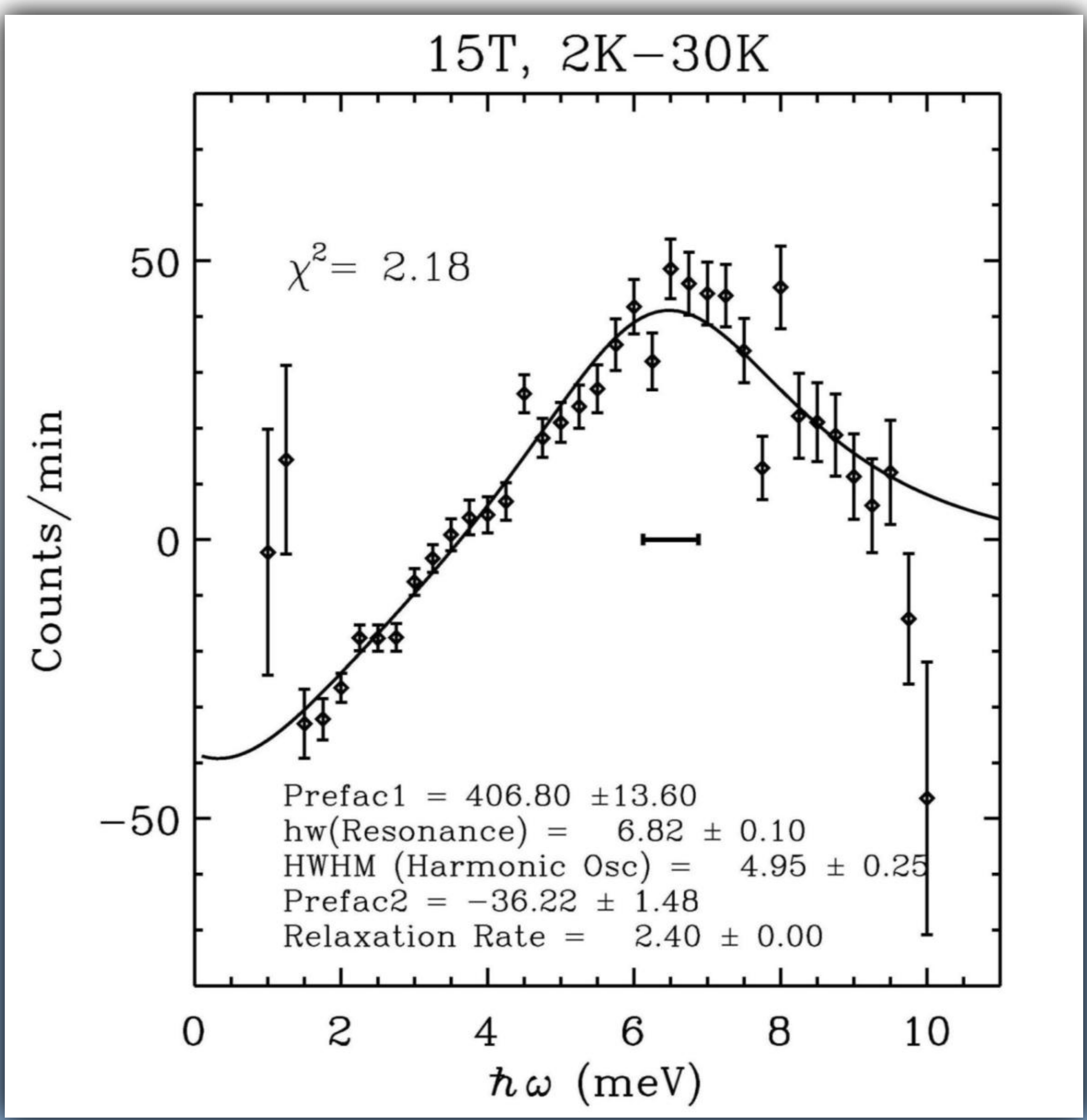


Hsu F et al. PNAS 2008;105:14262-14264

From Critical Fluctuations to Resonance



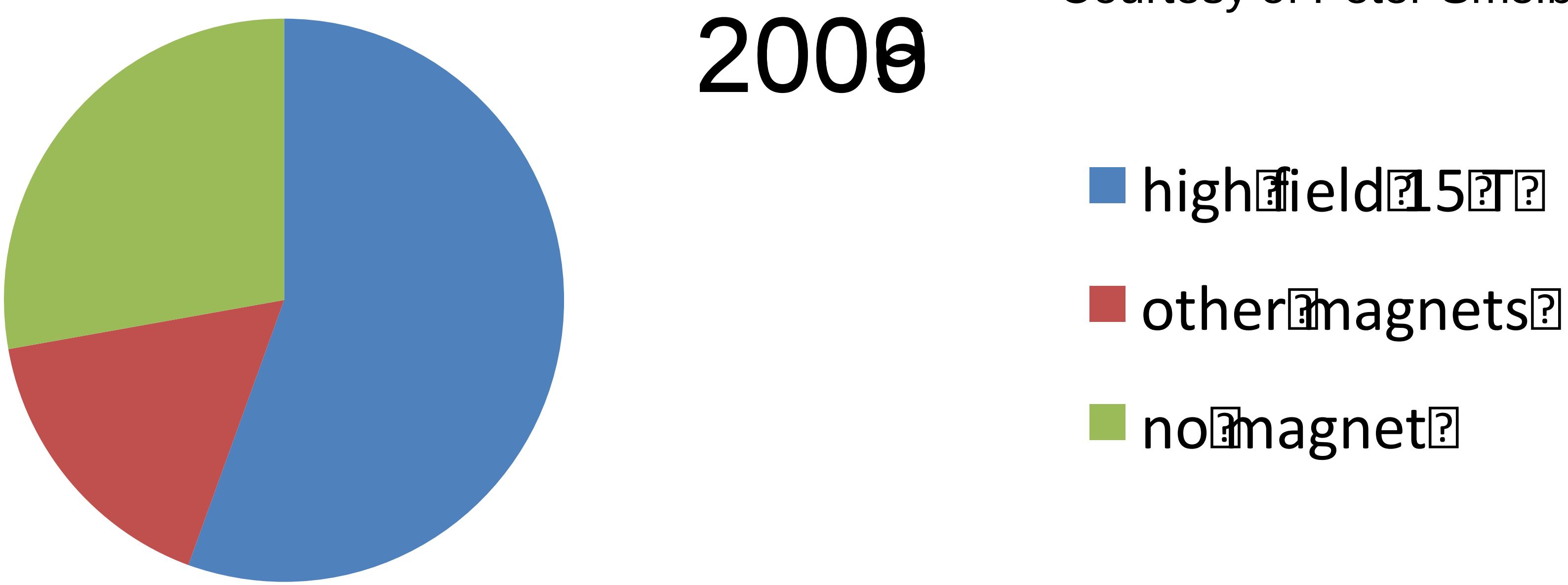
Field effect on spin resonance



- There is considerable fine structure in spectra collected at **Q**=(0.5,0.5,0)
- These features are not temperature dependent through T_c
- Possible explanations include phonons and magnons from magnetic second phase
- The predominant effect of applied field on the superconducting resonance is to broaden the line
- A possible origin is field enhanced effects of static disorder

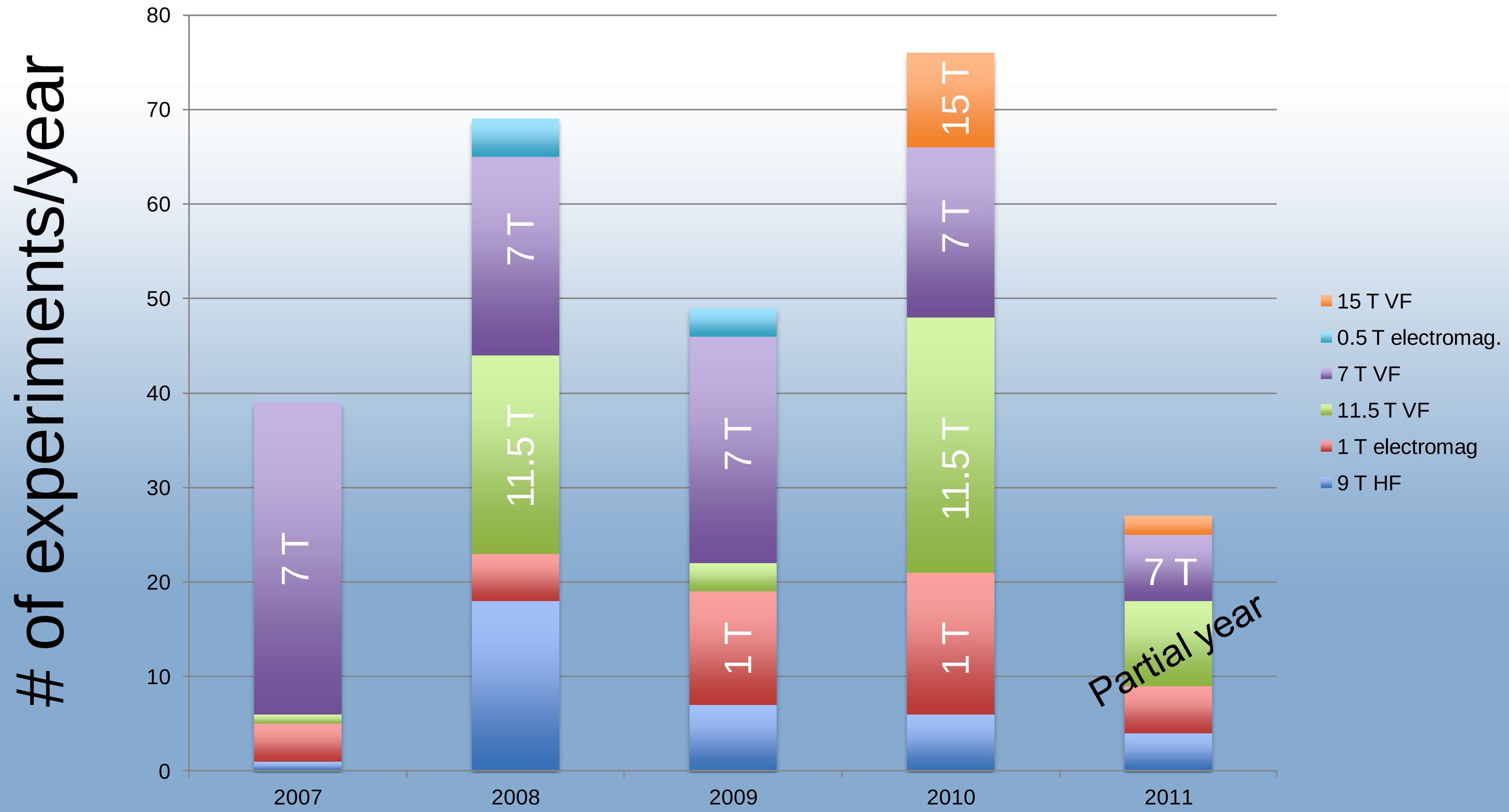
HZB: Strong user interest in neutron scattering at high fields

Courtesy of Peter Smeibidl

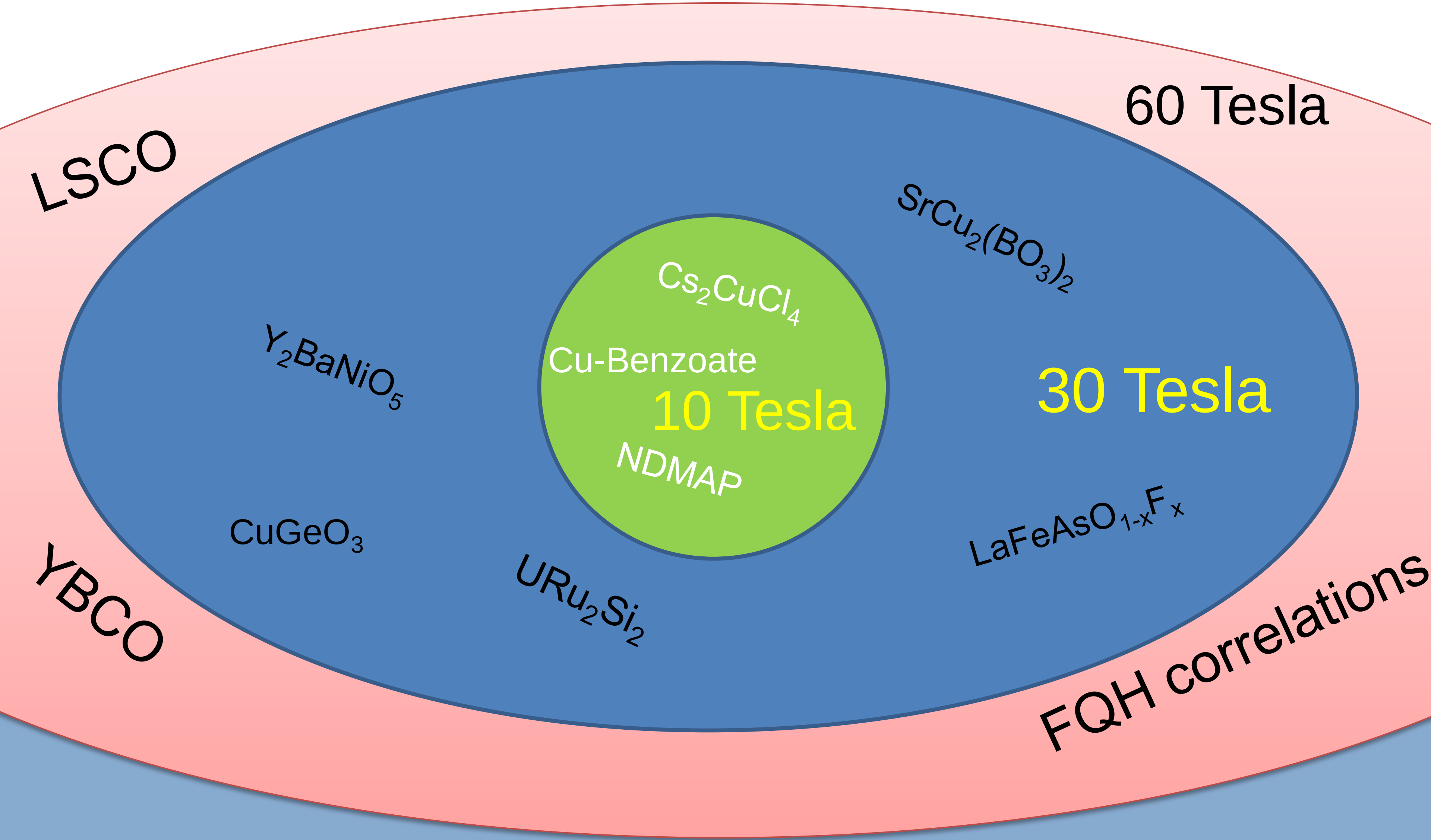


FLEX Triple axis instrument at Helmholtz Zentrum Berlin

NIST: Strong user interest in neutron scattering at high fields



The high field frontier

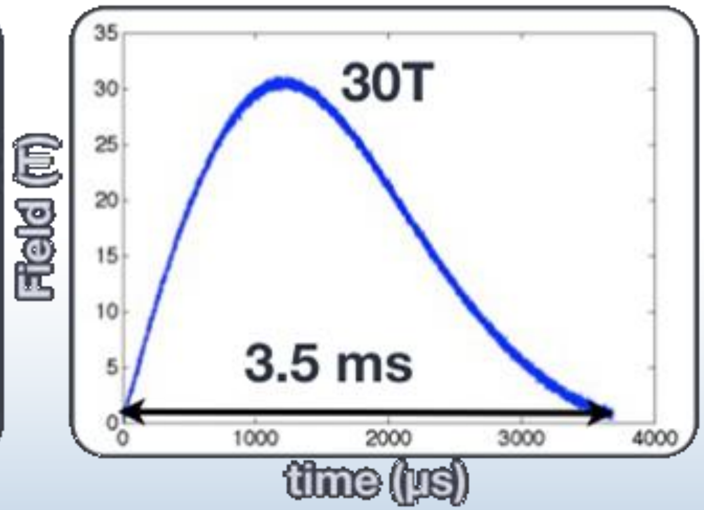


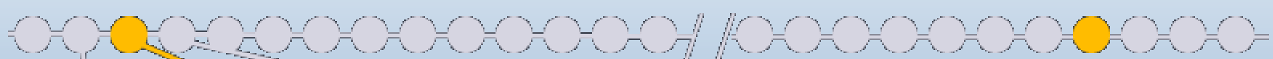
- Address progressively higher energy phenomena for applications
- A new dimensional in materials space allowing access to novel phases

