Progress with DRACULA ?

Diffractometer for
Rapid
ACquisition
Ultra
Large
Areas

DRAC, first presented at the ILL "Instrument Day" 26 Feb 2002
DRAC, highest priority for Instrument Committee 17 Oct 2003

Pilgrims Progress…

Pulsed Souces are Best?

No Available Beam Tube

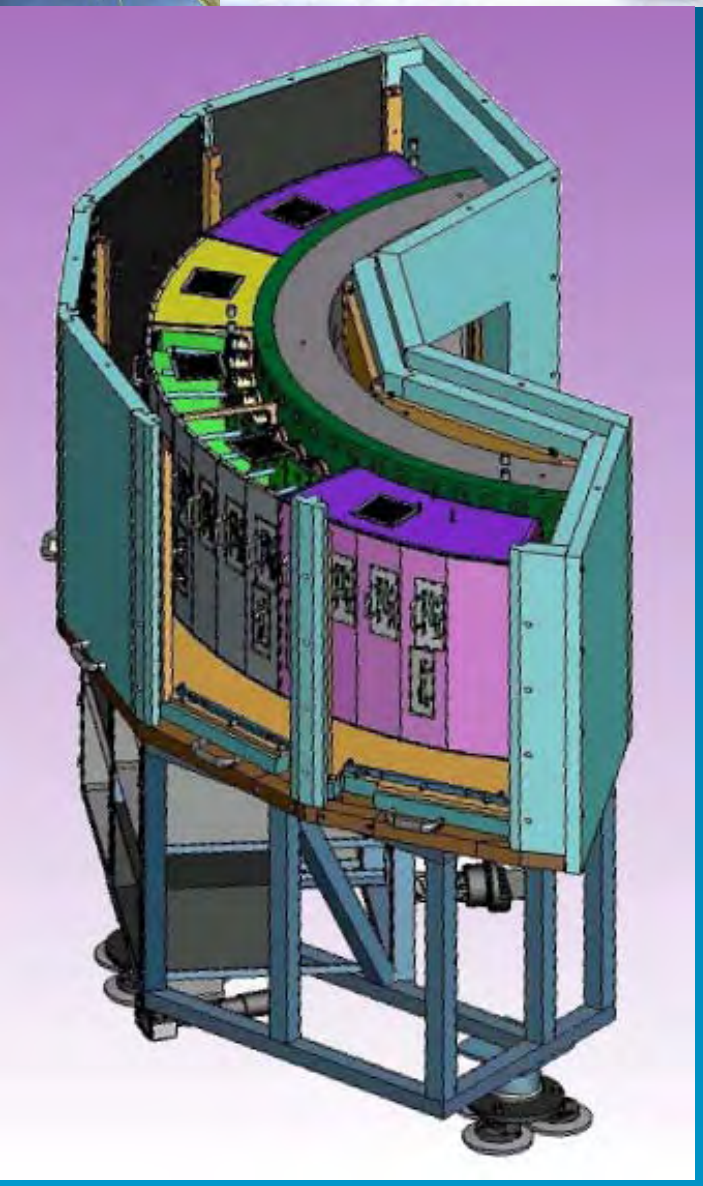
Undermines ESS ?

No Money/Manpower

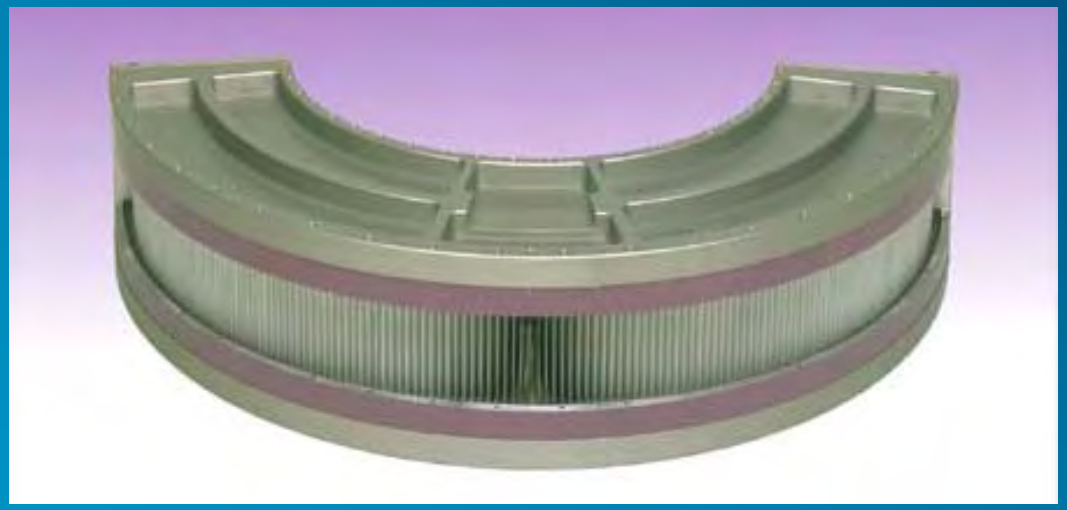
Too Many D's, upgrade D20?

No Pretty Pictures

DRACULA - What do we want to do ?

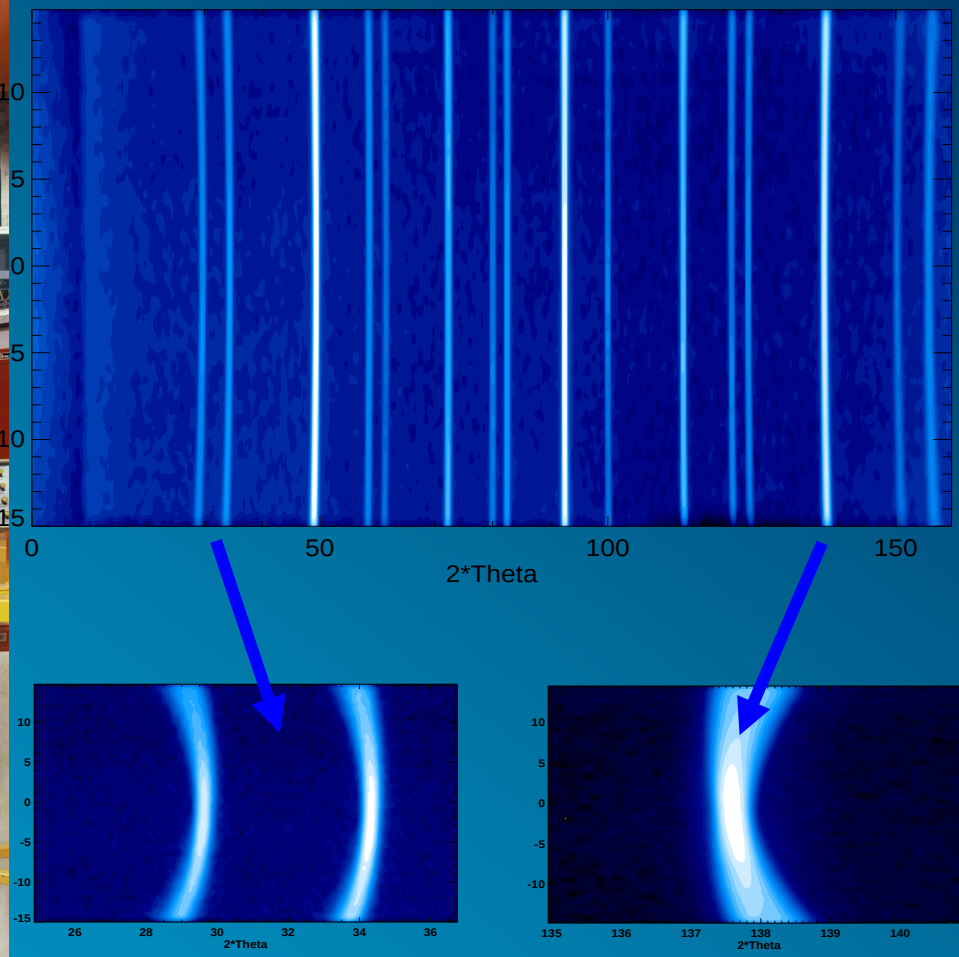


- Large, compact 2D area detector (cf D19)
- Order of magnitude faster than D20
- Competitive in speed to best SNS machine
- Moderate resolution over whole detector
- Special sample environments, v.small samples
- Radial collimator, Low background



2D detectors for CW Powder Diffraction Super-D2B, SPODI at FRM2, ANSTO…

UK-EPSRC Super-D2B project at ILL

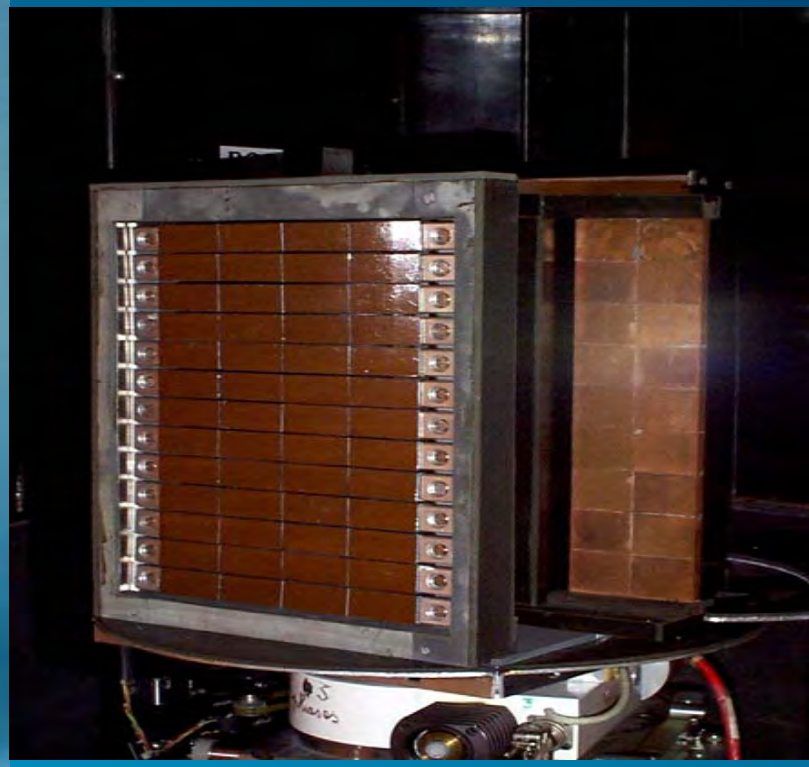


E.Suard, C.Ritter, A.Hewat, P.Attfield… (Edinburgh)

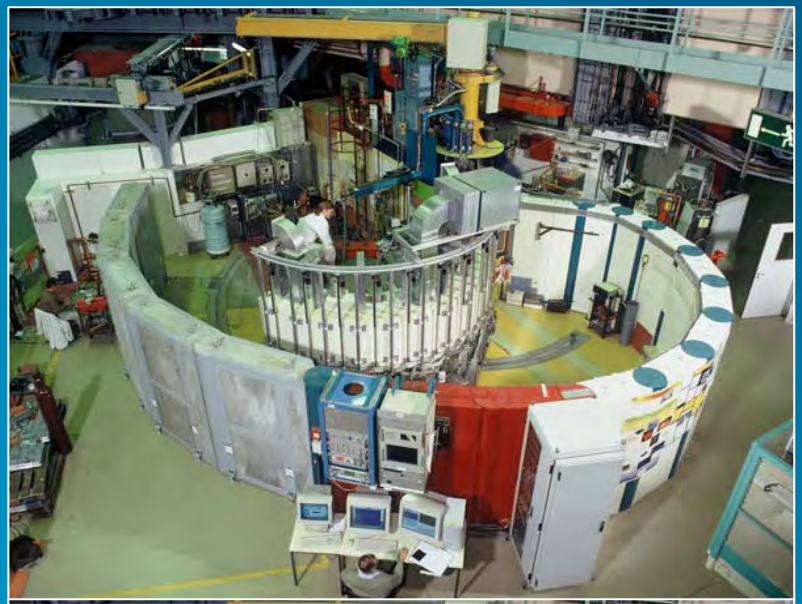
Big monochromators give very high flux D2B, D20, IN8…

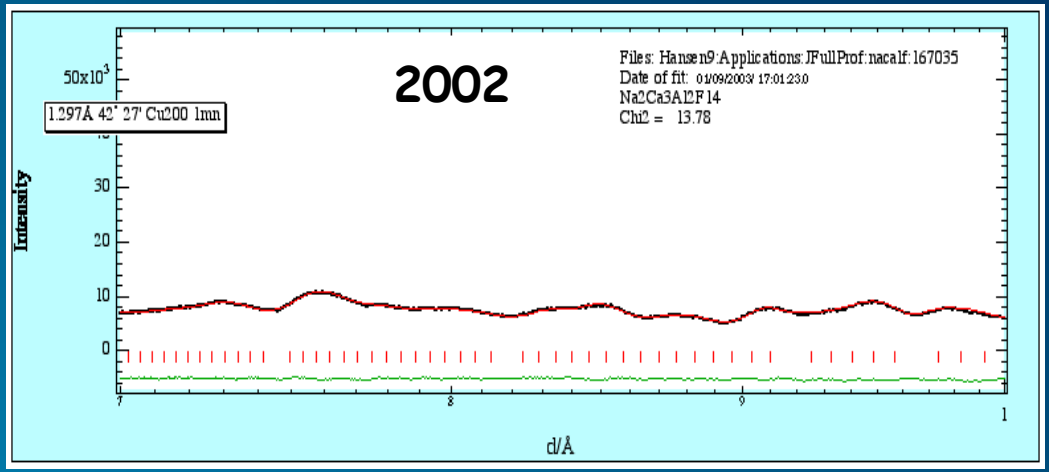
Very high flux on the sample

- D2B 1.0×10^{7} $n.cm^{-2}.sec^{-1}$
- D20 9.8×10^{7} $n.cm^{-2}.sec^{-1}$
- IN8 6.5×10^{8} $n.cm^{-2}.sec^{-1}$

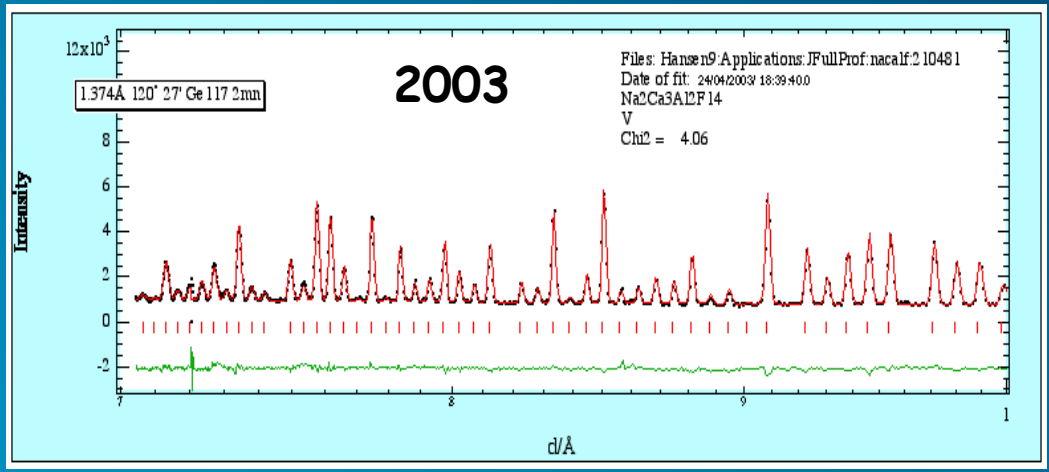


High flux compatible with good Resolution
High take-off option on D20



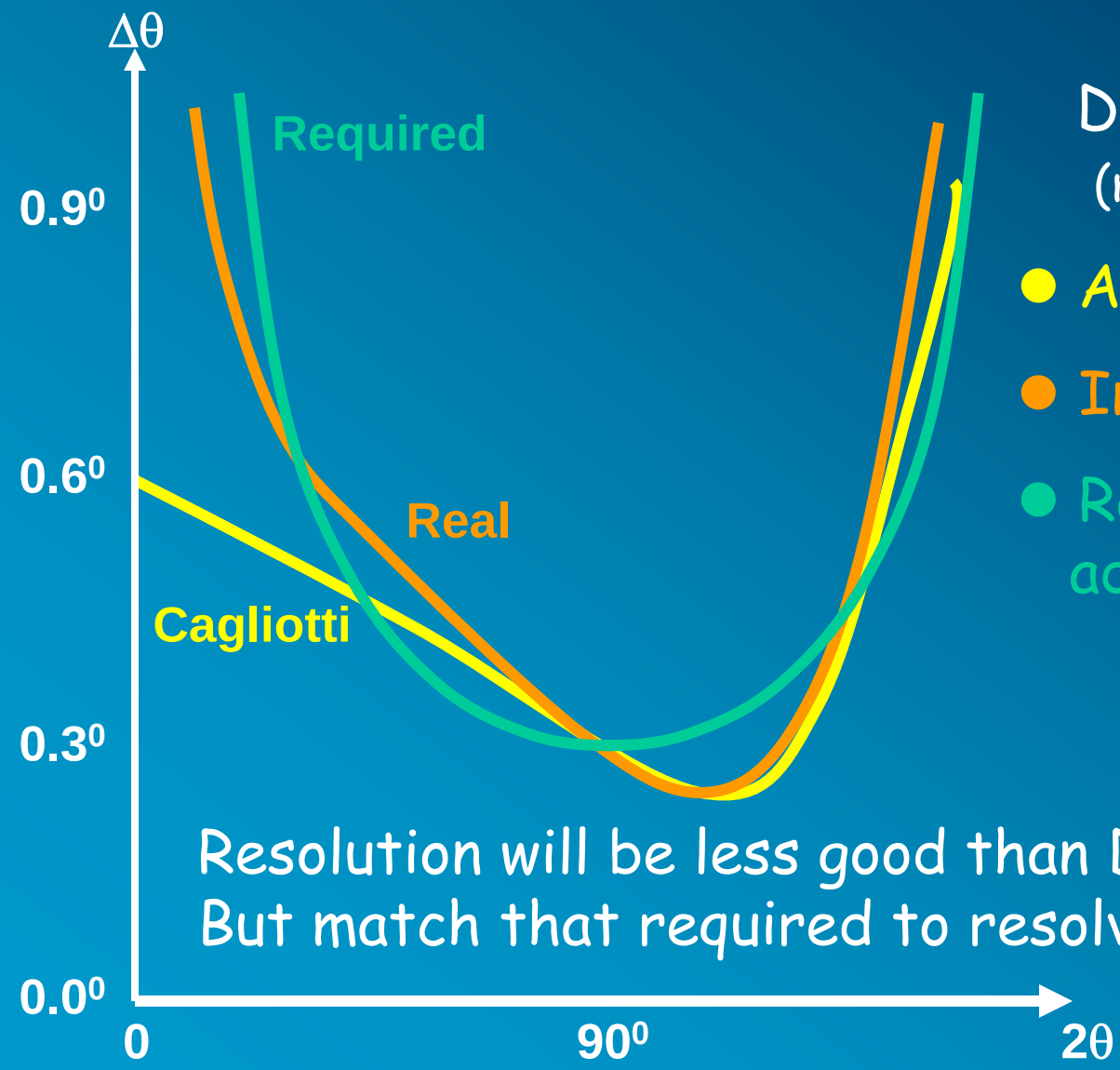


Before and After (data in 2 min.)



Higher D20 resolution since 2003

High flux compatible with good Resolution
DRACULA resolution matches peak density



DRACULA resolution
(minimum $\Delta d/d \sim 3*10^{-3}$)

- According to Caglioti
- Including vertical div.
- Required to resolve adjacent peaks

Resolution will be less good than D20 "hi-resolution"
But match that required to resolve adjacent peaks

Shoot the Dragon…

Pulsed Source are Best

I agree…

- Provided Europe has a high flux pulsed source (ESS)
- For very high resolution backscattering…
- But not for high intensity, moderate resolution
- We cannot compete with the American SNS if we only have ISIS, a medium flux pulsed source…

Comparison of TOF & CW Diffractometers

Jorgensen, J.D., Cox, D.E., Hewat, A.W., Yelon, W.B (1984)

"Scientific opportunities with advanced facilities for neutron scattering"

Shelter Island Workshop, 1984

Nuclear Instruments and Methods in Physics Research B12 (1985) 525-561

Efficiency for a given resolution = time averaged flux on sample
* sample volume
* detector solid angle

P.G. Radaelli, S. Hull , H.J. Bleif & A. M. Balagurov (2001)

ESS Instrumentation Group Reports

"Powder Diffraction Instruments"

Comparison of TOF & CW Diffractometers

The time-averaged **Flux*Detector** criterium

With big detectors we can compete with the SNS

The time-average sample flux is higher on a CW source.

	D20	GEM	DRACULA	SNS
Flux average on sample	5×10^7	$\sim2\times10^6$	$\sim10^8$	$\sim2.5\times10^7$
Detector solid angle	0.27 sr	4.0 sr	1.5 sr*	3.0 sr
Efficiency=Flux*Detector	1.7	1	18	9

* Based on new D19 detector: R=760 mm, h=400 mm, 800 linear resistive wires $30^o\times160^o$

A High Resolution SNS Powder Diffractometer

B.E.F. Fender and A.W. Hewat, Jan. 78

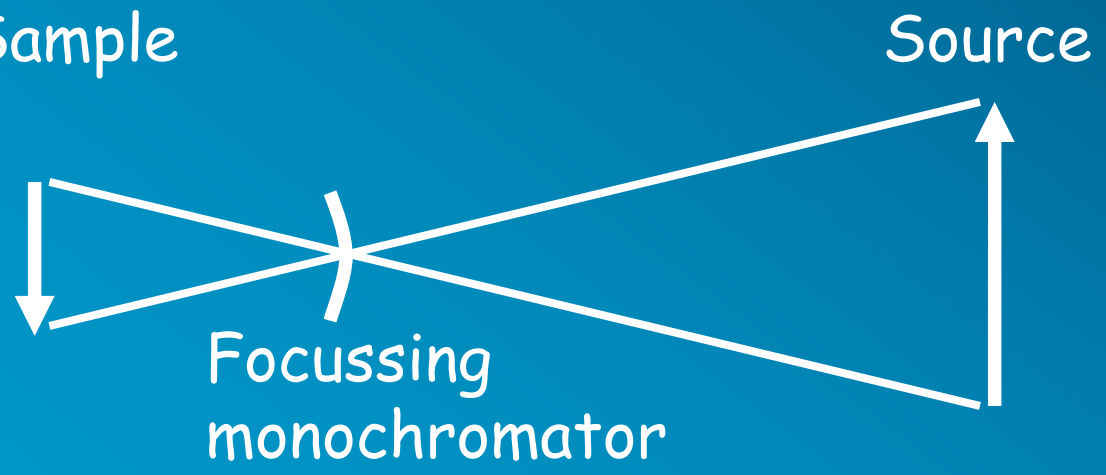
The HRPD proposed for the Rutherford SNS[1)] is essentially Steichele's original design[2,3,4)], as developed on a pulsed source by Windsor and Sinclair[5)],but with the following features. For comparison with a conventional HRPD see also Fender[6)] and Hewat[7)].

Constructed as HRPD at ISIS

Why is sample flux so high from a reactor?

A: Large vertically focusing monochromators ?

No ! Focusing in real space only gives a factor of x2 or x3

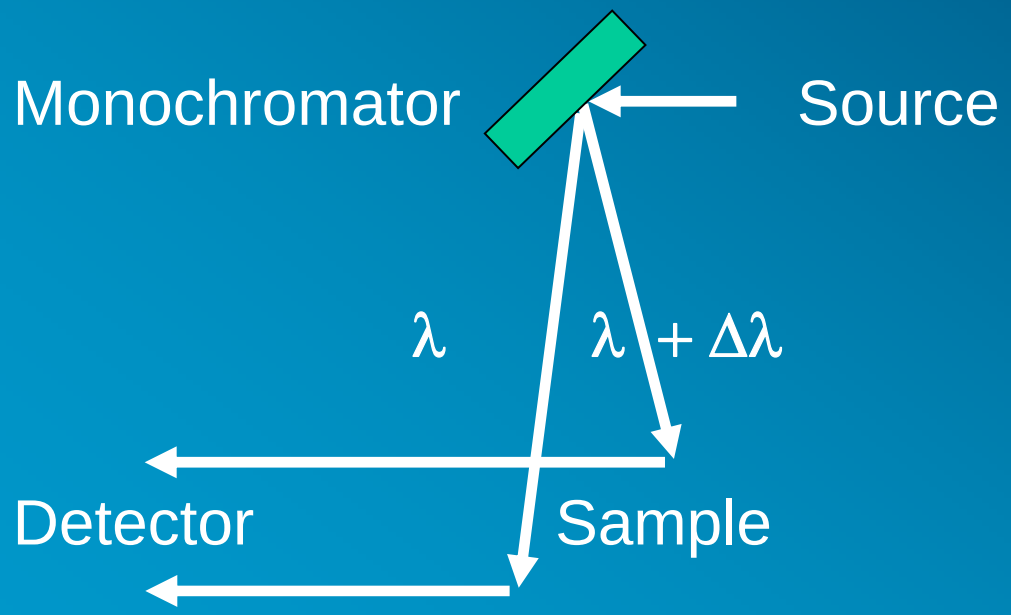


cf use of convergent guide with TOF

Why is sample flux so high from a reactor?