DC and pulsed fields at the SNS

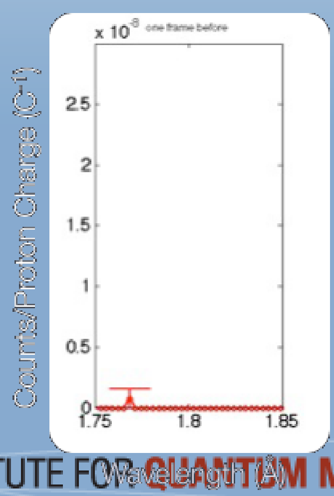
30 Tesla pulsed magnet
From Nojiri

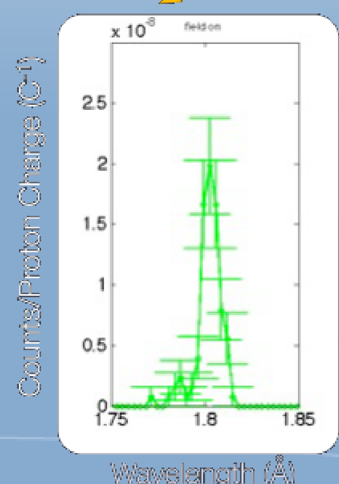
Pulse Field Neutron Scattering

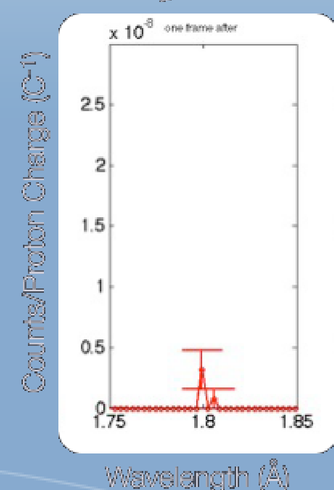




One field pulse per 18,000 neutron pulses
300 field pulses needed for one Bragg peak



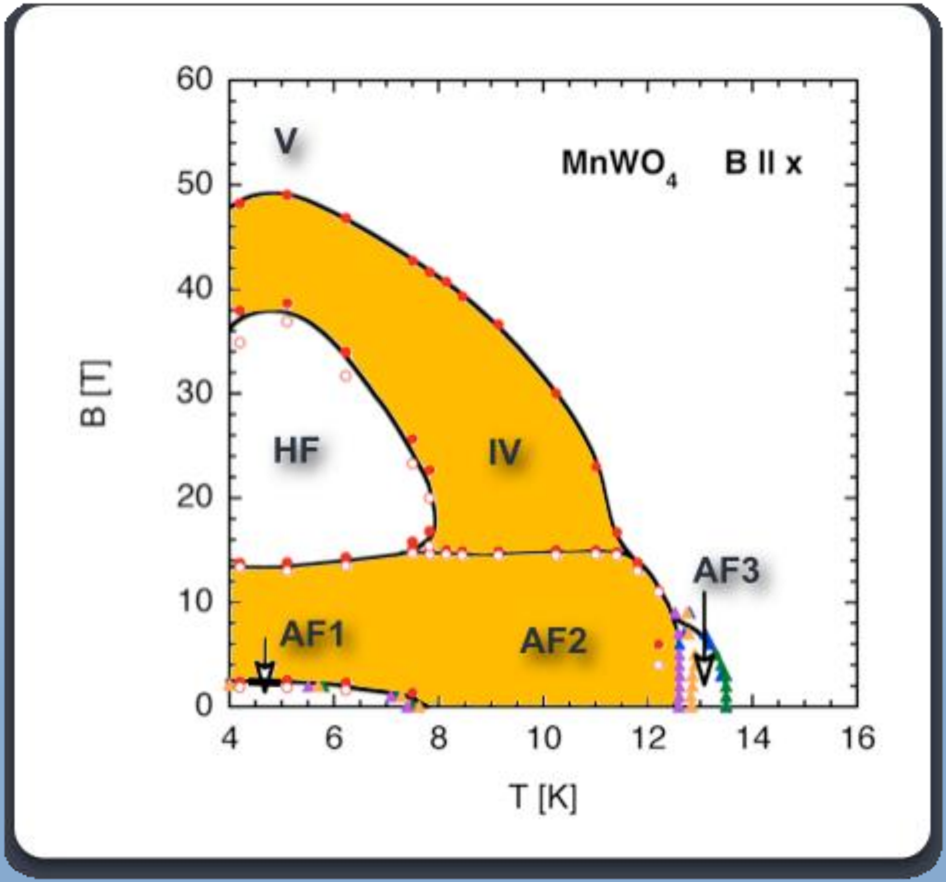




Kate Ross &
Bruce Gaulin,
McMaster U.

First neutron diffraction at 30 Tesla

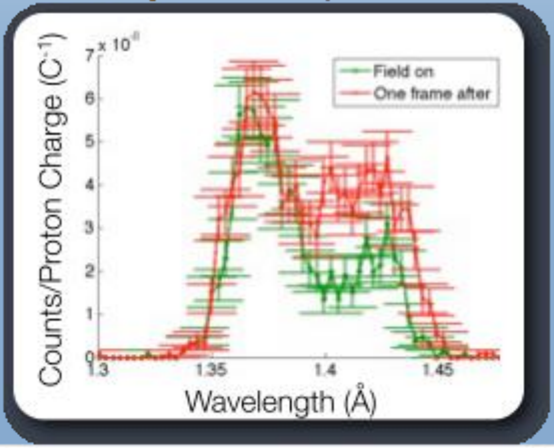
K. Ross, B. Gaulin, G. Granroth, L. Santodonato, P. Peterson,
A. Parizzi, H. Nojiri, S. Yoshii, M. Yasui, K. Okada, M. Matsuda,
B. J. Carlo, and J. Ruff.



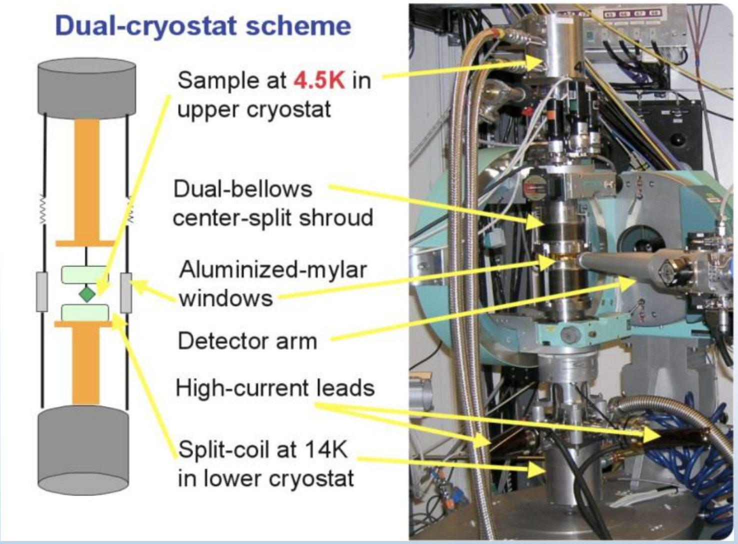
In IV phase, IC structure with same ordering wavevector as AF2

In HF phase, commensurate order with no sign of IC phase (phase fraction less than 1%)

Suppression of asymmetric intensity in HF phase?

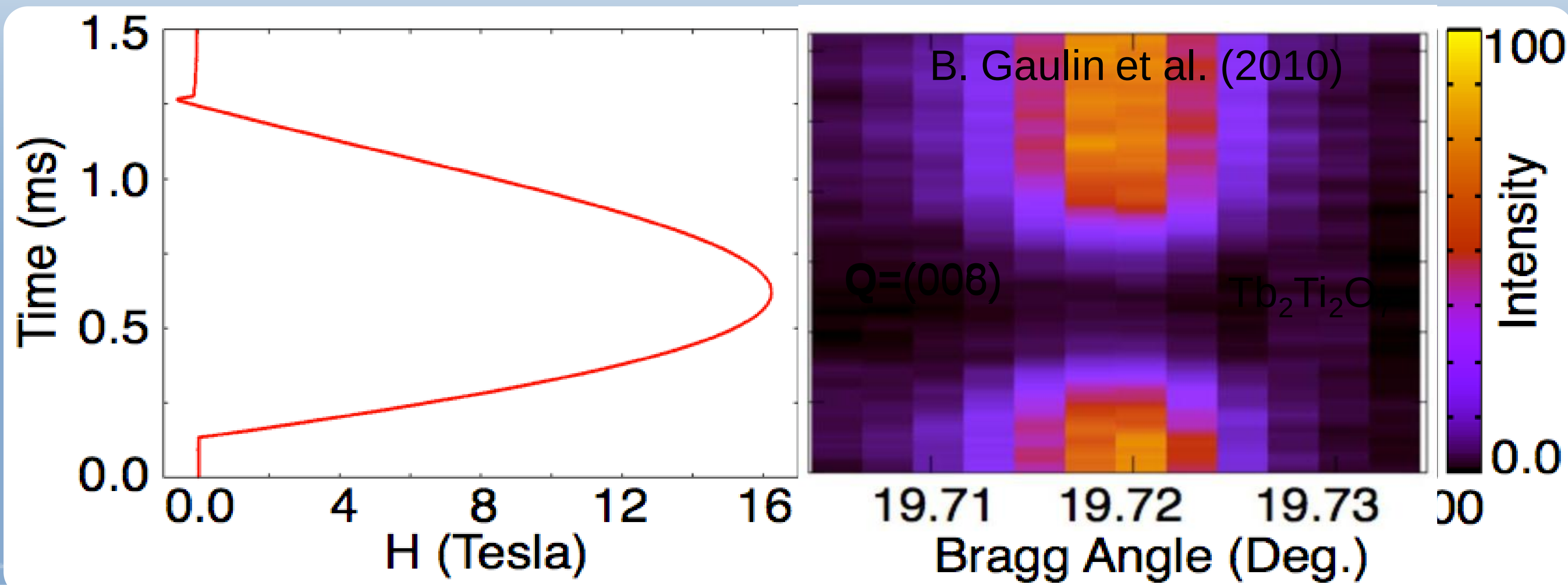


A different story for synchrotron x-rays



- **Magnetostriction resolvable to the μs**
- **Pulsed fields to 30 Tesla at 4.5 K -300 K**

Islam *et al.* Rev. Sci. Instrum. (2009)



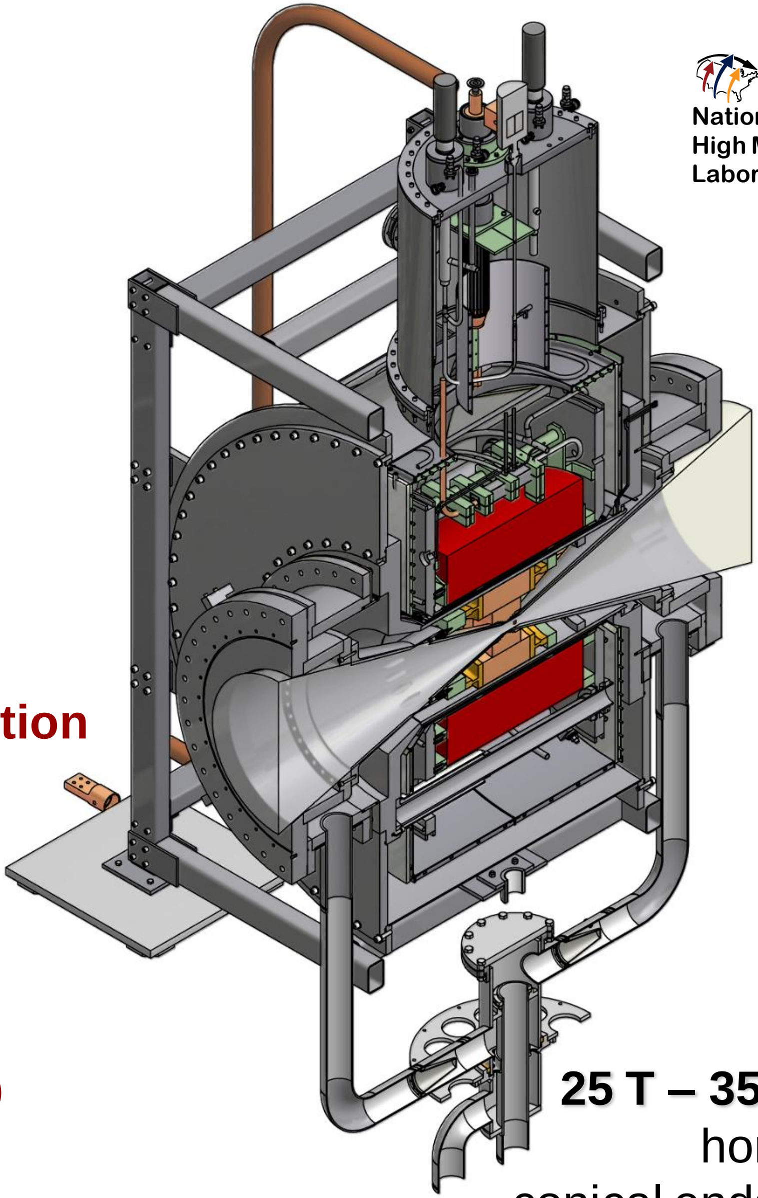
High DC field Neutron Scattering

- For conducting materials need DC field
- 10^4 duty cycle gain compared to pulsed magnets
- 10^2-10^3 gain in sample volume
- Enables inelastic scattering
- larger sample volume

Magnet	ZEEMANS	HZB	NHMFL
Outsert magnet technology	Nb_3Sn + NbTi CICC	Nb_3Sn CICC	Nb_3Sn CICC
Outsert field contribution (T)	13	13	13
Insert magnet technology	YBCO + Nb_3Sn	Florida-Bitter	Florida-Bitter
Room temperature bore (mm)	50	50	40
Conical Angle (degrees)	30-40	30	4
On-axis field (T)	25 - 30	25	36
Power (MW)	0	4	13

Hybrid Magnet + Infrastr.