A: Large wavelength-band focusing monochromators ?

Yes ! Focusing in reciprocal space can give a factor of x10

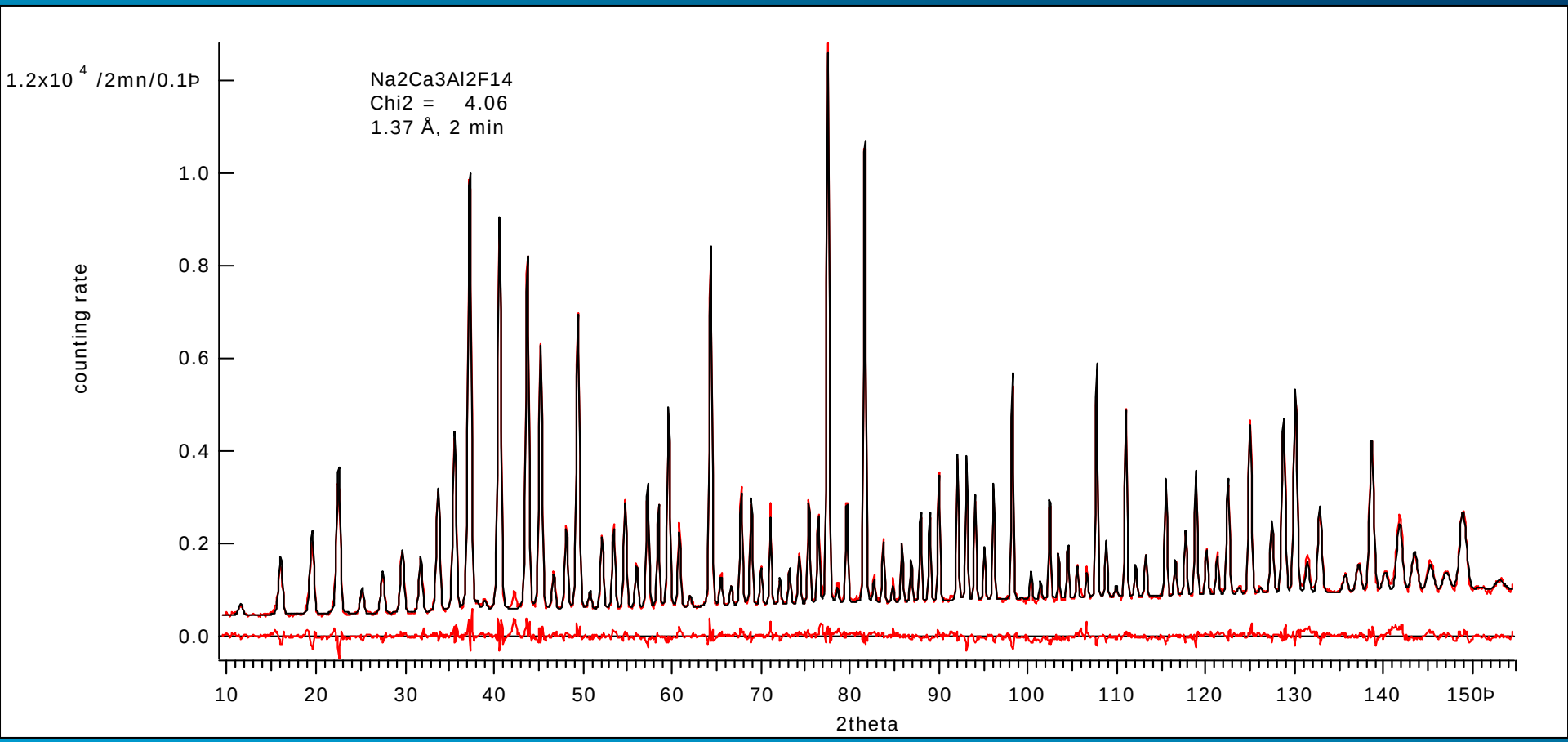


$\Delta d/d \sim 0.1\%$ for $\Delta\lambda/\lambda \sim 1\%$

D20 – Good Resolution but still very fast

Thomas Hansen (2003) ILL News, June 2003

2 minute D20 data for a ~700 mm^3 sample of $Na_2Ca_3Al_2F_{14}$



Shoot the Dragon…

Don't believe it !

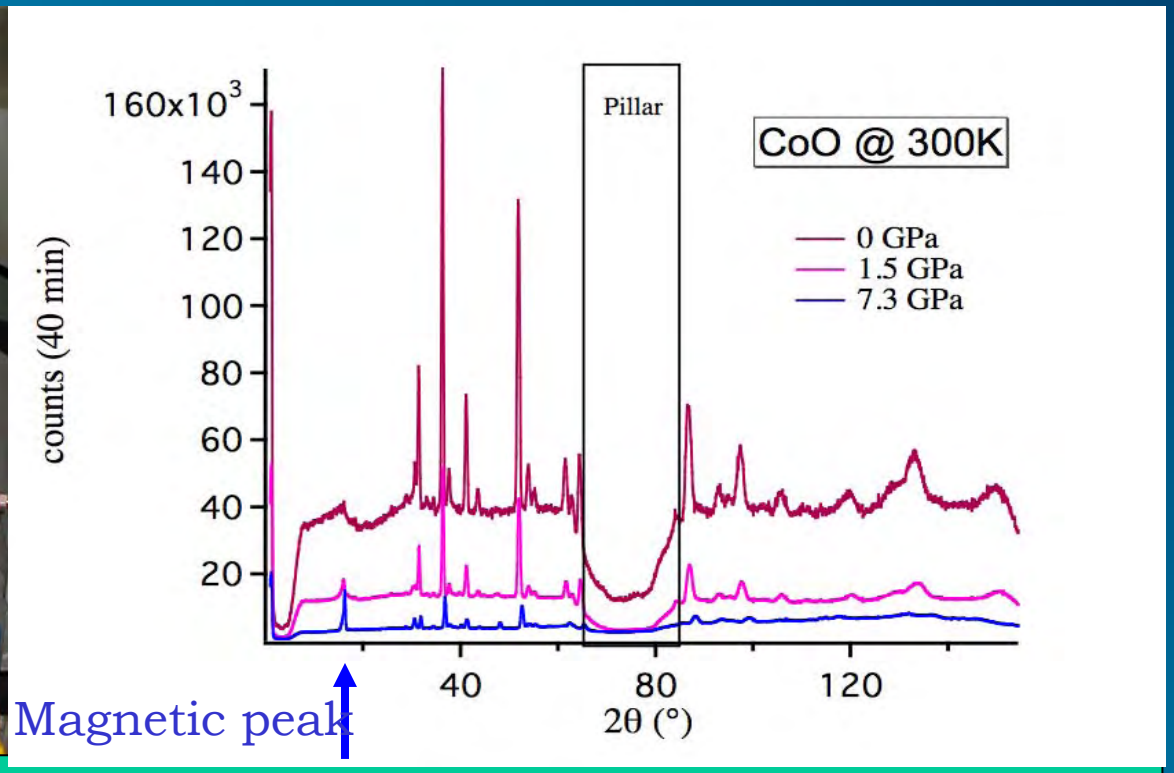
- We have EPSRC money unclaimed from the D2B project
- We have CCLRC money promised for new instruments…
- We propose using detectors already developed for D19
- The D19 detector is being built by ILL staff on CDD's
- CCLRC money is for the "full cost" materials+manpower

Applications –Small Samples, Low Background

D20 with "large" Paris-Edinburgh Pressure Cell (50 Kg)

Kernavanois et al. (2003) Advanced Millennium Pressure Project

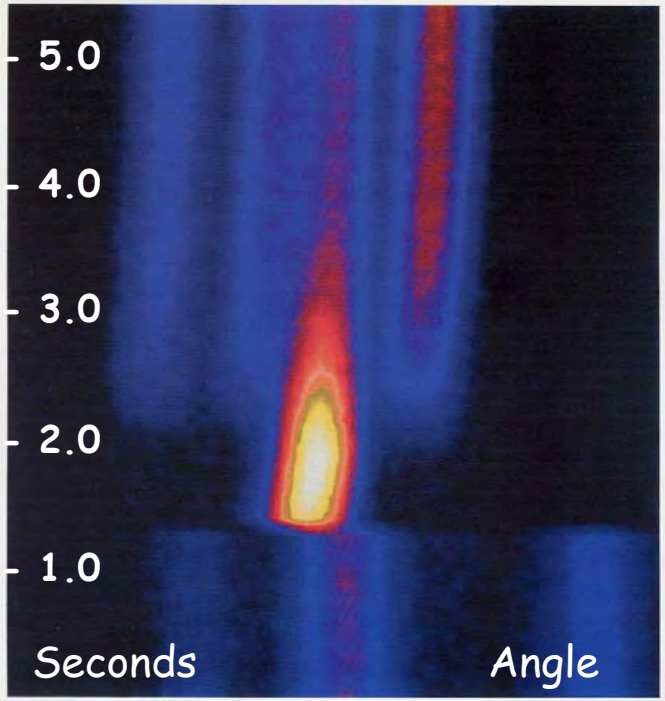
40 minute D20 data for a 100 mm^3 sample of CoO at 7.3 GPa



BUT low temperatures -> smaller cells -> 1-10 mm^3 samples

Applications –Small Samples, Fast Detectors

Very fast chemical and electrochemical kinetics

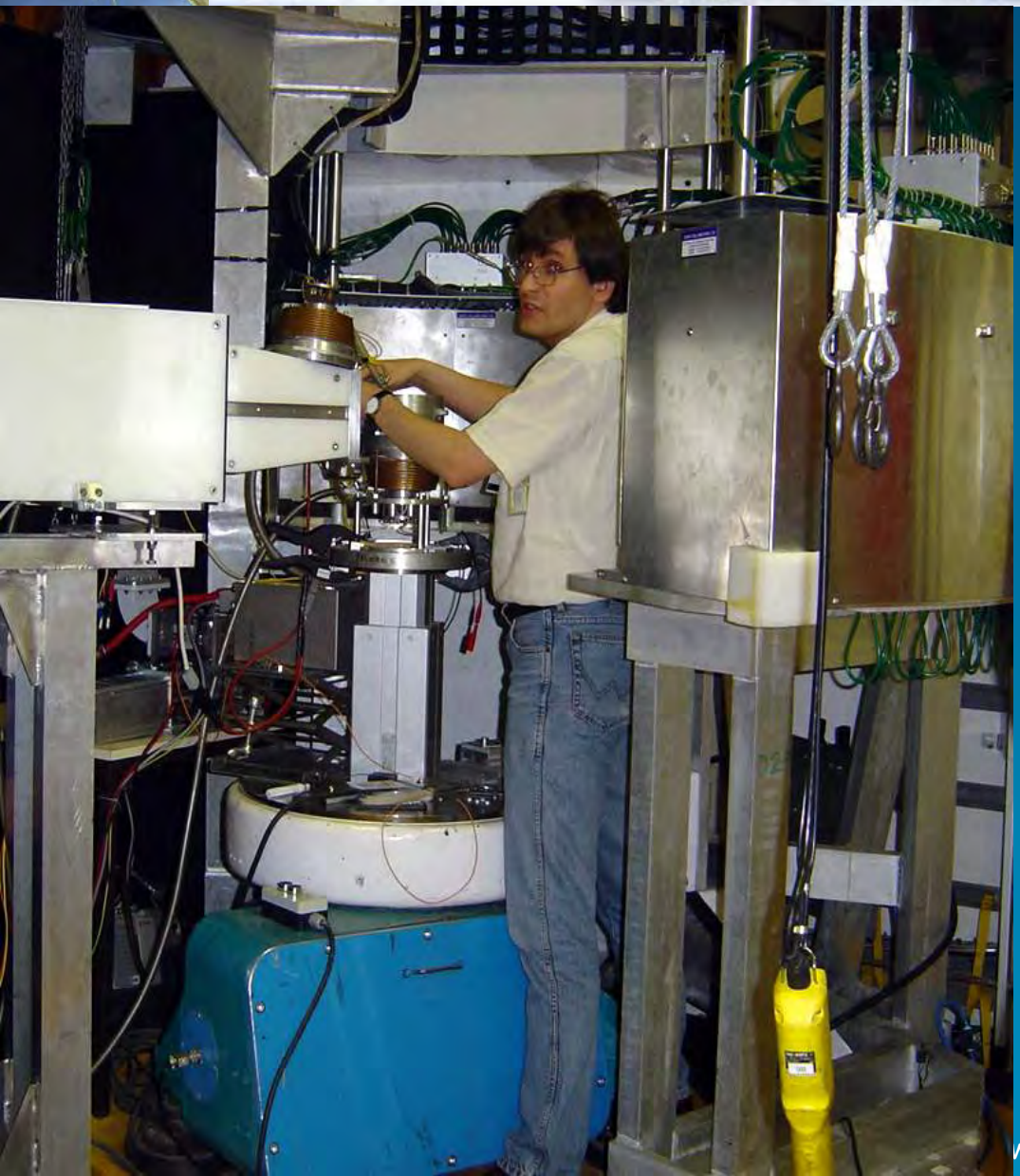


Journal of the American Ceramic Society
Incorporating Advanced Ceramic Materials and Communications
Volume 85 Number 10 October 2002

- The explosive SHS reaction was studied in real time with neutrons
- The reaction is exothermic, & heats the sample to 2200°C in <1 sec
- The complete diffraction pattern (left) is collected at 300 ms intervals - **A World Record**

D.Riley, E.Kisi, T.Hansen, A.Hewat (2002)

Applications –Samples, Complex environments



High-T Microwave Furnace Super-D2B (Boysen et al.)

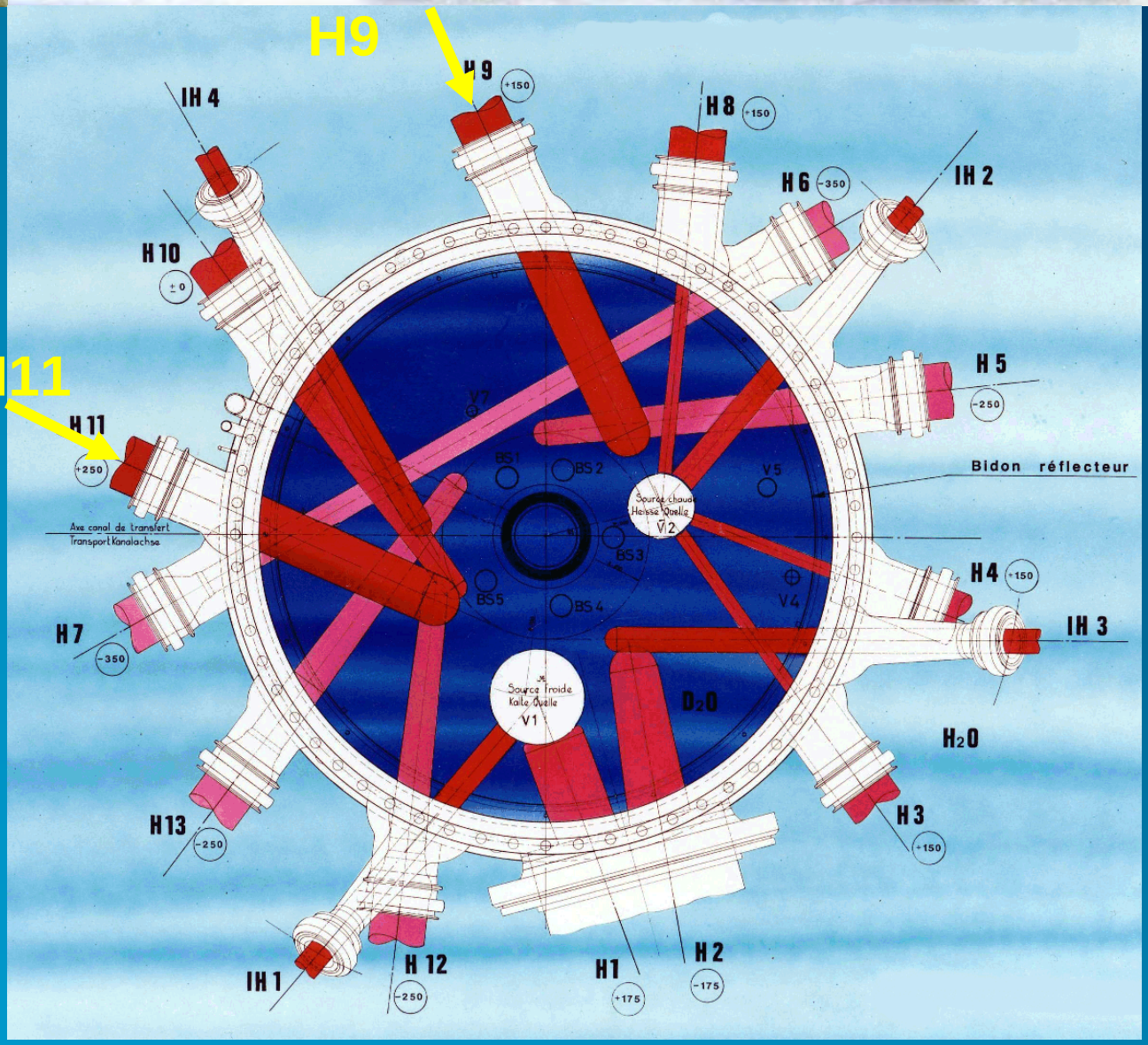
…with Carsten Korte from Giessen (2004)

Shoot the Dragon…

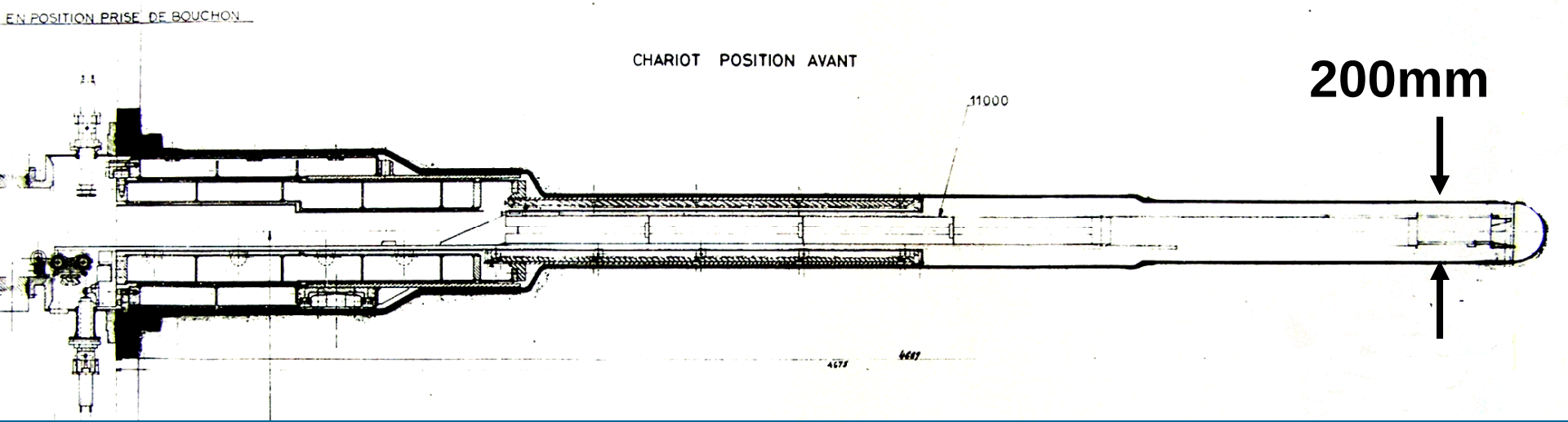
No Available Beam Tube

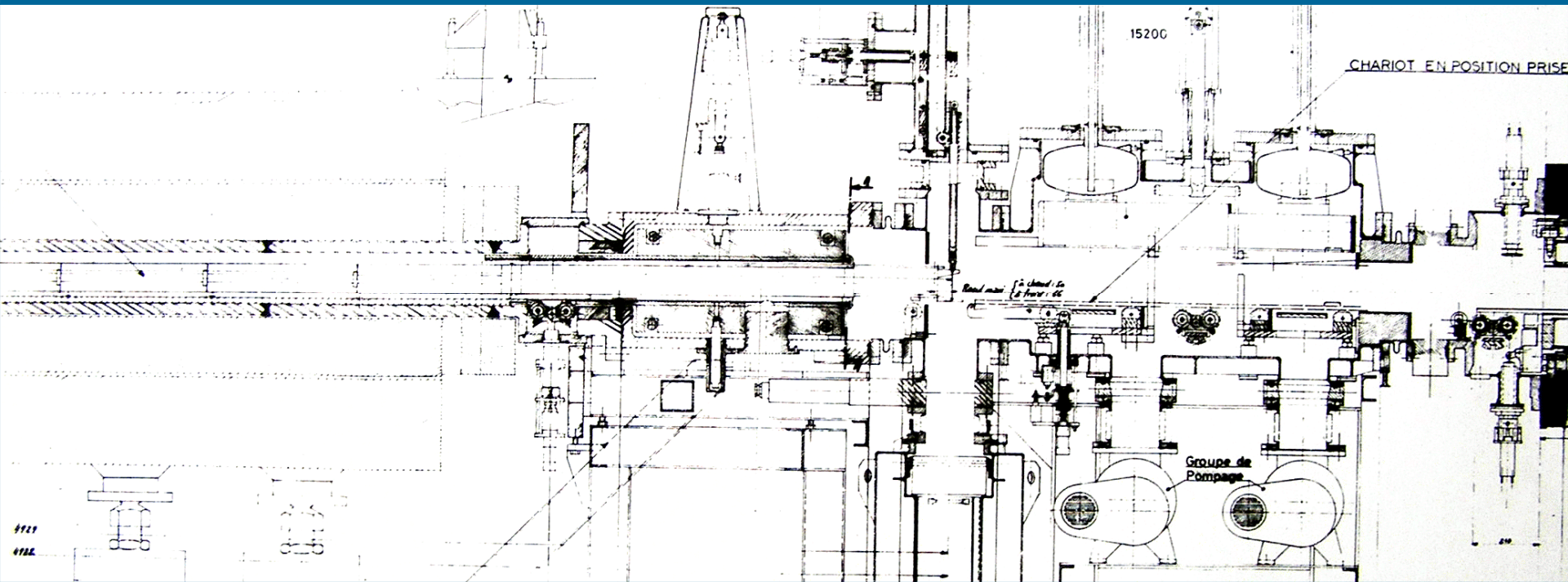
- DRACULA needs a high flux thermal beam tube
- The H9 beam tube has ILL's highest flux
- Similar requirements for TOMOGRAPHY & DRACULA

High Flux ILL Thermal Beam-Tubes

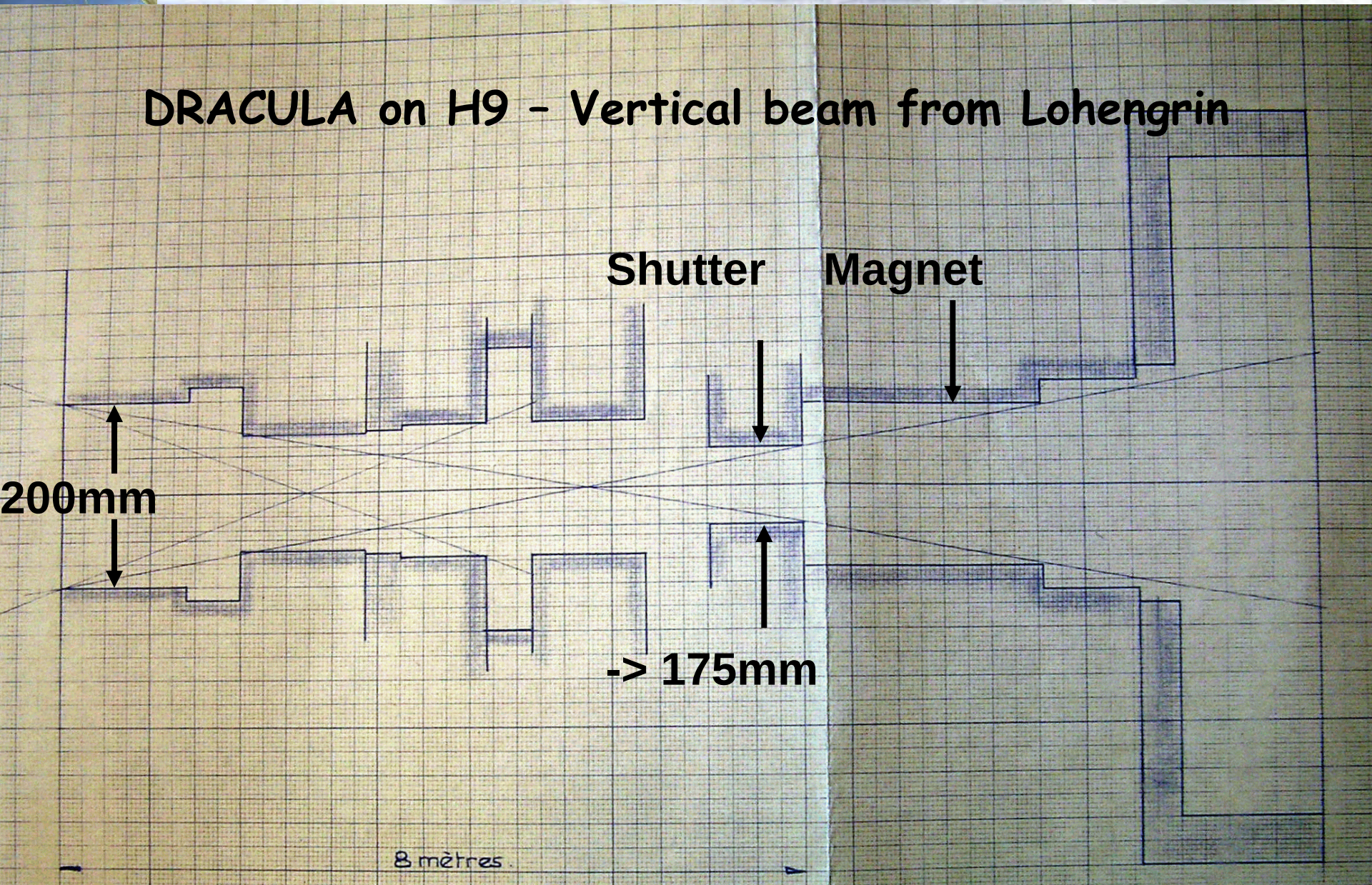


DRACULA on High Flux Beam Tube H9
H9 similar dimensions to H11 (D20)