National
High Magnetic Field
Laboratory



Challenges

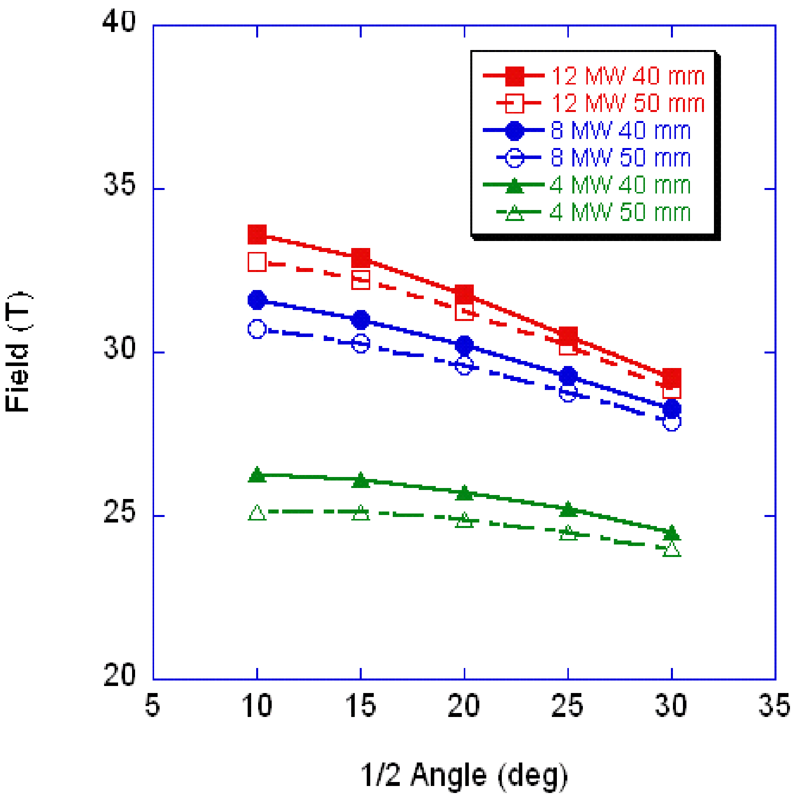
- Design and Construction
- Operation

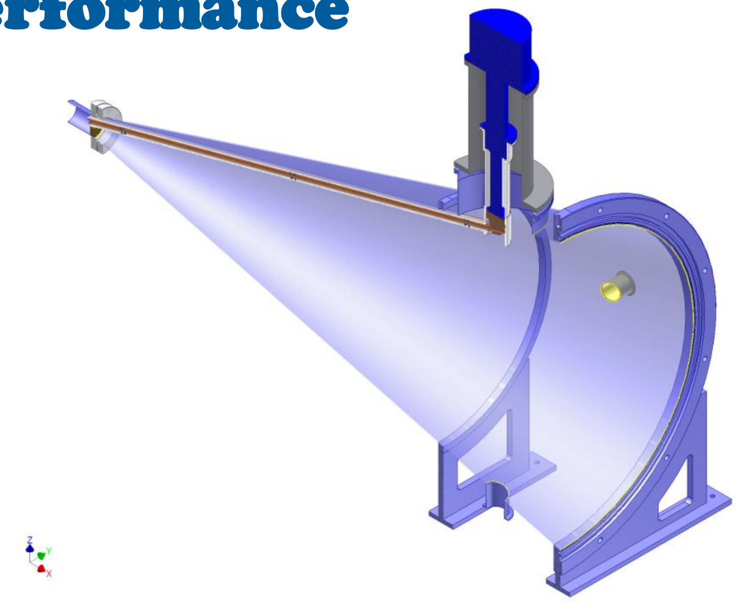
Series-Connected System

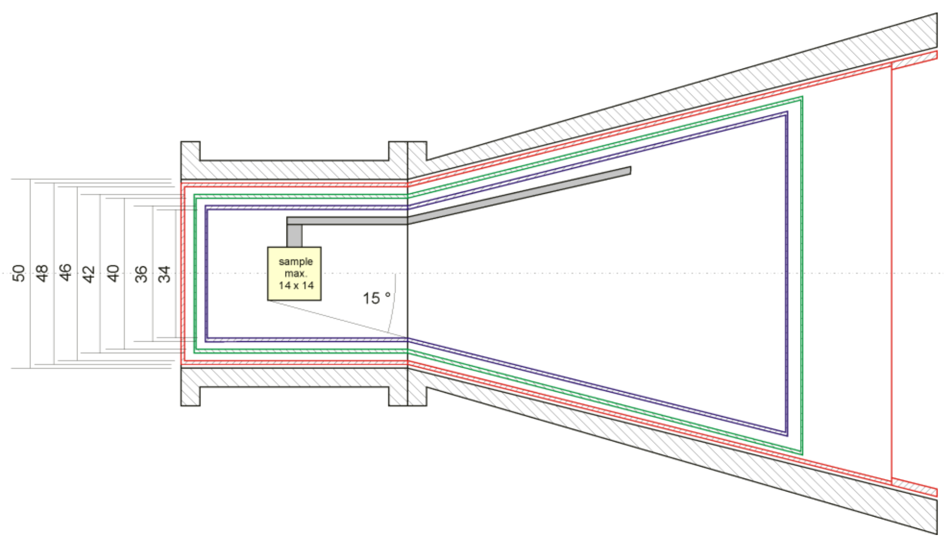
- SC coil (Cable-in-Conduit)
- resistive Bitter coil

25 T – 35 T Hybrid Magnet
horizontal field
conical ends (30°opening angle)

Neutron Scattering Experiment System Performance







Geometry

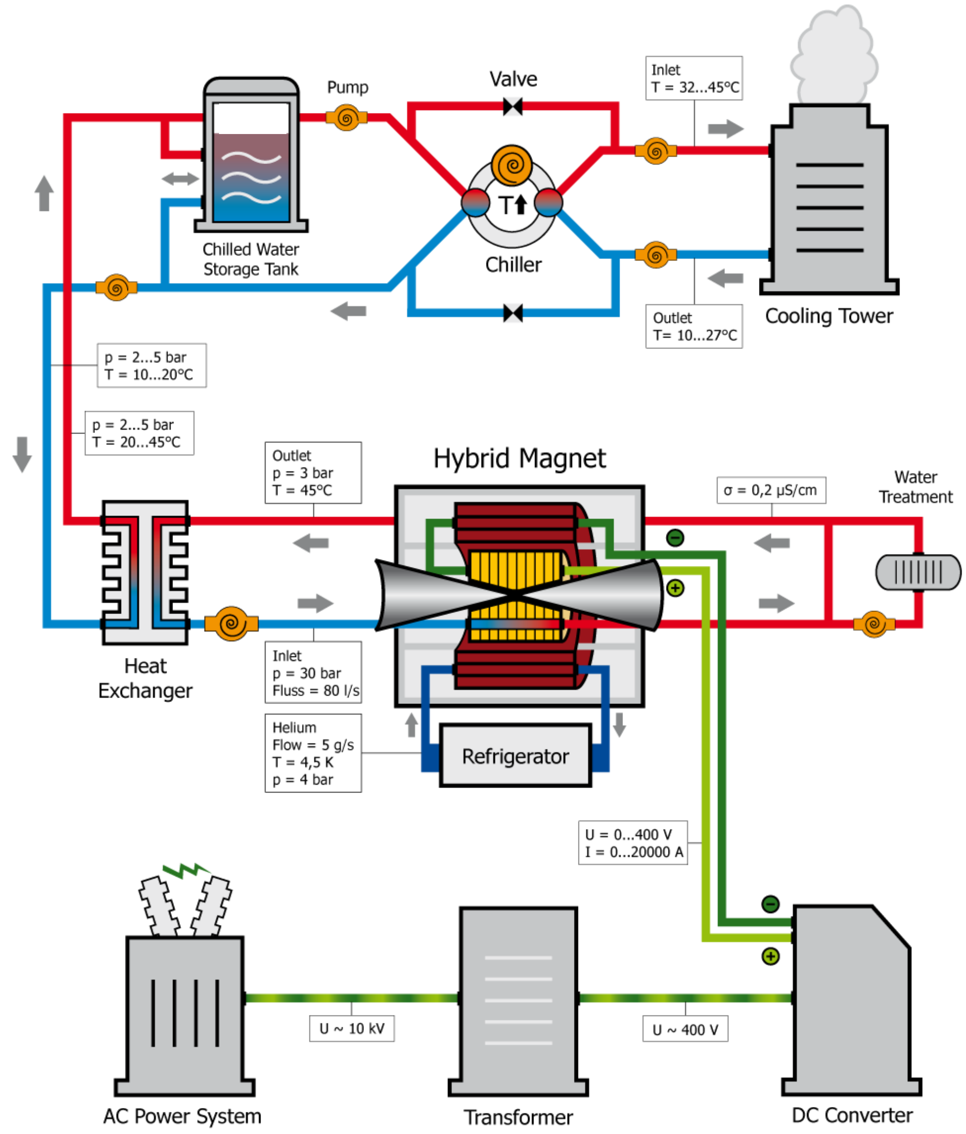
- 15 mm – 20 mm sample size
- 30°conical angle
- 50 mm bore

Hybrid Magnet System + Infrastructure

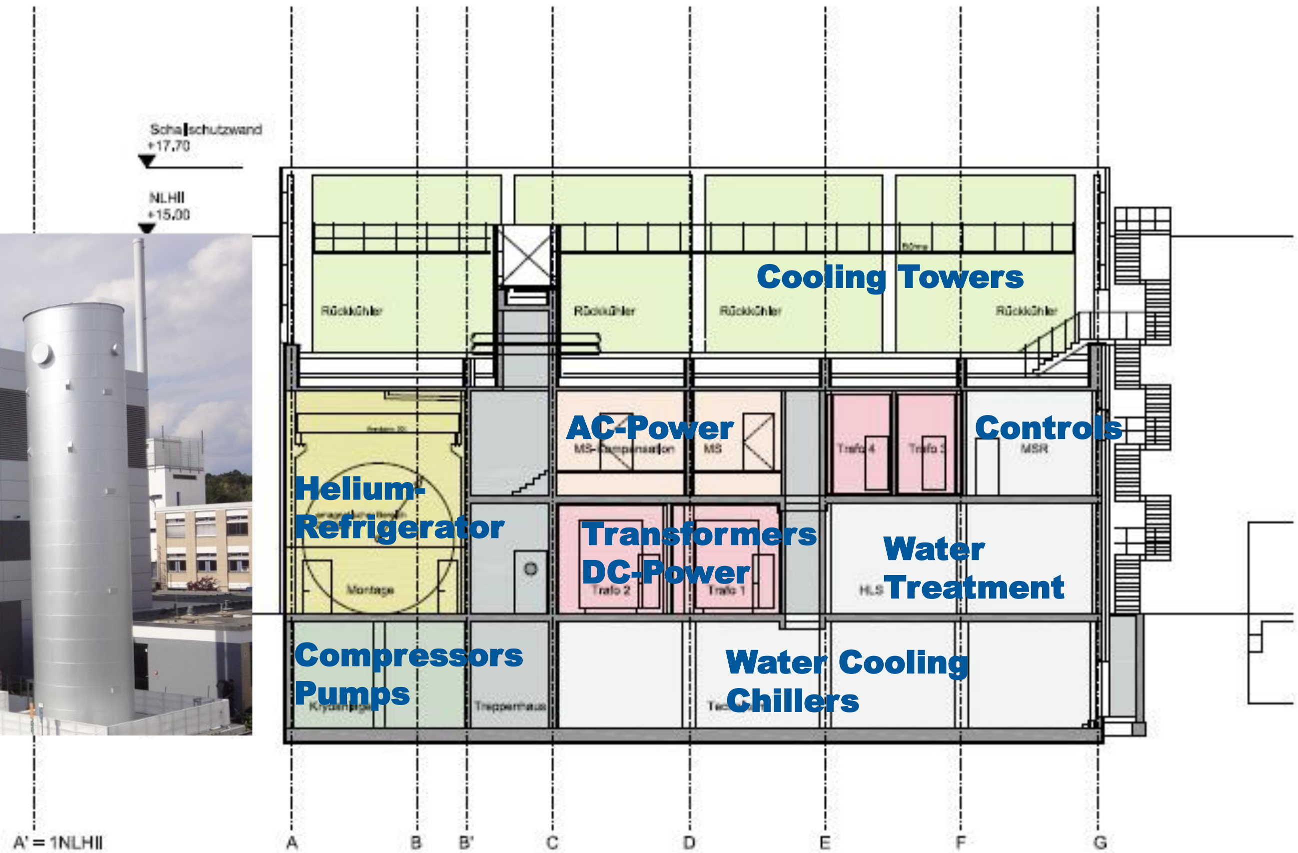
Power:
20 kAmp,
200 / 400 V
4 / 8 MW
Temporal stability: 10 ppm

Water:
4 / 8 MW
40 / 80 l/sec, 30 bar
0.2 µS

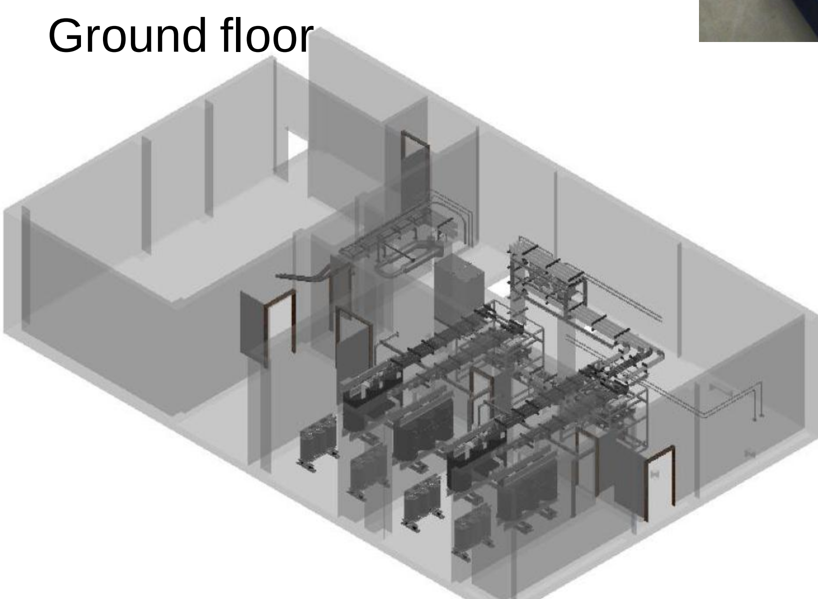
Cryoplant:
Supercrit. He:
10 g/sec, 4 bar, 4.3 K
Liq. N_2:
15 - 20 g/sec, 77 K

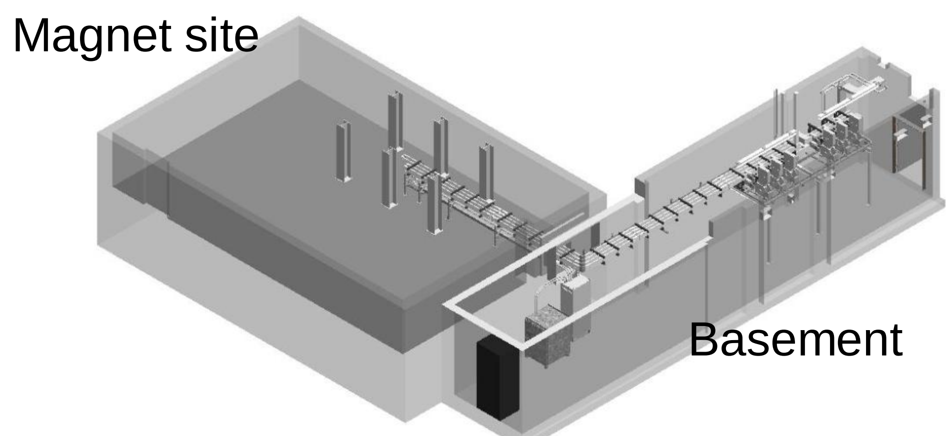


Infrastructure Building



Power Supply
(8 MW)





Water Cooling - primary loop

Built and operated by external company

- **Status proj. phase 1: 4.4 MW cooling power**
- **2 MW chiller**
- **3 MW + 2.4 MW cooling towers**
- **250 m^3 cold water buffer**
- **Hardware complete**

Commissioning started Mar. 2012 in combination with power supply

Helium Refrigerator

Contract Feb. 2010 (Linde)
(16 months delivery)

Detailed design complete
(Sept. 2010)

Installation: 3 Q / 4 Q 2011

Commissioning complete: 1Q 2012

Cryostat Fabrication

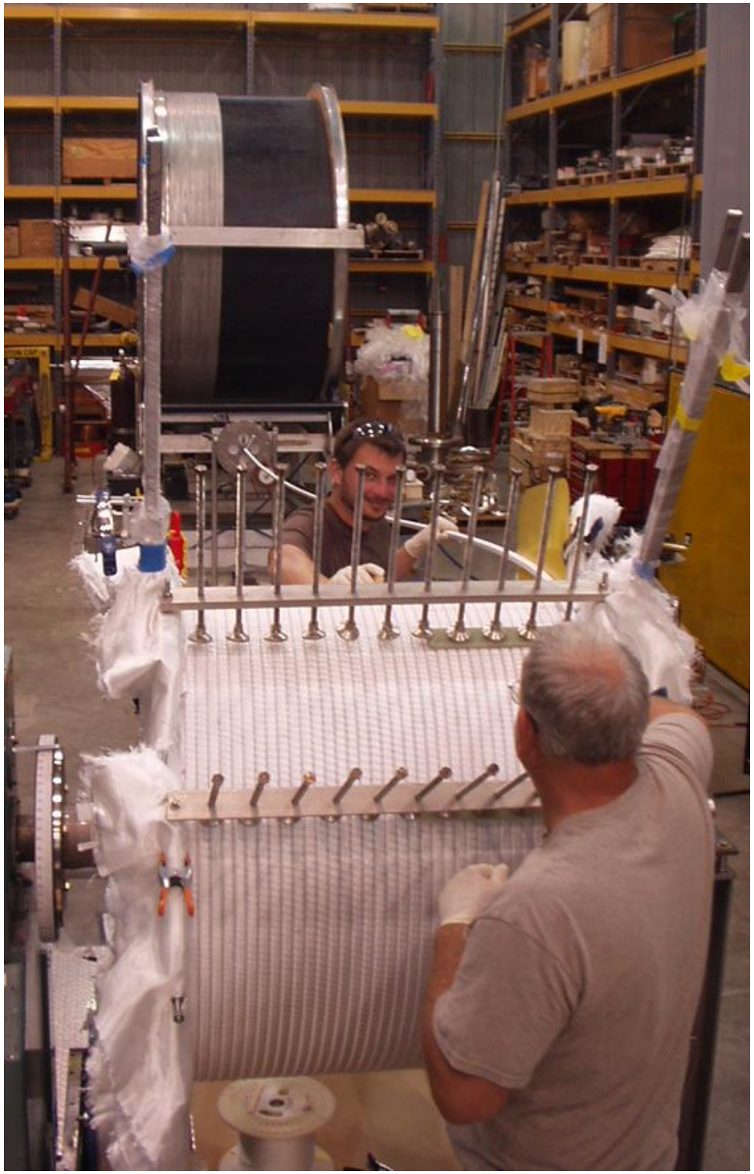
Contract Dec. 2010 (Criotec)
(16 months delivery)

Detailed design complete
(June 2011)

Fabrication well underway

Completion: 3Q 2012

Coil Winding at NHMFL

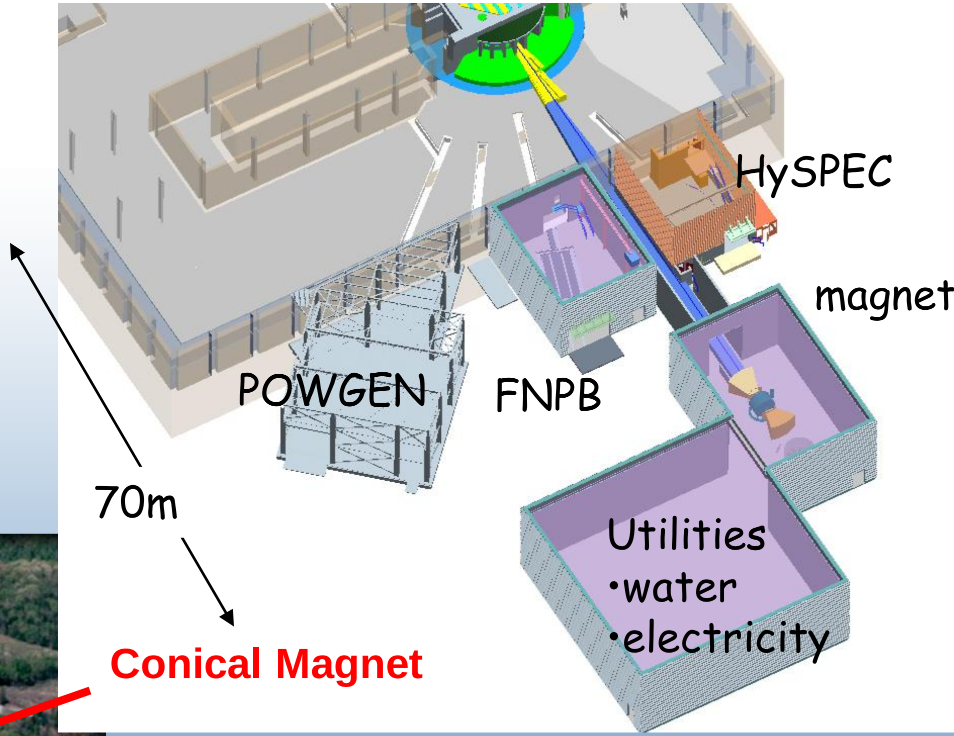


- HZB 25 T Magnet Schedule
  - 17 of 18 layers of HZB coil Complete May 2012!
  - Joints between conductors complete Aug. 2012
  - Reaction Complete Oct. 2012
  - Impregnation complete Jan 2013
  - Cold Mass Assembled March 2013
  - Arrive Europe May 2013
  - Cryostat Assembled Oct. 2013
  - Arrives HZB Dec 2013
  - 25 T March 2014

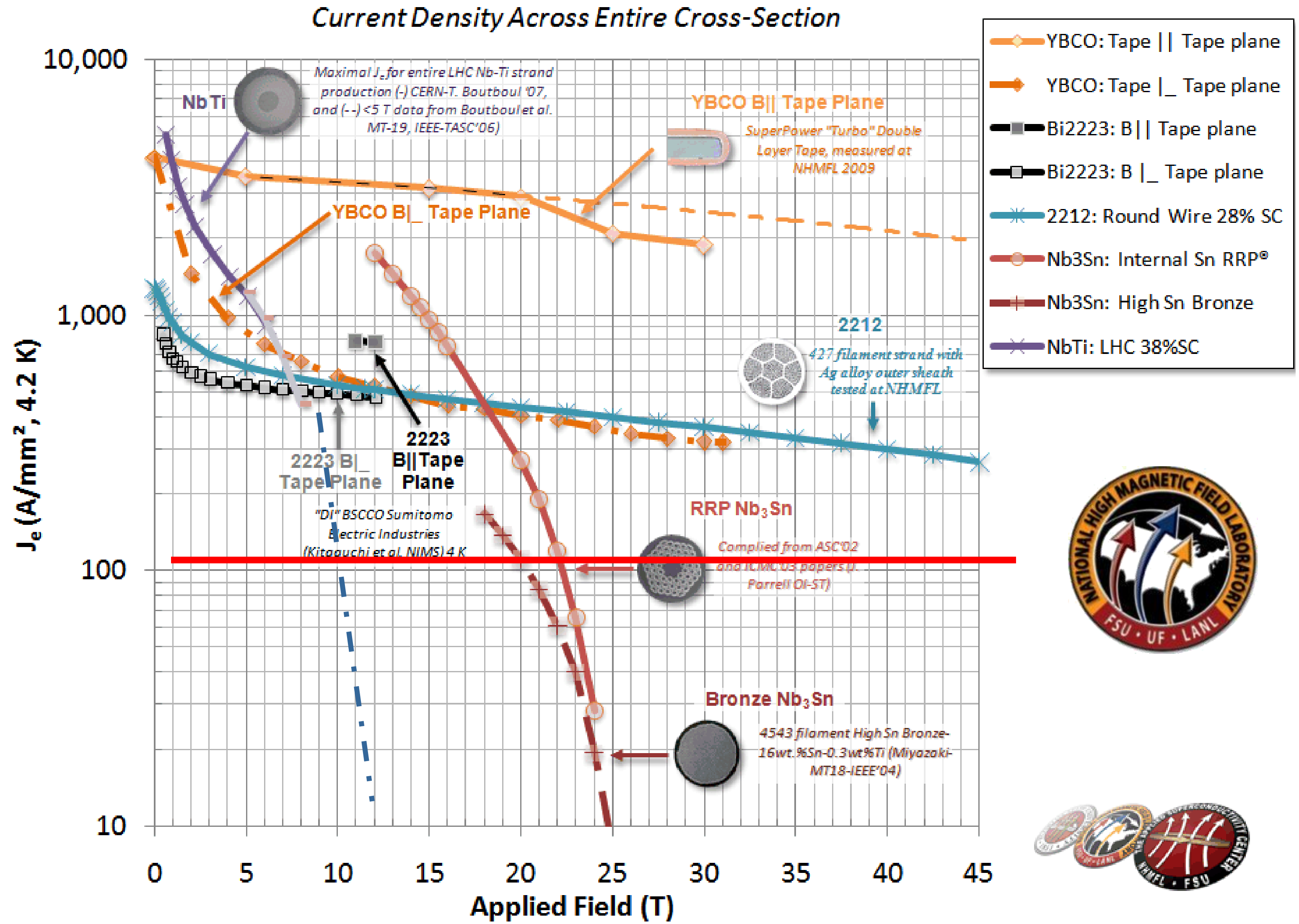
High field scattering at SNS

The ZEEMANS project :

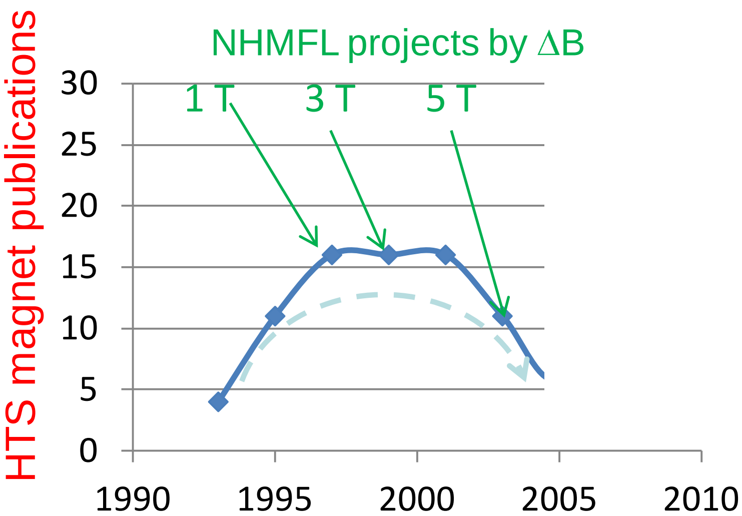
- 30 Tesla
- All forms of neutron scattering
- Brightest neutron source
- Fully superconducting