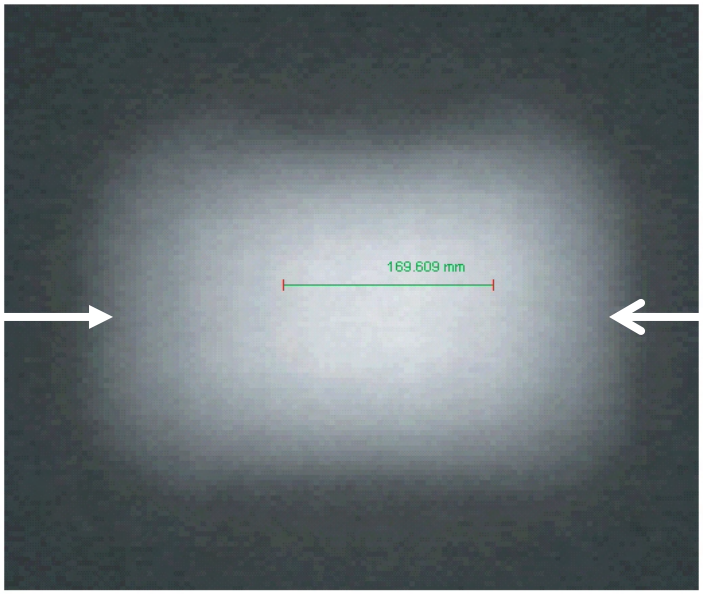


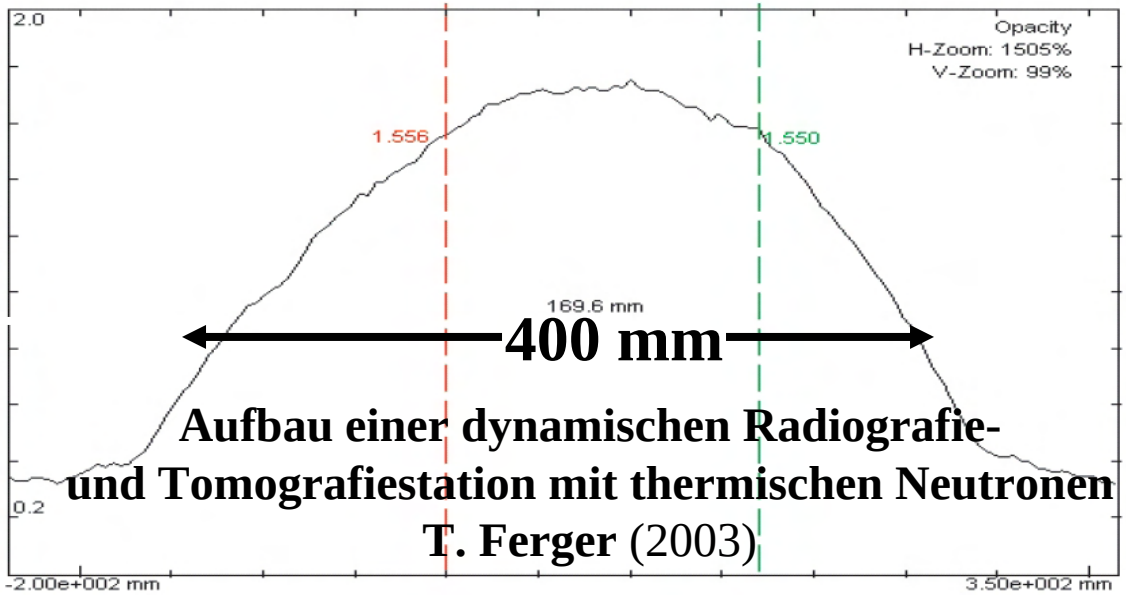


DRACULA on High Flux Beam Tube H9
H9 similar dimensions to H11 (D20)



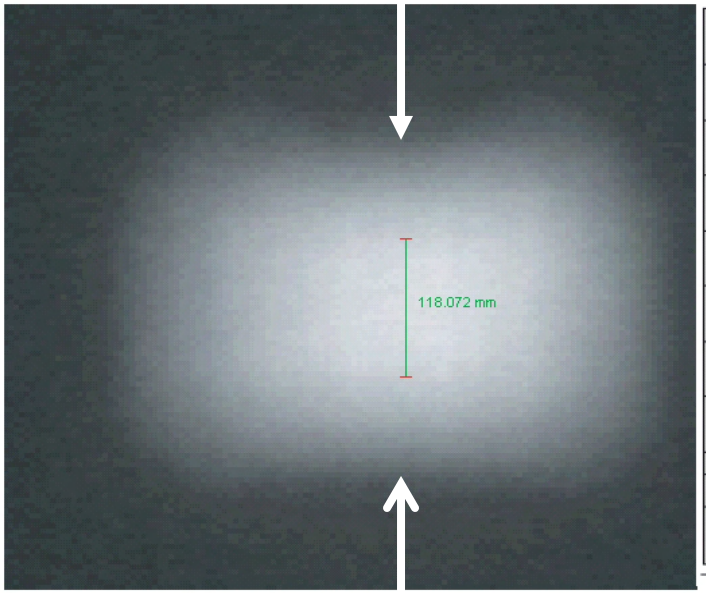
DRACULA on High Flux Beam Tube H9
H9 similar dimensions to H11 (D20)

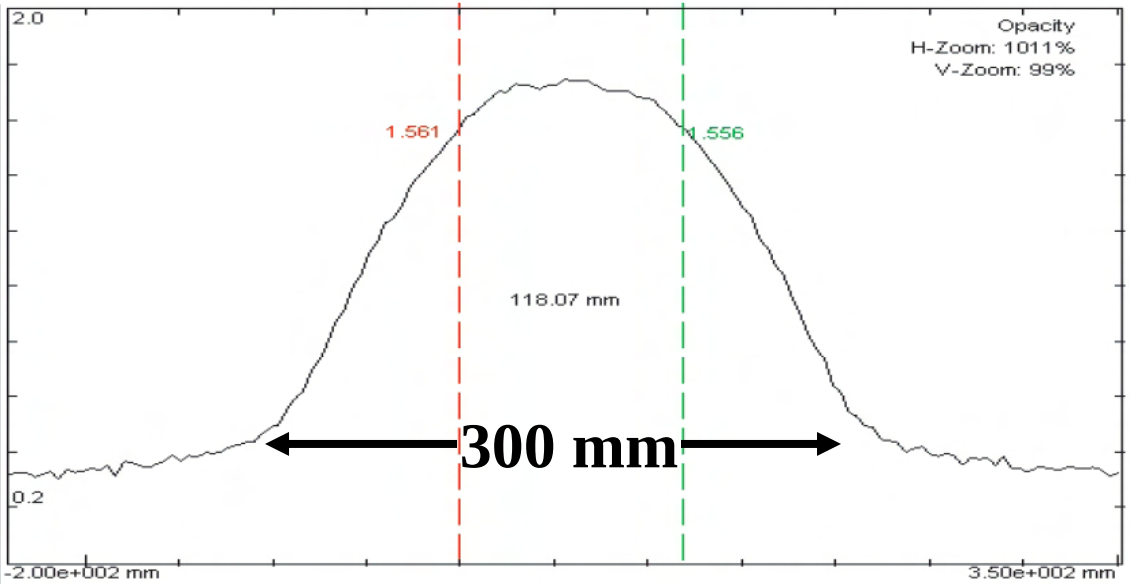




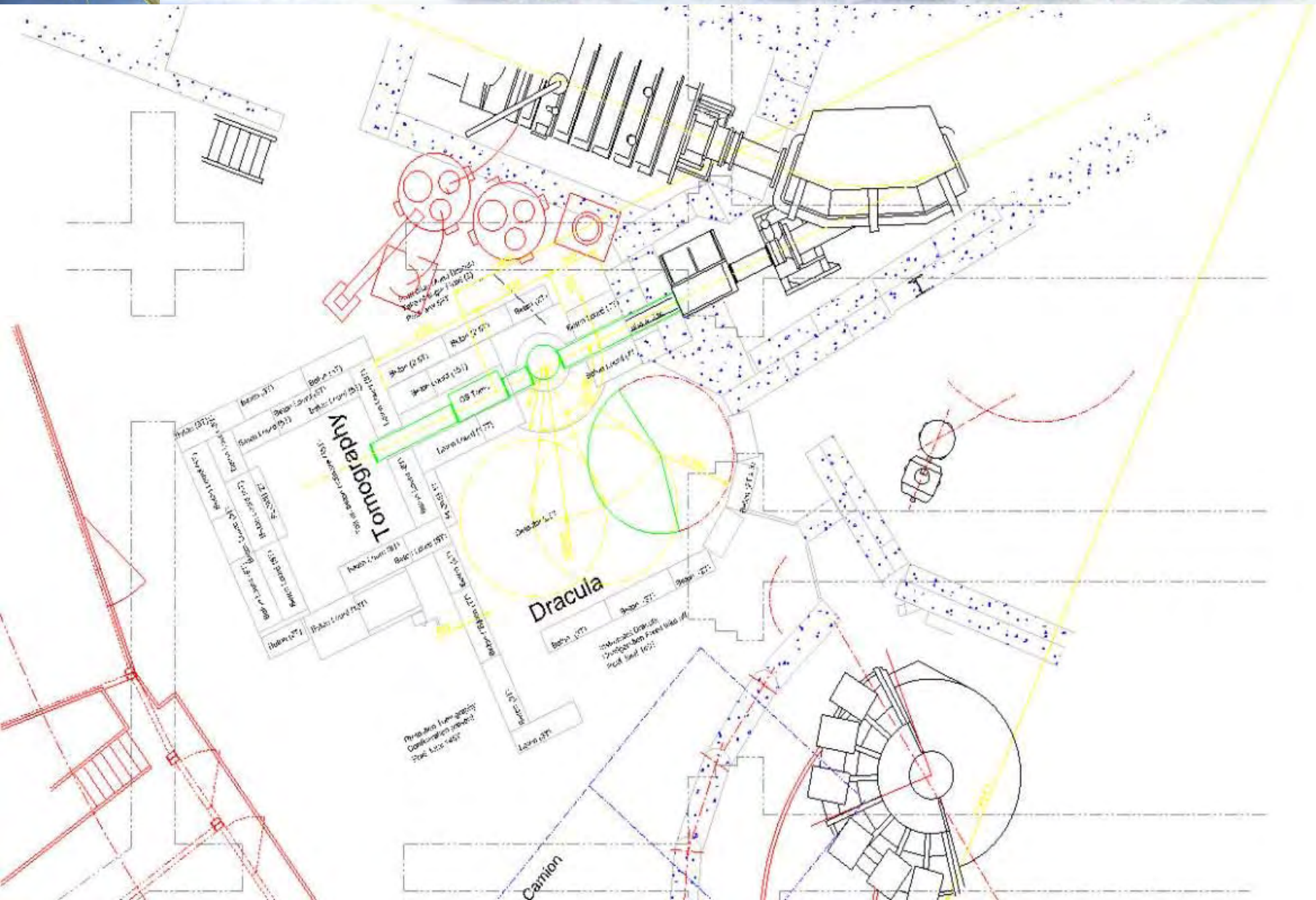
Aufbau einer dynamischen Radiografie- und Tomografiestation mit thermischen Neutronen