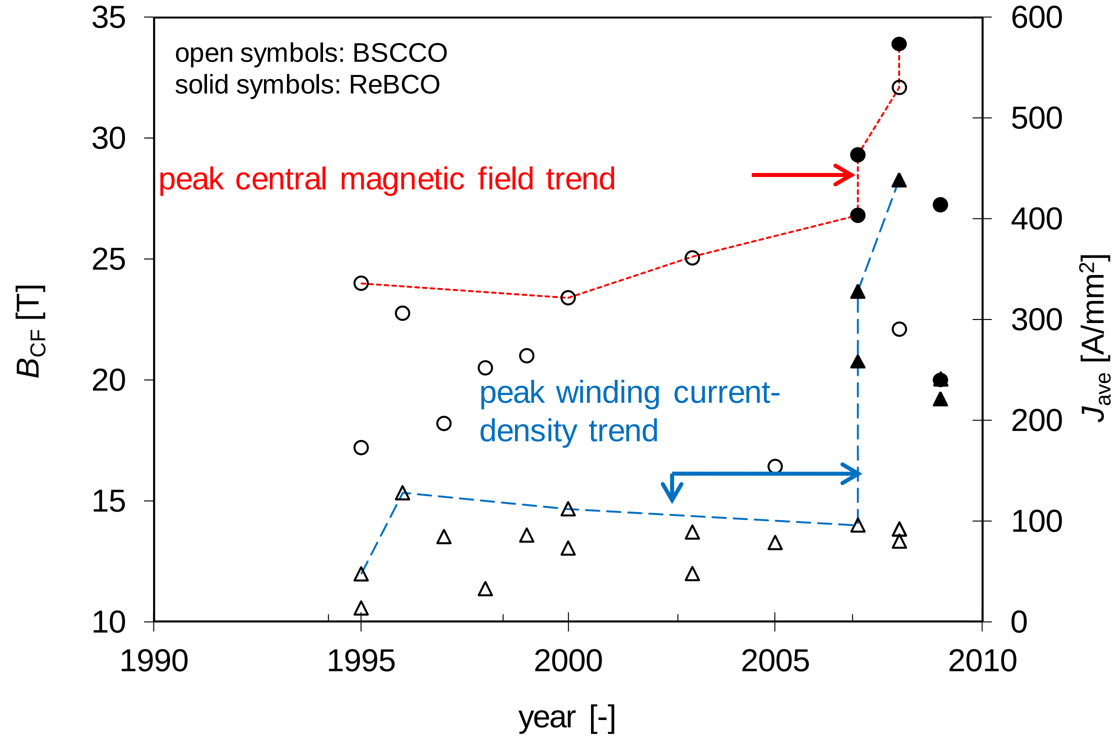


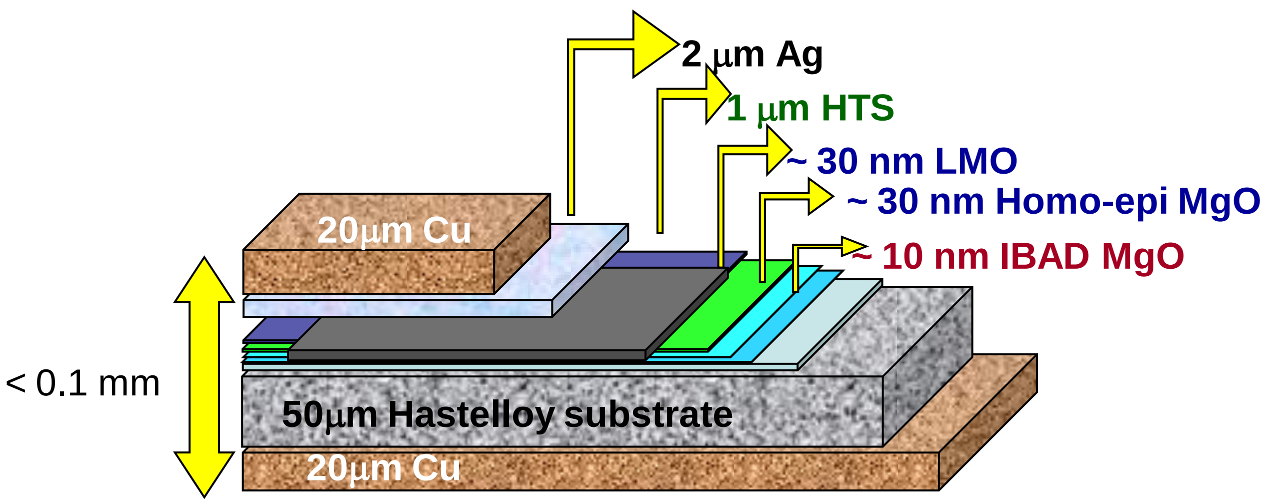
Developments in conductors for superconducting magnets



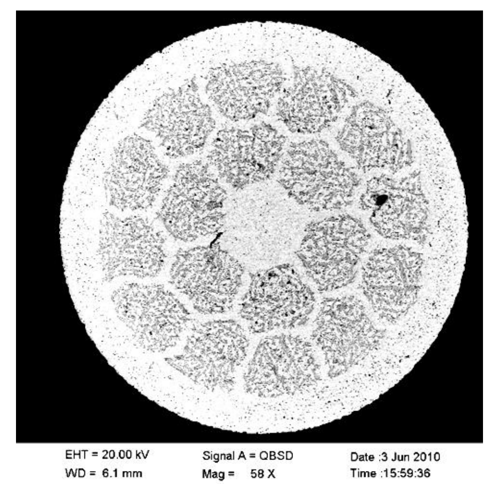
High T_c Coils







2G YBCO Tape - SuperPower



Bi2212 Round Wire - OST

MagLab/HZB/ESS Collaboration

The European Spallation Source (similar to the SNS in Oak Ridge, Tennessee) will be built outside Lund Sweden.

It is intended to be operational in 2020 and will replace European sources which are expected to reach the end of their life.

The MagLab is collaborating w/ ESS and the Helmholtz Zentrum Berlin by conducting a conceptual design of the next-generation neutron-scattering magnets.

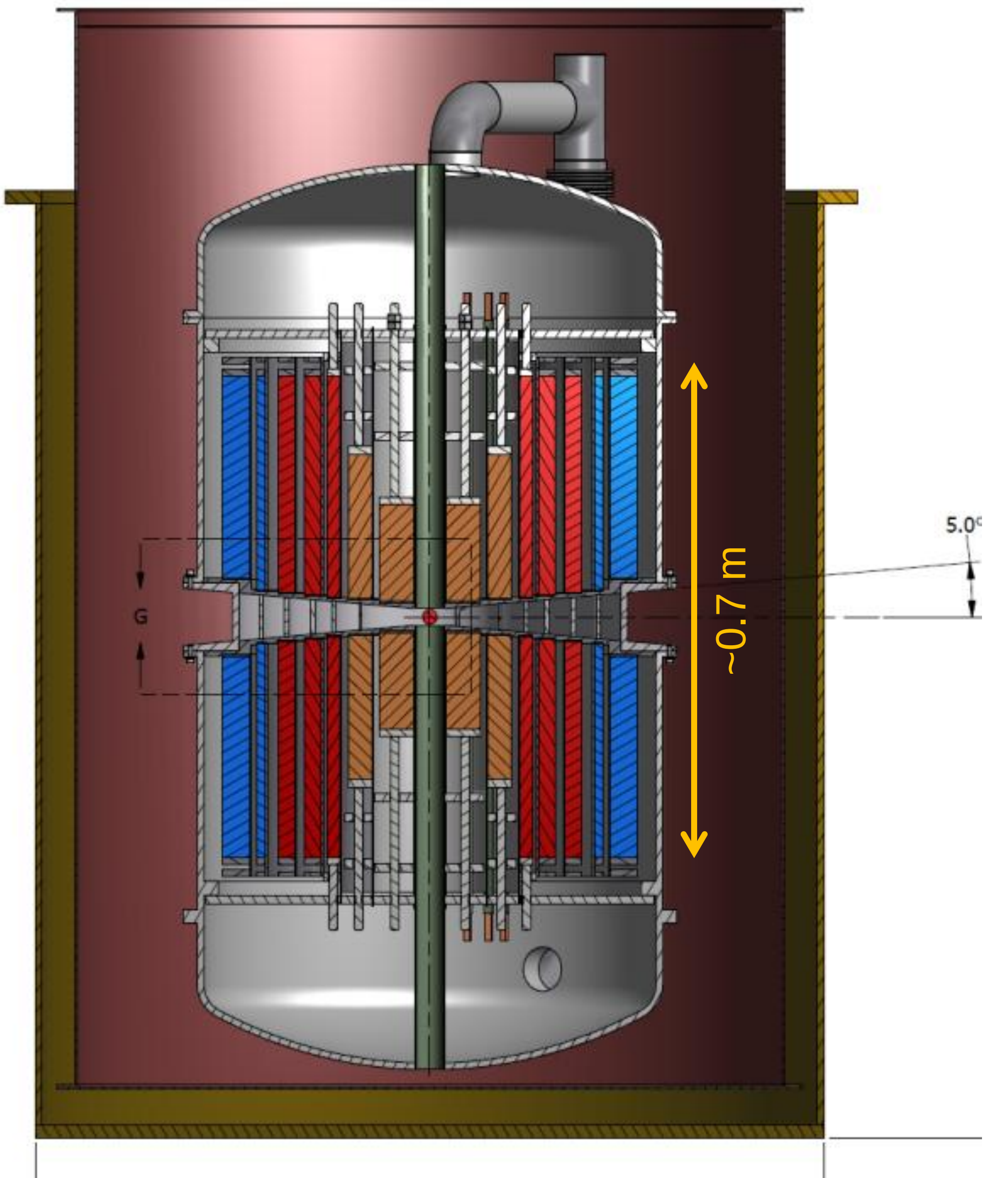
M.D. Bird, S. Bole, I. R. Dixon, A. V. Gavrilin,
W. D. Markiewicz, P. Smeibidl, J. Toth, H. W. Weijers, M. White

HELMHOLTZ CENTRE BERLIN for Materials and Energy

EUROPEAN SPALLATION SOURCE

MagLab/ESS Concepts

25 T Superconducting Split Magnet



Mid-Plane Spacers

Split magnets provide field perpendicular to the beam which lies in the scattering plane.

The scattered neutrons typically pass through aluminum rings which separate the two halves of the magnet.

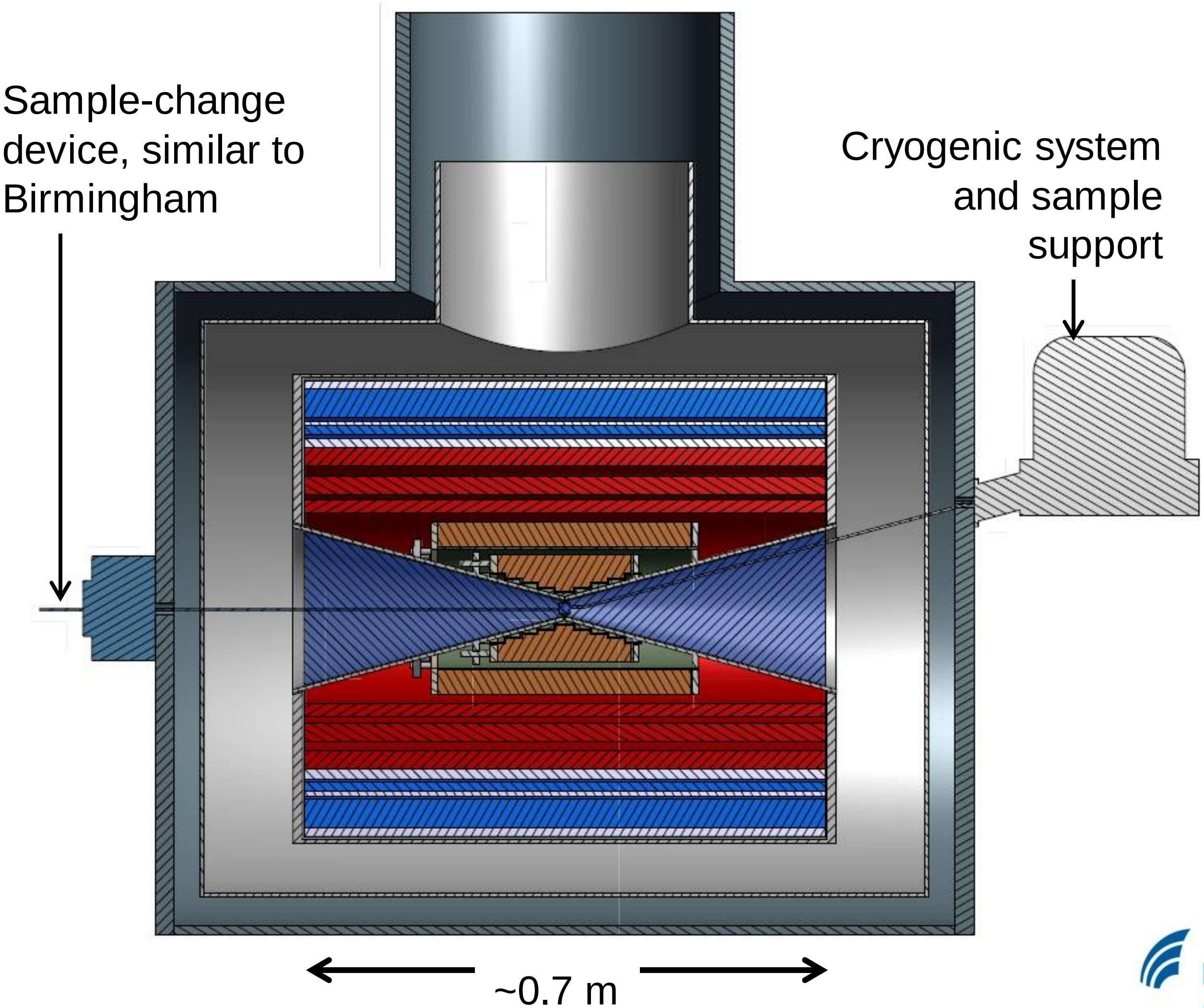
The sample is easily introduced from the top.

HELMHOLTZ CENTRE BERLIN for Materials and Energy

EUROPEAN SPALLATION SOURCE

MagLab/ESS Concepts

30 T Superconducting Conical Magnet



The conical configuration might attain higher field but changing samples requires more sophisticated manipulation.

Sample temperature control is also more challenging.

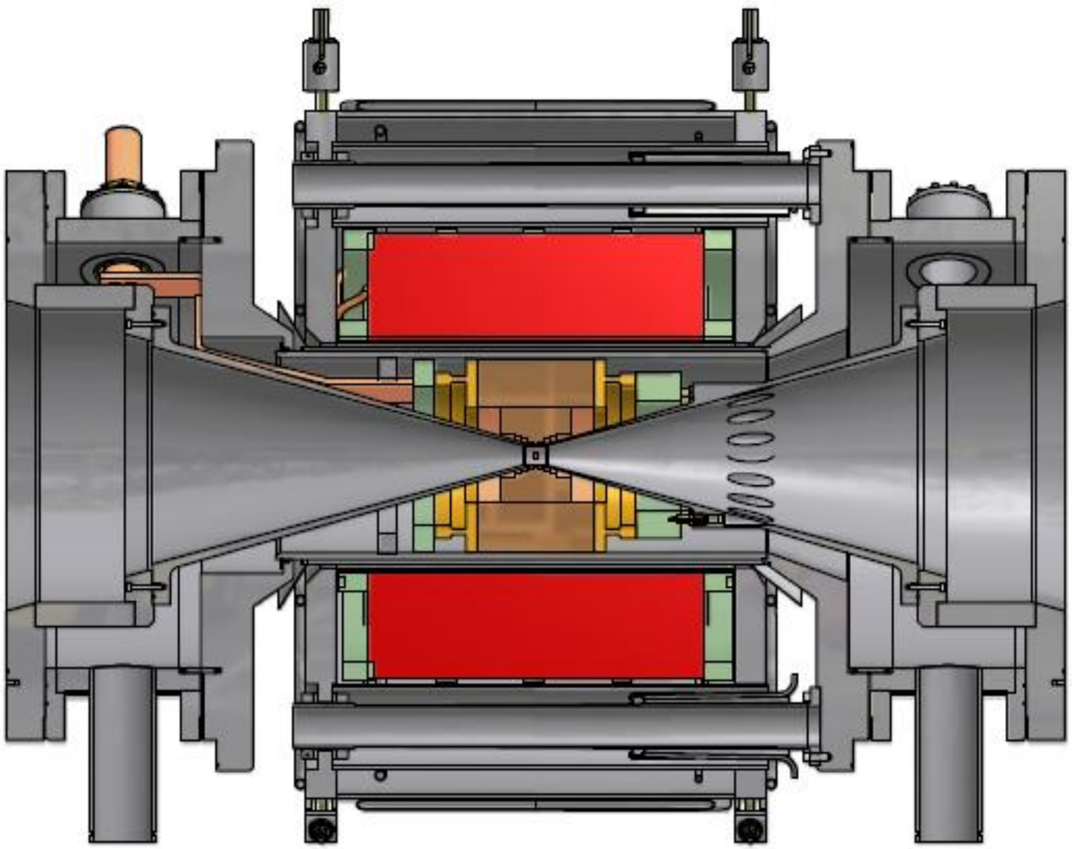
Phase I Design Report to be submitted mid-2012. Phase II funding discussion to start later in 2012.

HELMHOLTZ CENTRE BERLIN for Materials and Energy

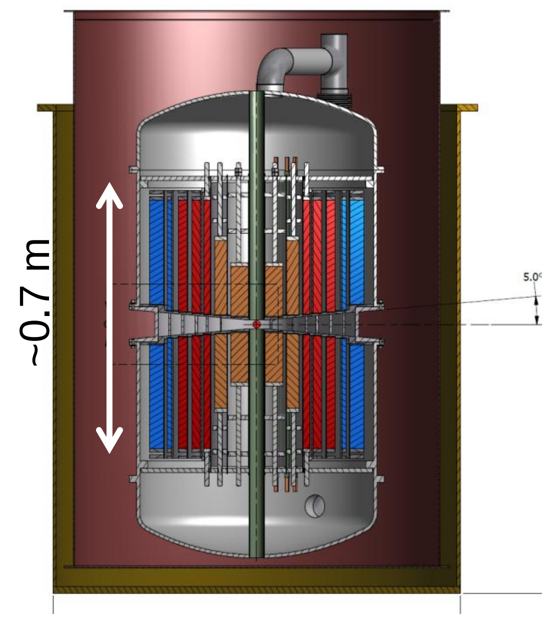
EUROPEAN SPALLATION SOURCE

25 T Neutron Magnet Concepts

Present-Generation
Conical Scattering Magnet
HZB, 25 T, 4.4 MW Hybrid



Next-Generation
Split Scattering Magnet
ESS, 25 T, 0 MW, YBCO



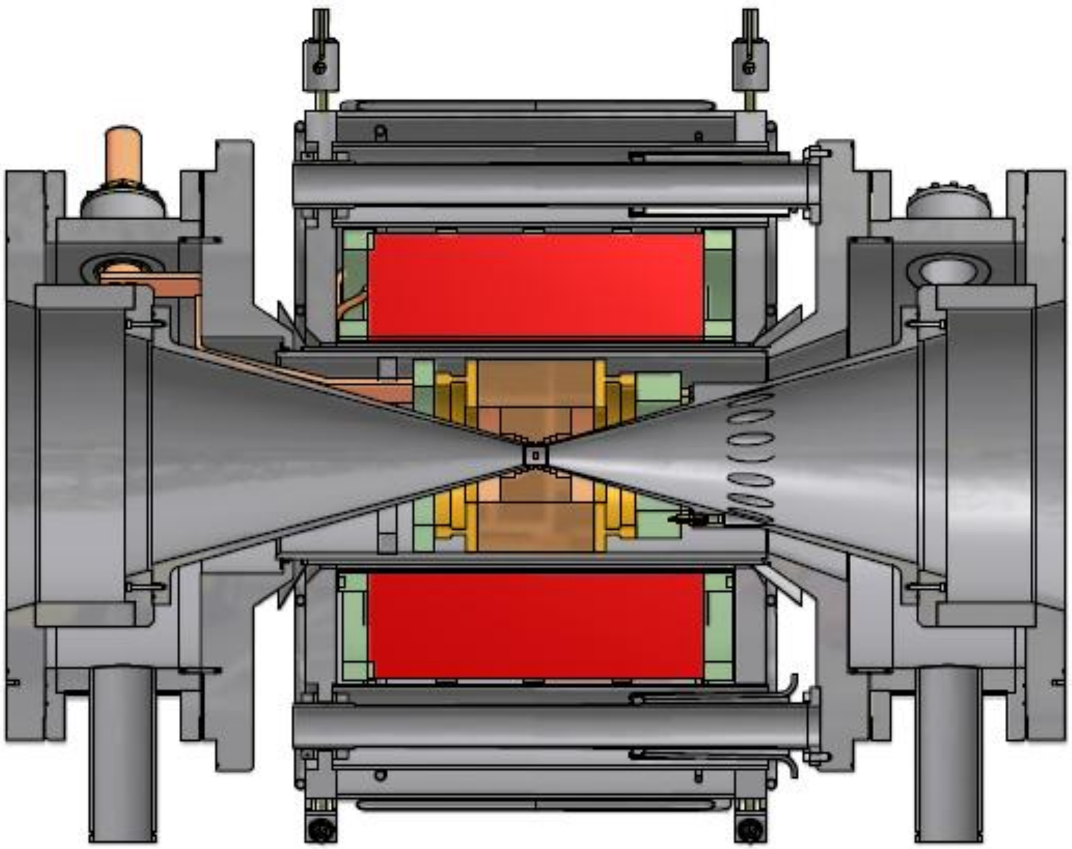
HELMHOLTZ CENTRE BERLIN for Materials and Energy

EUROPEAN SPALLATION SOURCE

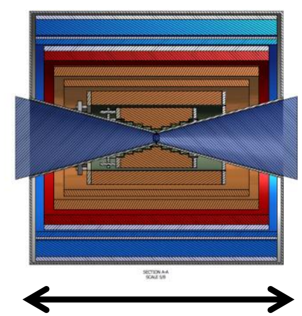
25 T magnet cold-masses … sc

30 T Conical Magnet Concepts

Present-Generation
Conical Scattering Magnet
HZB, 30 T, 8 MW Hybrid



Next-Generation
Conical Scattering Magnet
ESS, 30 T, 0 MW, YBCO



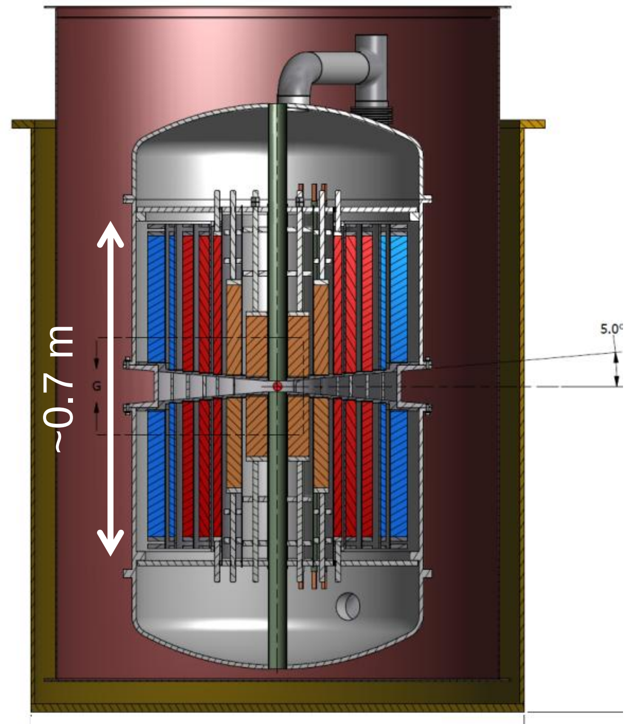
~0.7 m

30 T magnet cold-masses & support stru ar cale.

Split Neutron Magnet Concepts

FAT SAM @ SNS
16 T, Nb_3Sn & NbTi

Next-Generation
Split Neutron Scattering
Magnet
ESS Concept, 25 T
YBCO, Nb_3Sn & NbTi



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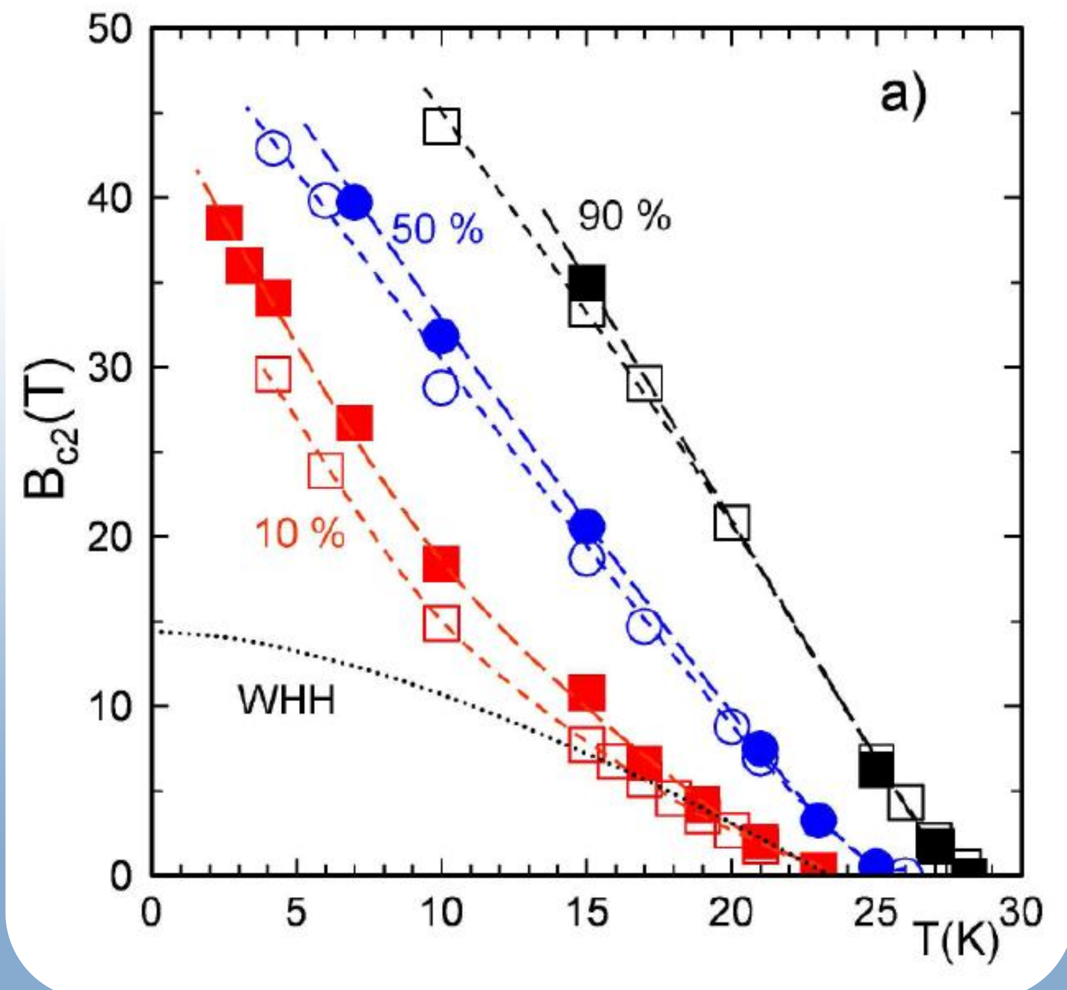
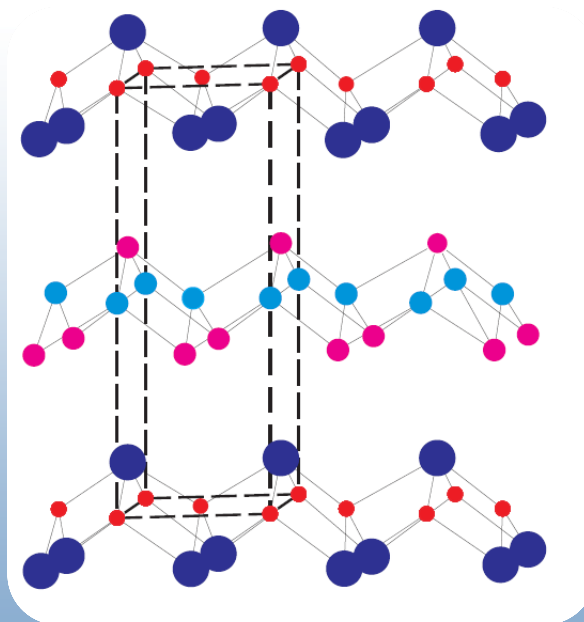
for Materials and Energy

EUROPEAN
SPALLATION
SOURCE

High field vortex matter: $La(O_{1-x}F_x)FeAs$



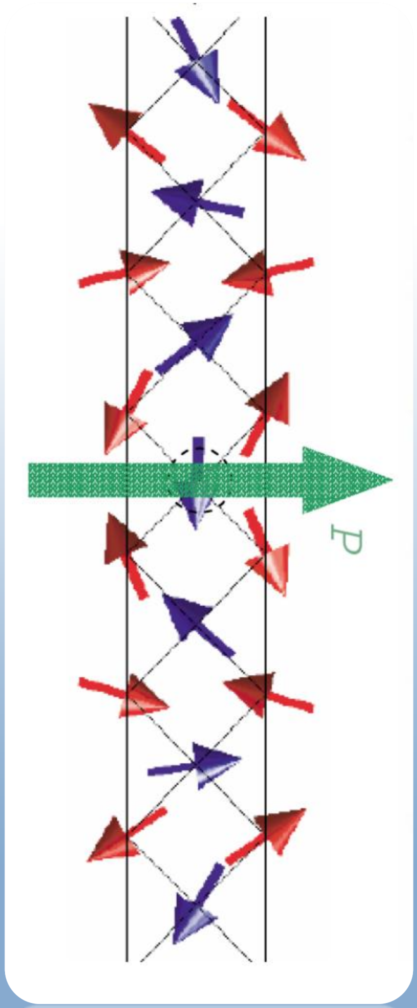
F. Hunte et al. (2008)