T. Ferger (2003)

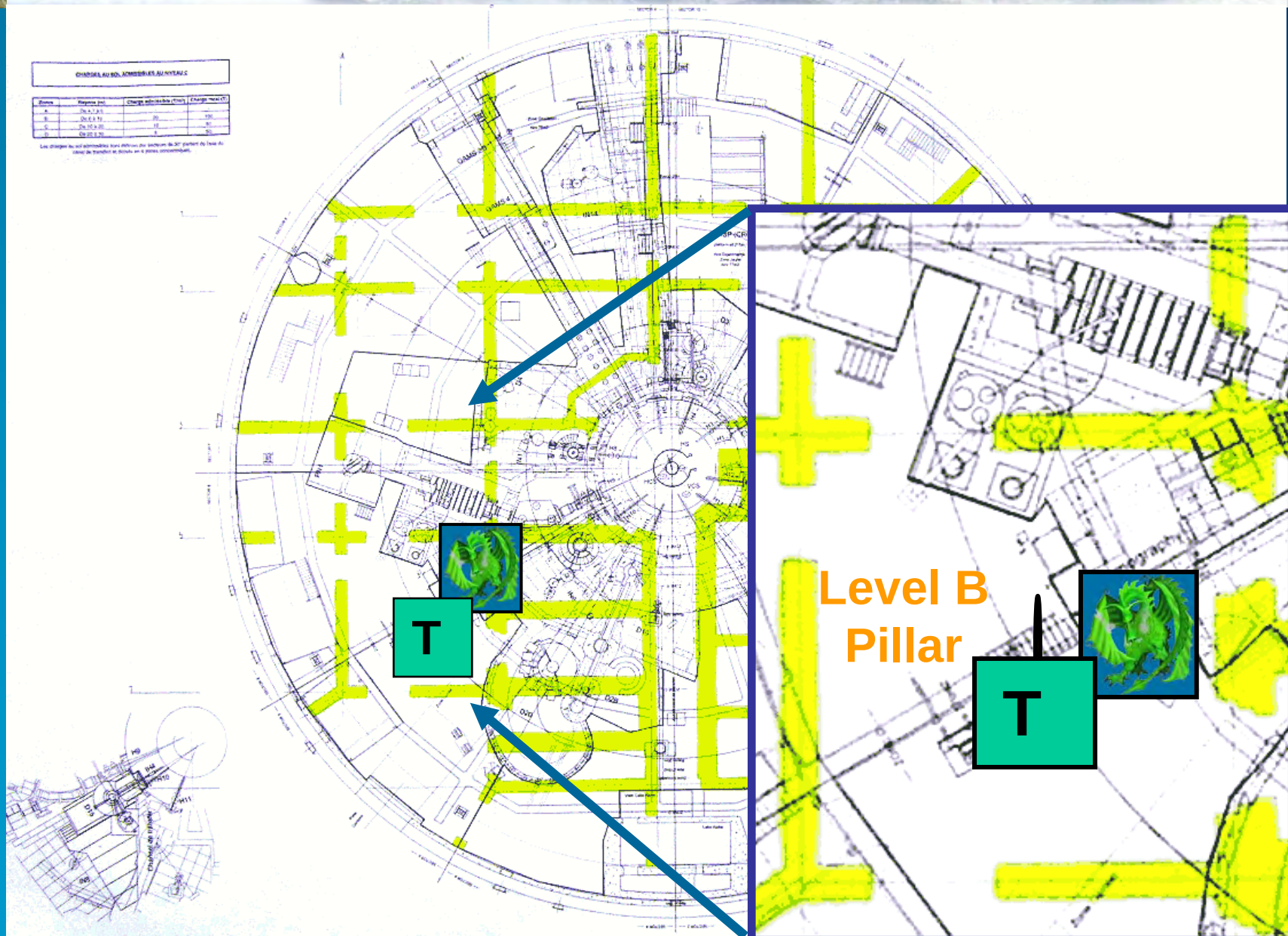




DRACULA+TOMOGRAPH on Beam Tube H9
Level C Foor Plan



DRACULA+TOMOGRAPH on Beam Tube H9
Level B Support



DRACULA+TOMOGRAPH on Beam Tube H9

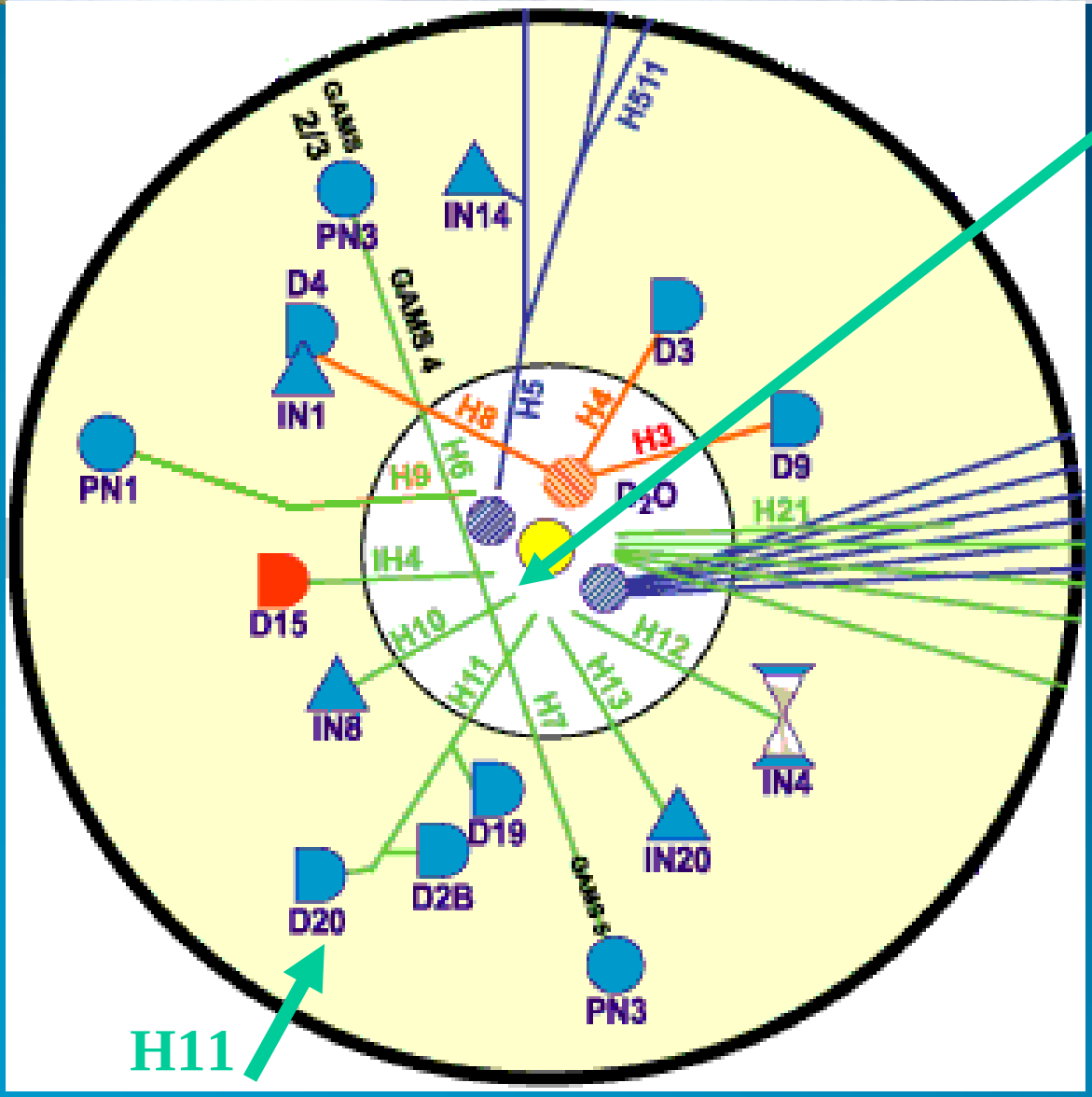
DRACULA on H9 (co-existing with Tomography station)

- Tomography would be moved back ~4m
- Tomography could be supported using a pillar in level-B
- A detailed floor load calculation has been commissioned
- Tomography would benefit by having better resolution
- Tomography would benefit from a better, larger casemate
- Dracula monochromator would absorb ~15% of white beam

Convert D20 to DRACULA ?
Need for a High Flux Thermal Beam Tube

- D20 has only recently been finished & is now working well
- D20 is the ILL's most requested machine (57 proposals)
- Only 2 modern powder machines for 22% of ILL proposals

Publications on ILL Thermal Beam-Tubes from ILL WWW pages



Most ILL Beam Tubes are Thermal

Number of Papers (1999-2004)

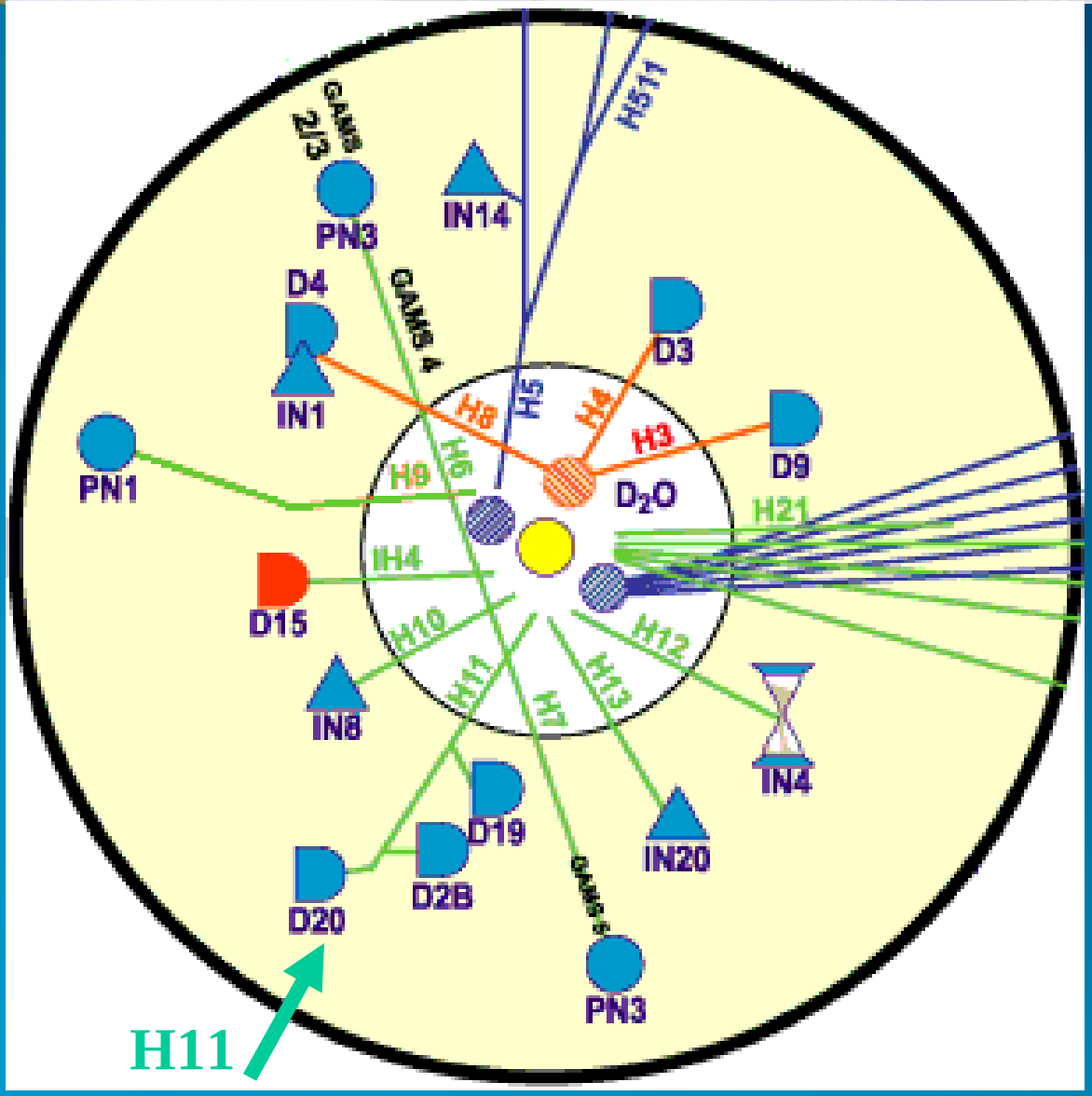
- H6+7 (GAMS) 12
- H9 (PN1) 14
- H10 (IN8) 61
- H12 (IN4) 22
- H13 (IN20) 69