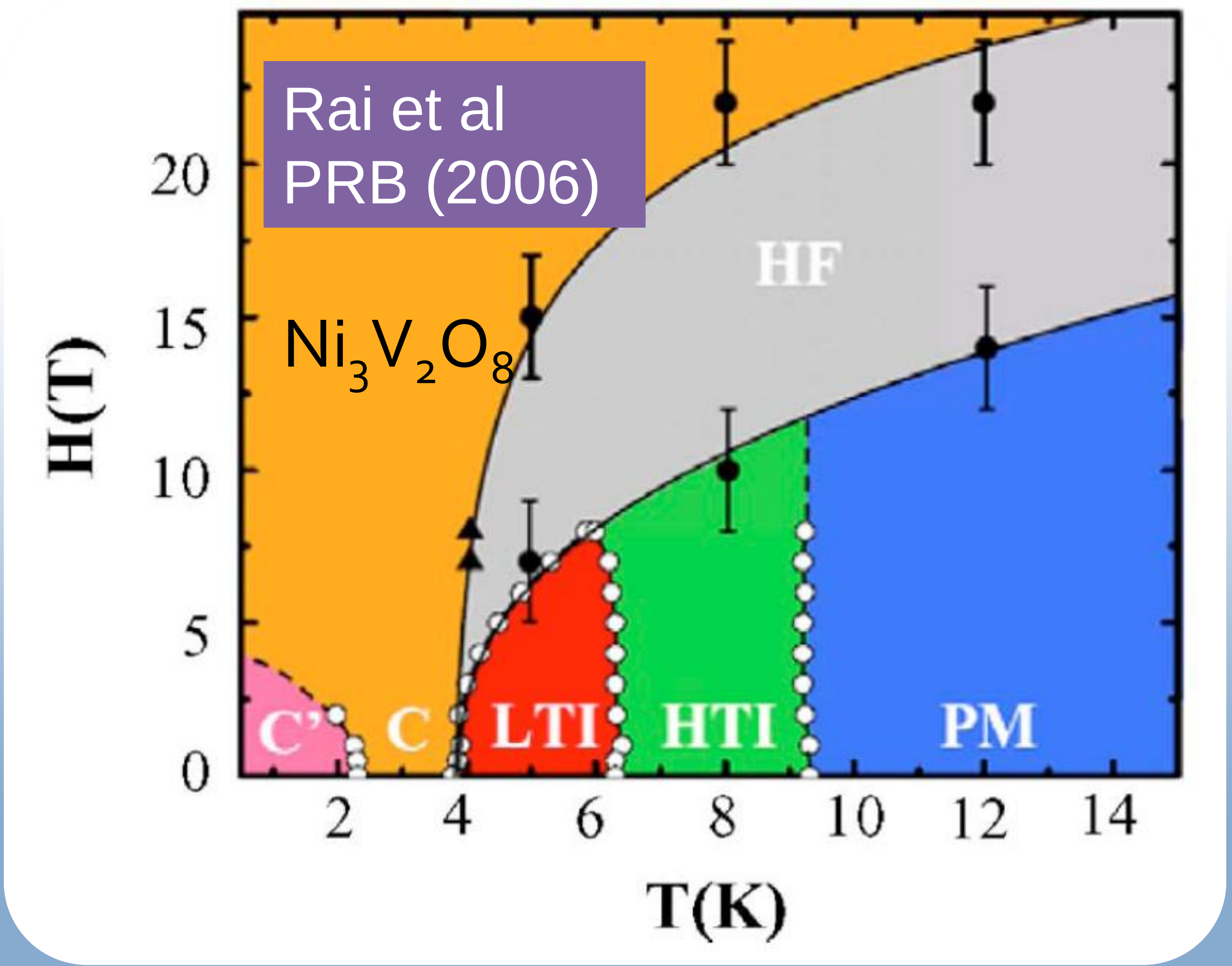


Technological use of superconductivity requires control of field induced vortext matter

Explore vortex matter in Higher field, higher T_C superconductors

Multiferroic oxides



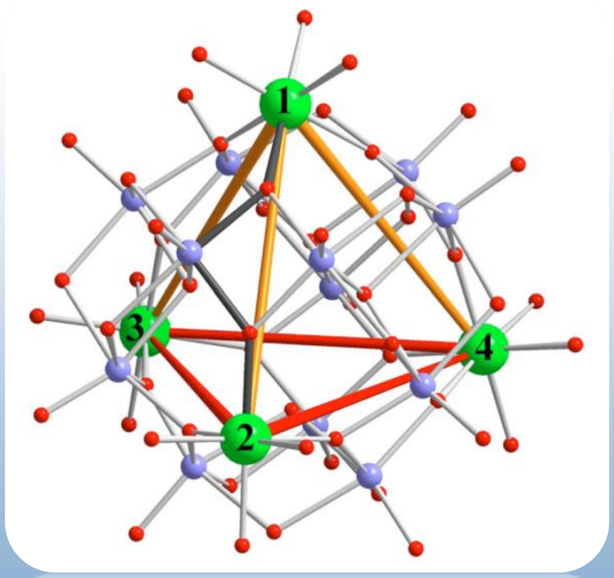


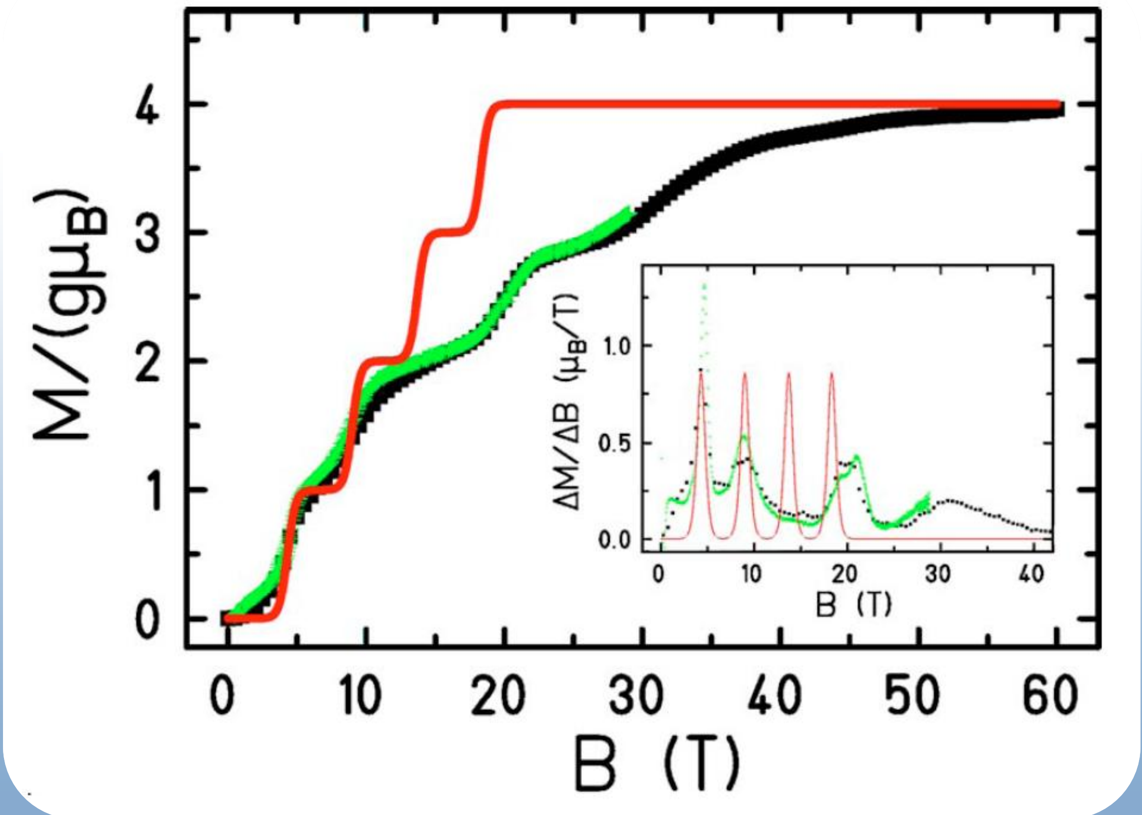
Materials that couple E&M are poorly understood and have great potential

Directly probe magneto-elastic coupling responsible for multiferroic response

Modified Exchange in Molecular Magnets

$\{Ni_4Mo_{12}\}$

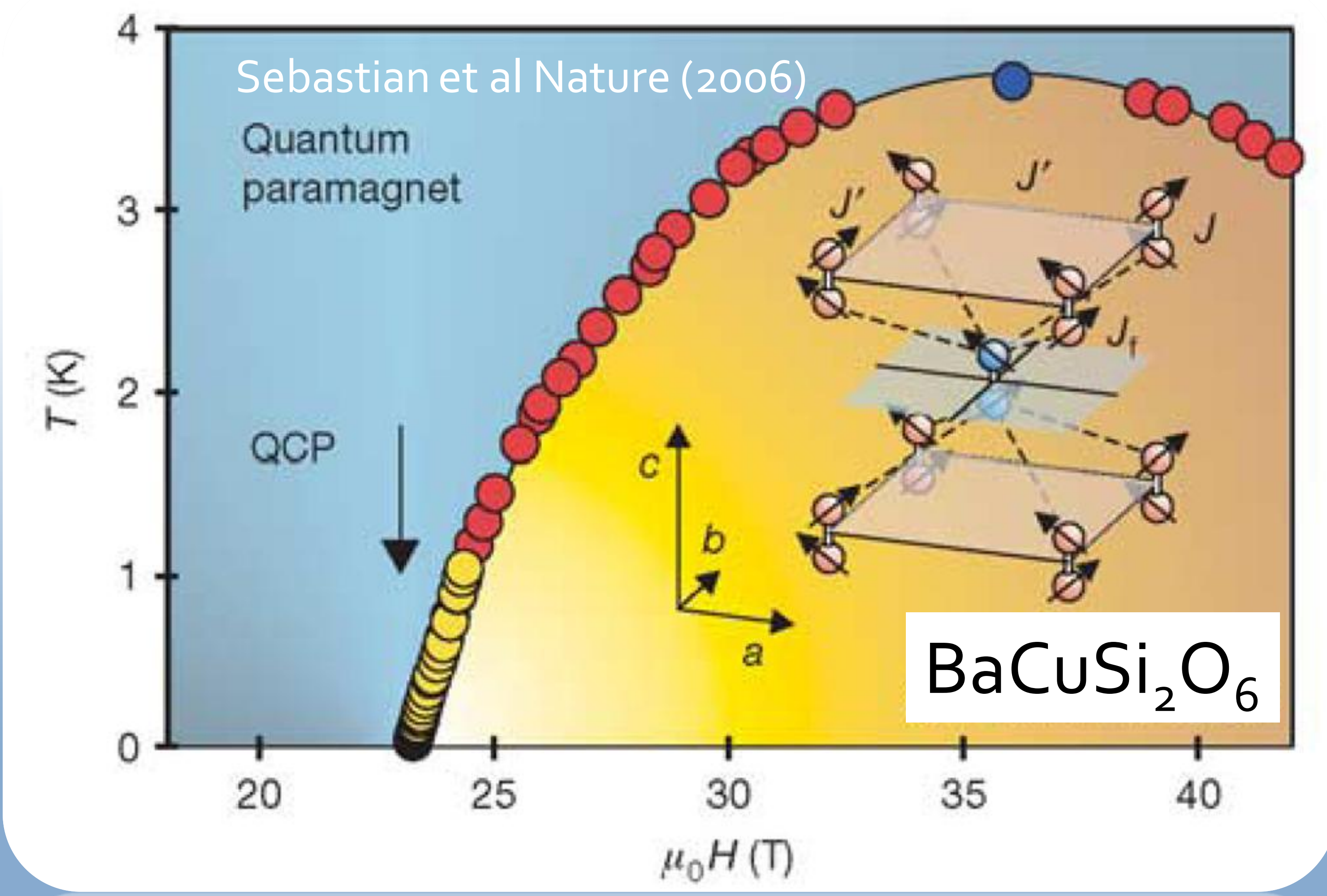




Molecular magnets are coherent quantum system with potential q-bit applications

Probe field induced changes of chemical & magnetic structure and interactions in molecular magnets

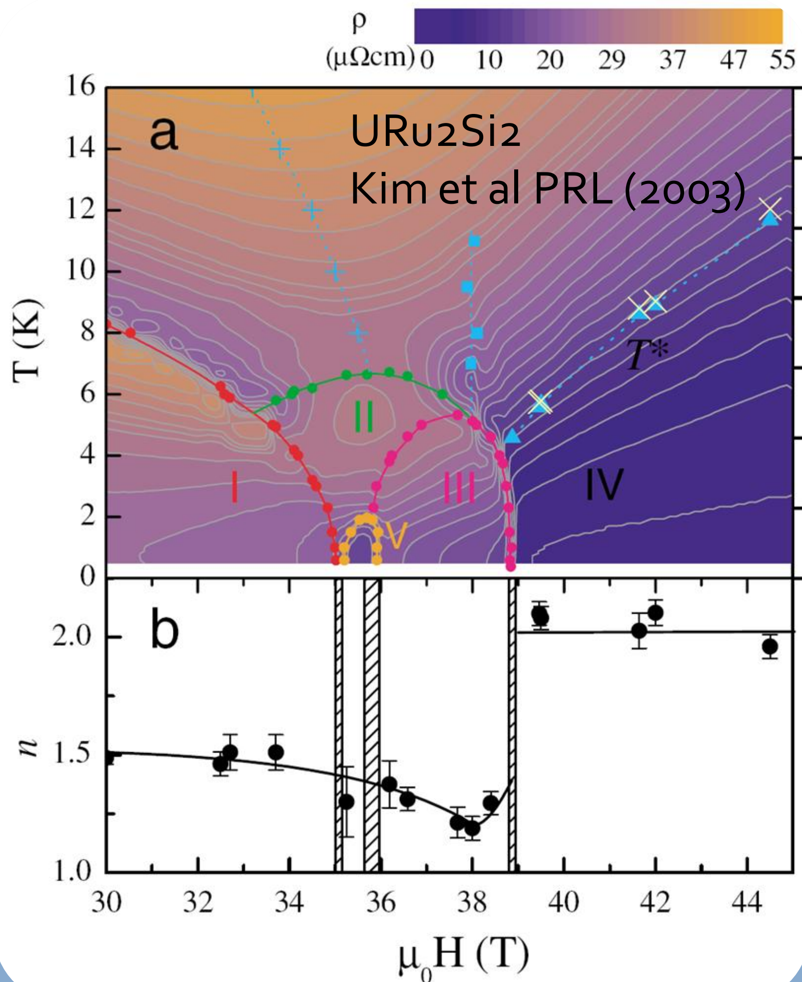
High Field Quantum Criticality



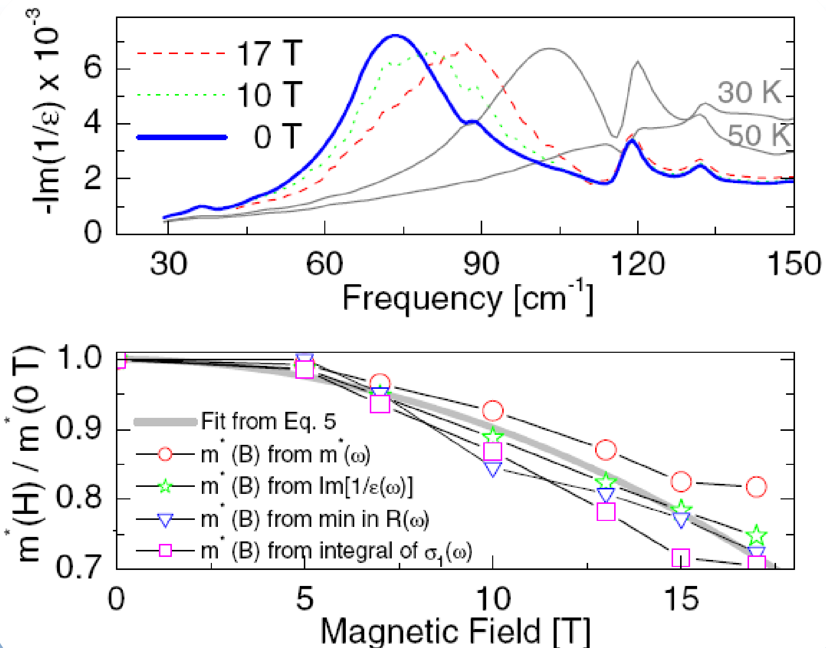
Much theory but few experiments access unique properties at quantum criticality

Determination of B-induced structure + static & dynamic quantum critical exponents

Field tuning heavy fermions



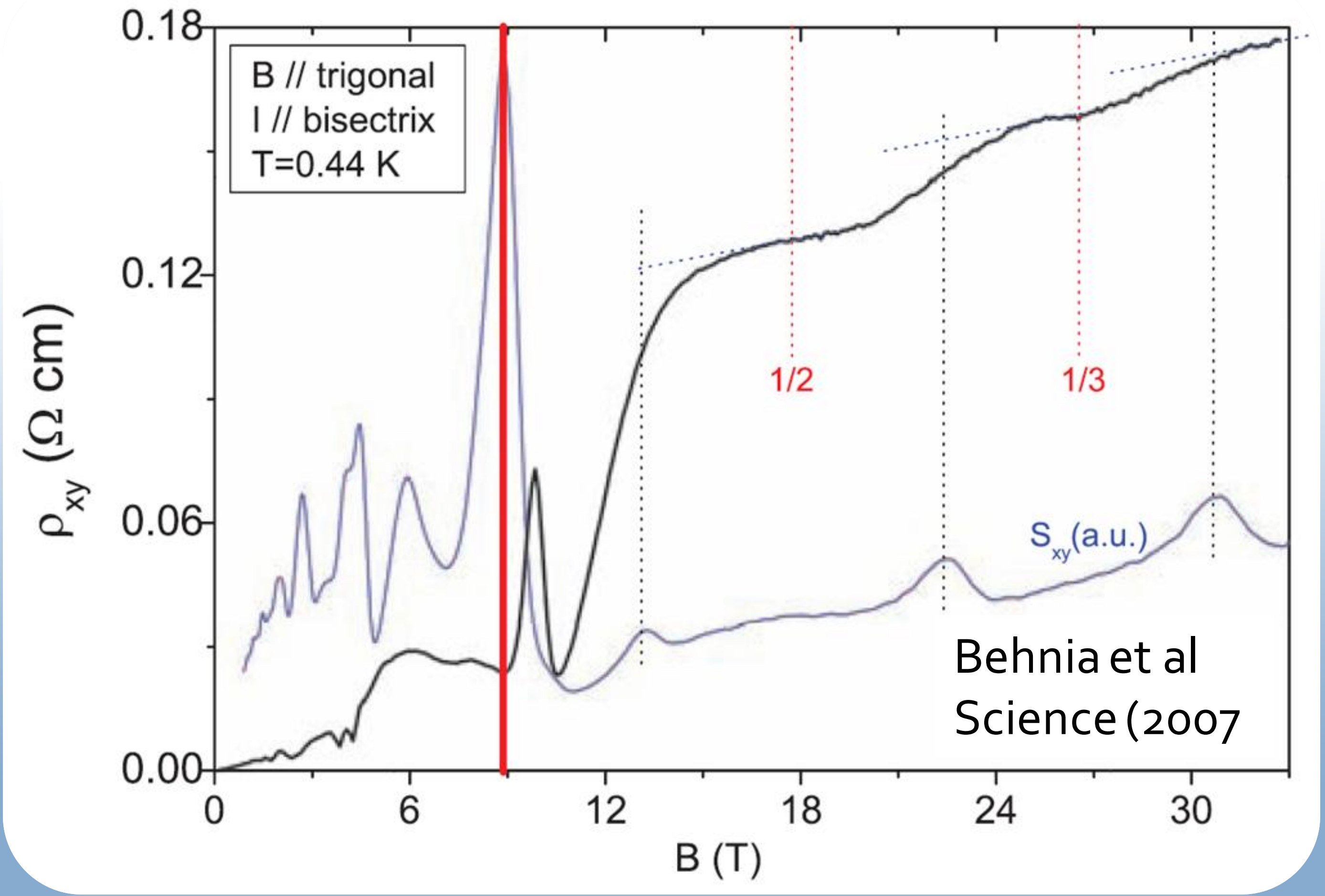
CeRu4Sb12
Dordevic et al PRL (2006)



Heavy fermion systems expose the complexity of strongly interacting electrons

Examine novel correlated phases with neutrons for a better overview of correlated physics

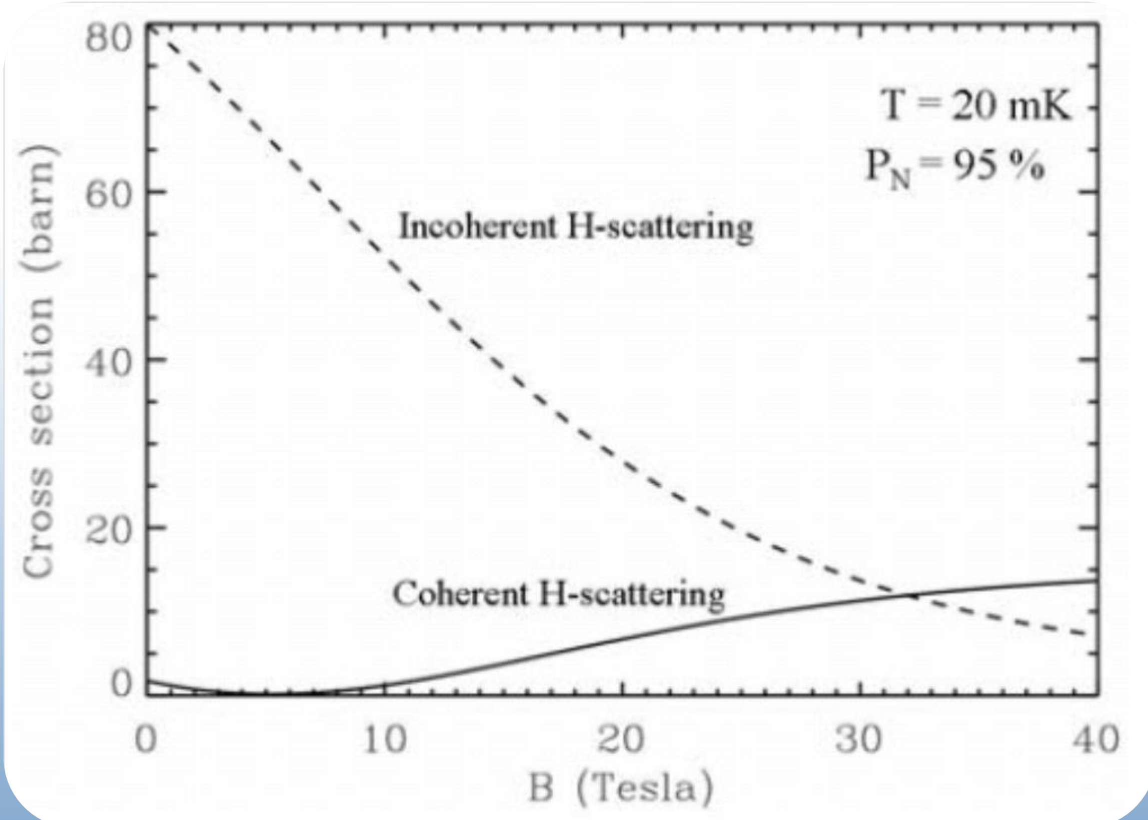
Fractionalized Quasi Particles in Bi

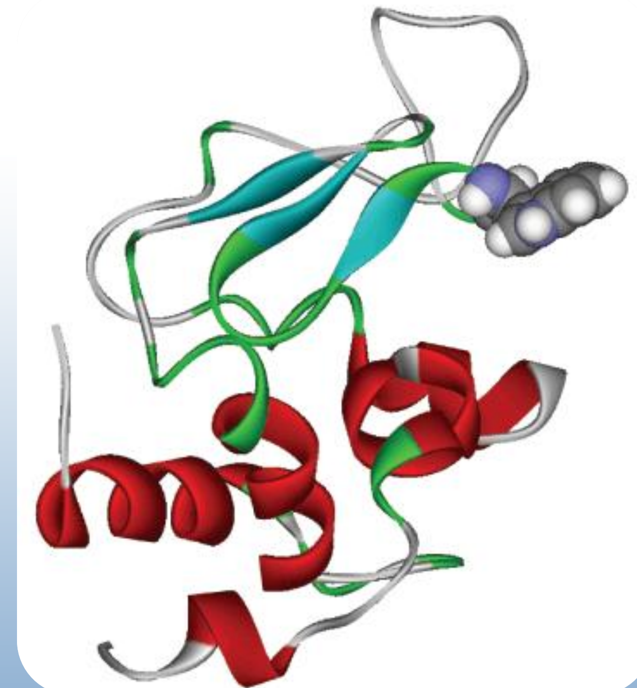


By forcing electrons into degenerate Landau levels, high fields induce novel correlated metallic phases

Neutrons can probe the correlated state wave function through diffraction and inelastic scattering

Locating hydrogen in protein structure

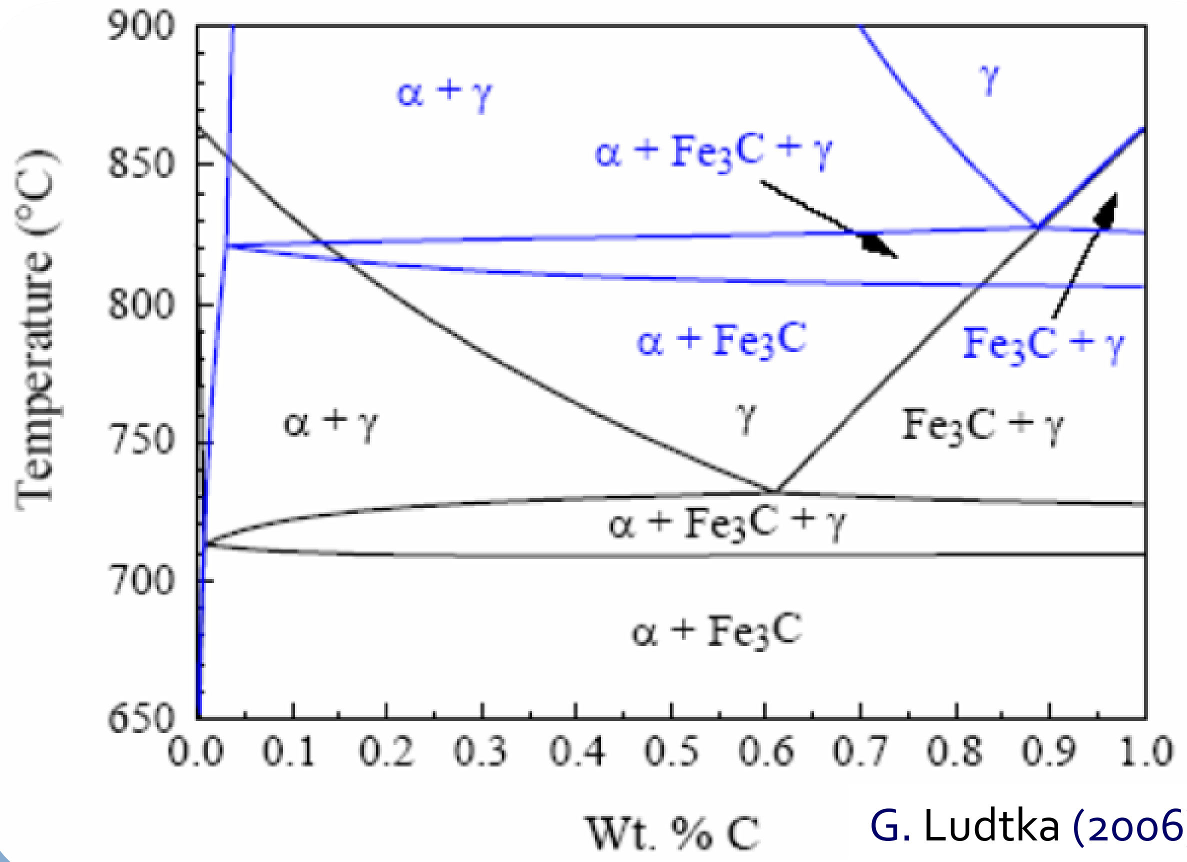




Materials functionality in biology
is critically dependent on hydrogen

By aligning proton spins
determine hydrogenous
structures without
multiple phasing crystals

High field metallurgical processing



G. Ludtka (2006)

Even minute improvements in structural properties of metals can have enormous technical implications

Determine structure and magnetization of field stabilized high temperature metallurgical phases

Conclusions

- The magnetic field is one of few controlled non-thermal perturbations to explore hard condensed matter
- Neutron scattering is essential to understand the field induced properties
- High field neutron scattering now reaches 17 Tesla and has made key contributions to quantum magnetism and superconductivity
- Higher fields enable a broader range of science including protein crystallography and metallurgy
- Pulsed field systems reach beyond 30 Tesla and are particularly promising for synchrotron sources
- New hybrid technologies are being implemented in Europe for >25 T steady state neutron scattering
- Much more compact fully superconducting YBCO based systems are now feasible for neutron scattering.
- Lower costs, higher fields, and better access for neutrons make these recent developments very exciting
- Combined with developments in sources and instrumentation a new generation of high field neutron scattering can impact a broad range of materials science
- By virtue of the NHMFL the US leads the relevant magnet technologies but there is no funded project to make scientific use of this for neutron scattering