Total 178

- H11 (D's) 458

Source: ILL library

Publications on ILL Thermal Beam-Tubes from ILL WWW pages



PRL, Phys.Rev., JACS, Nature, Science

Number of Papers (1999-2004)

- H6+7 (GAMS) 5
- H9 (PN1) 4
- H10 (IN8) 20
- H12 (IN4) 2
- H13 (IN20) 14

Total 45

- H11 (D's) 115

Source: ILL library

Highly cited ILL neutron diffraction papers

http://www.ill.fr/dif/citations/

Large number of citations for ILL neutron powder work

- **922 (D2B)** Hwang HY, Cheong SW, Radaelli PG, Marezio M, Batlogg B (1995) **Phy.Rev.Lett. 75**, 914.
  Lattice effects on the magnetoresistance in doped LaMnO3.
- **856 (D2B)** Cava RJ, Hewat AW, Hewat EA, Batlogg B, Marezio M, Rabe KM, Krajewski JJ, Peck WF, Rupp LW (1990) **Physica C. 165**, 419.
  Structural anomalies oxygen ordering and superconductivity in oxygen deficient Ba2YCu3Ox.
- **501 (D1A)** Capponi JJ, Tournier R, Chaillout C, Hewat AW, Lejay P, Marezio M, Nguyen N, Raveau B, Soubeyroux JL, Tholence JL (1987) **Europhysics Letters. 3,** 1301.
  Structure of the 100K superconductor Ba2YCu3O7 between 5-300K by neutron powder diffraction.
- **435 (IN8)** Rossat-Mignod, J. M., L. P. Regnault, et al. (1991) **Physica C 185-189**: 86-92.
  Neutron scattering study of the YBa2Cu3O6+x system.
- **367 (D16)** Deteresa JM, Ibarra MR, Algarabel PA, Ritter C, Marquina C, Blasco J, Garcia J, Delmoral A, Arnold Z (1997) **Nature 386,** 256-259
  Evidence for magnetic polarons in the Magnetoresistive materials
- **337 (D1A)** Fitch, A. N., H. Jobic, et al. (1986). **Journal of Physical Chemistry 90**, 1311-1318
  Localization of benzene in sodium-Y, zeolite by powder neutron diffraction.
- **335 (IN6)** Buchenau, U., M. Prager, et al. (1986). **Physical Review B 34**, 5665-5673.
  Low-Frequency modes in vitreous silica.
- **332 (IN13)** Doster, W., S. Cusack, et al. (1989) **Nature 337**: 754-756.
  Dynamical transition of myoglobin revealed by inelastic neutron scattering.
- **321 (D2B)** Radaelli PG, Cox DE, Marezio M, Cheong SW, Schiffer PE, Ramirez AP (1995) **Phys.Rev.Lett. 75,** 4488
  Simultaneous structural, magnetic, and electronic-transitions in La(1-x)Ca(x)MnO3 with x=0.25 and 0,5
- **319 (D2B)** Radaelli PG, Cox DE, Marezio M, Cheong, SW (1997) **Phys.Rev. B55,** 3015
  Charge, orbital, and magnetic ordering in La(0.5)Ca(0.5)MnO3

Shoot the Dragon…

If we want ESS we have to…

- **Show we are making best use of what we already have**

Shoot the Dragon…

No Pretty Pictures

Pretty Pictures: "C'est magnifique, mais ce n'est pas la guerre"

General Bosquet, watching the British Light Brigade charge Russian guns

If we want ESS we have instead to…

- **Satisfy our users and earn their support**
  ie Numbers of groups, proposals, publications, citations

DRACULA – Small Samples, Low Background

Can we obtain all d-spacings with a 2Θ range of 60°-120° ?
(i.e. with a very small scattering volume)

Use a large focusing Ge monochromator near 90° to obtain several λ

[115] -> 1.54Å;		d= 0.889Å - 1.54Å
[113] -> 2.44Å;	(graphite filter)	d= 1.39Å - 2.44Å
[111] -> 4.61Å;	(beryllium filter)	d= 2.66Å - 4.61Å

Powder Diffraction on Pulsed Sources

A High Resolution SNS Powder Diffractometer

B.E.F.Fender & A.W.Hewat (1978)

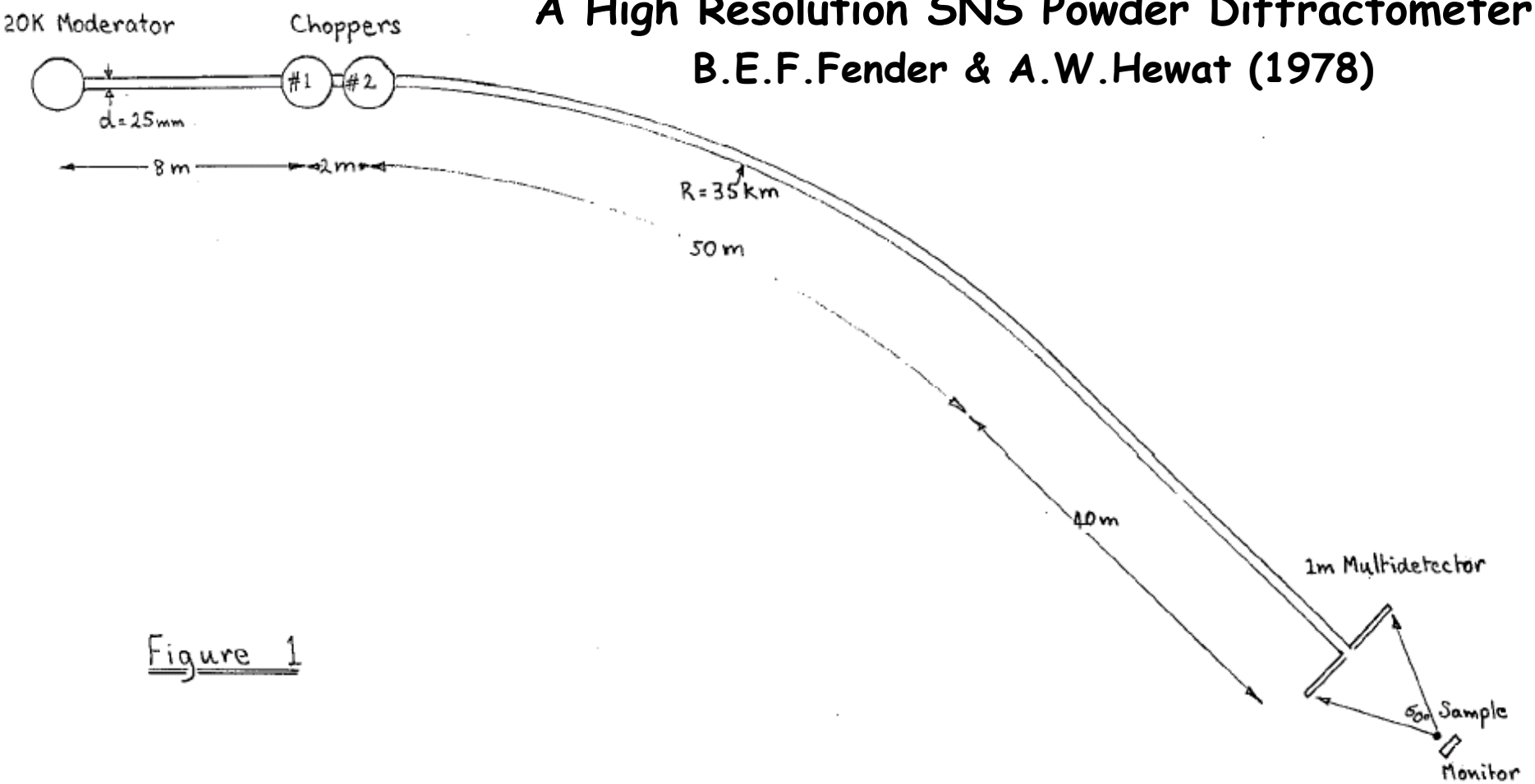


Figure 1

Powder Diffraction on Pulsed Sources

Advantages over Steichele's Munich machine

i) Higher resolution, because with a 20 K moderator the pulse width is as low as 7λ μsec for $\lambda < 2.2$ Å, giving $\frac{\Delta t}{t} = 2.8 \times 10^{-4}$ at $\ell = 100$ metres. (Steichele has 100 μsec at 143 metres giving $\frac{\Delta t}{t} \sim \frac{3\times10^{-3}}{\lambda}$).

ii) Constant resolution for $\lambda < 2.2$ Å, since the neutron velocity is $\frac{4000}{\lambda}$ m.sec^{-1} (Steichele has $\frac{\Delta d}{d}$ increasing with $\frac{\sin\theta}{\lambda}$, whereas one would like it to decrease as on a conventional HRPD).

iii) A larger $\Delta\lambda$(0.8 Å) can be used without frame overlap, because of the shorter flight path. (Steichele must be limited to $\sim$0.5 Å). This means that sufficient data for most crystal structure problems could be collected with a single $\Delta\lambda$ slice.

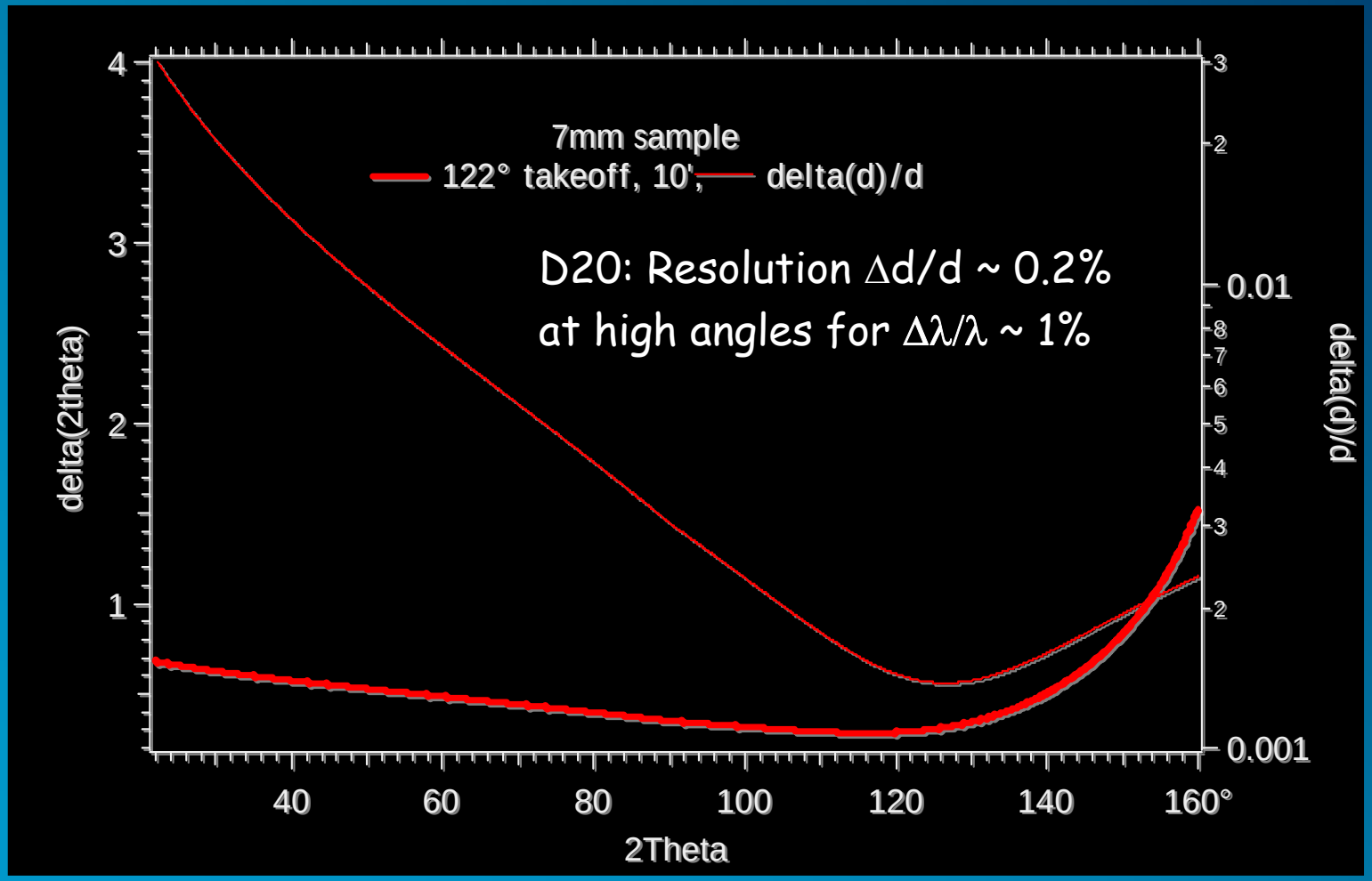
iv) Of course, much higher intensity (a factor of $\sim 10^3$), eliminating the need for the very large samples ($\sim$100 cm^3) used at Munich, and making possible experiments on quite small (< 1 cm^3) hydrogenous samples of complex structure, dilute precipates in alloys etc.

Disadvantage

(i) The pulse shape from the SNS moderator has a rapid increase, corresponding to the time for thermalization, followed by an exponential decay as the moderator empties. This is less favourable than the clean triangular pulse

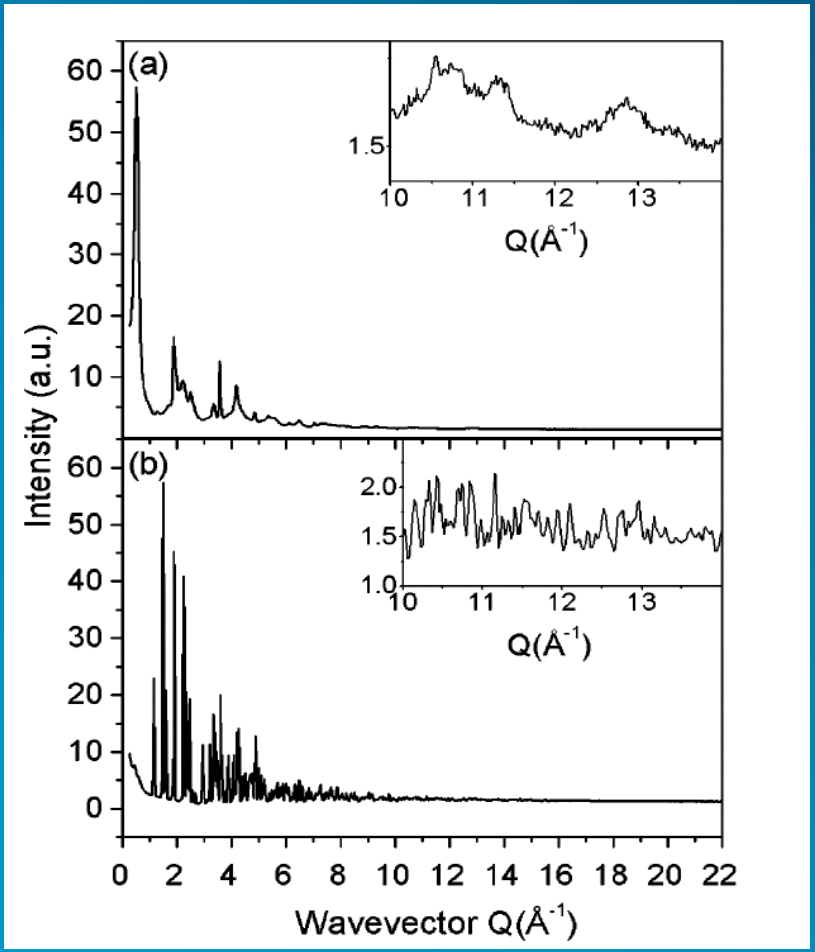
Does large $\Delta\lambda/\lambda$ mean low resolution ?

A: No. Resolution is INDEPENDENT of $\Delta\lambda/\lambda$ at focusing angle.

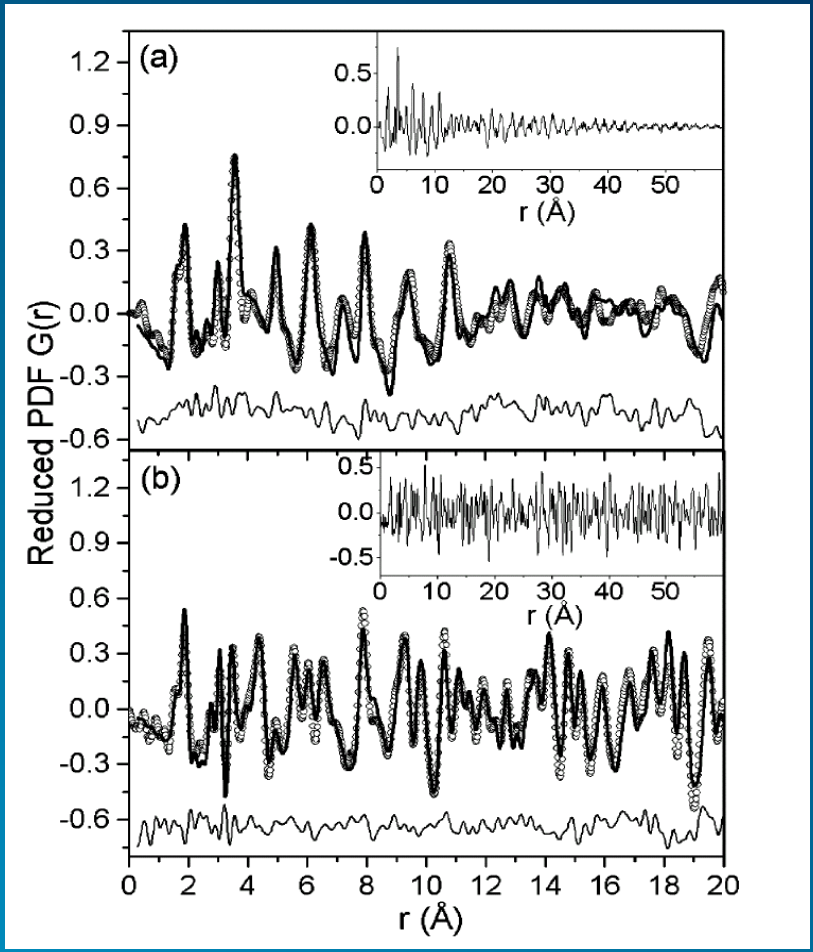


PDF and "Beyond Crystallography" Partially Ordered Materials - Super-D9 ?

1) Diffraction a) aerogel b) crystalline

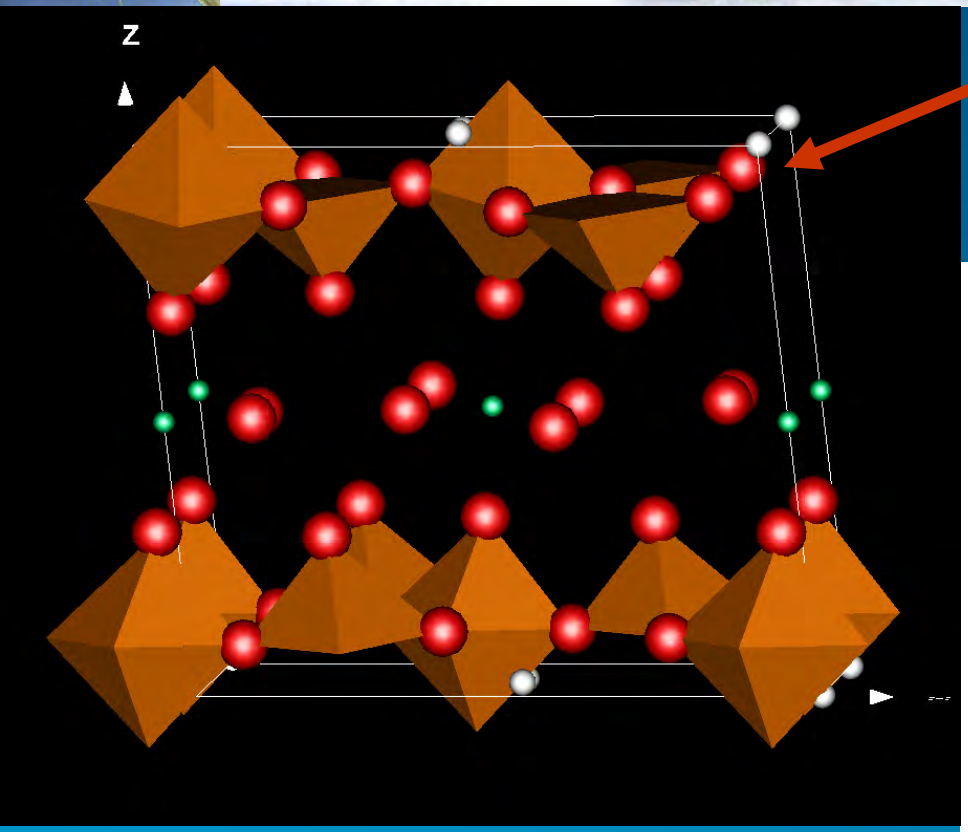


2) PDF a) aerogel b) crystalline



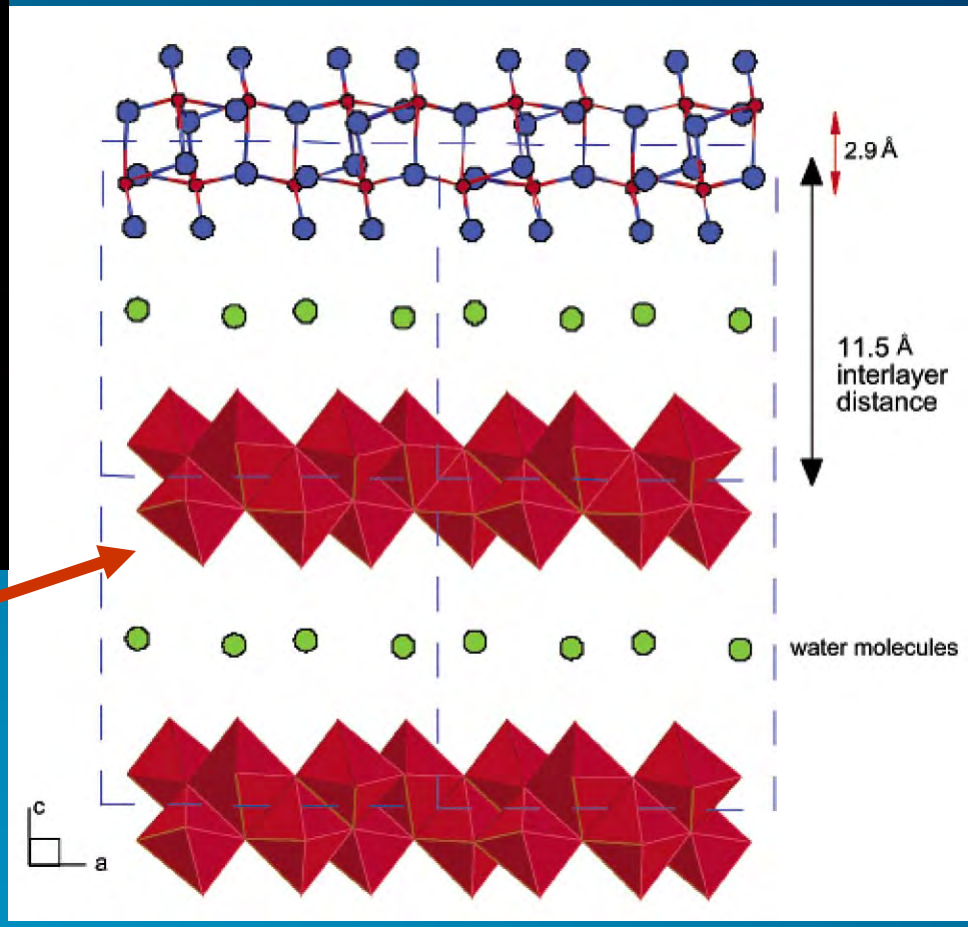
BOTH PDF functions have strong peaks to large r

PDF and "Beyond Crystallography" Partially Ordered Materials - Super-D9 ?



Hydrated Vanadium Oxide Crystal

Y.Oka, T.Yao and N.Yamamoto (1997)



$V_2O_5.nH_2O$ Xerogel

V.Petkov, P.Trikalitis, E.Bozin, S.Billinge, T.Vogt, and M.Kanatzidis (2002) JACS


Their's not to make reply,
Their's not to reason why,
Their's but to do and die:
Into the valley of Death
Rode the six hundred.
"C'est magnifique, mais ce n'est pas la guerre" General Bosquet

DRACULA cost and feasibility

- Dracula would use tested D19-type 2D-detector
- Dracula would also be useful for non-powder diffraction
- Dracula casemate optics would be similar to that of D20
- Dracula mono. would be less high, but horizontally focussing
- Cost and time-scale for Dracula can be easily calculated
- Dracula is a "no-risk" project that will give ILL a big lead

DRACULA – Strategy or Submission ?

DRACULA – Strategy or Submission ?

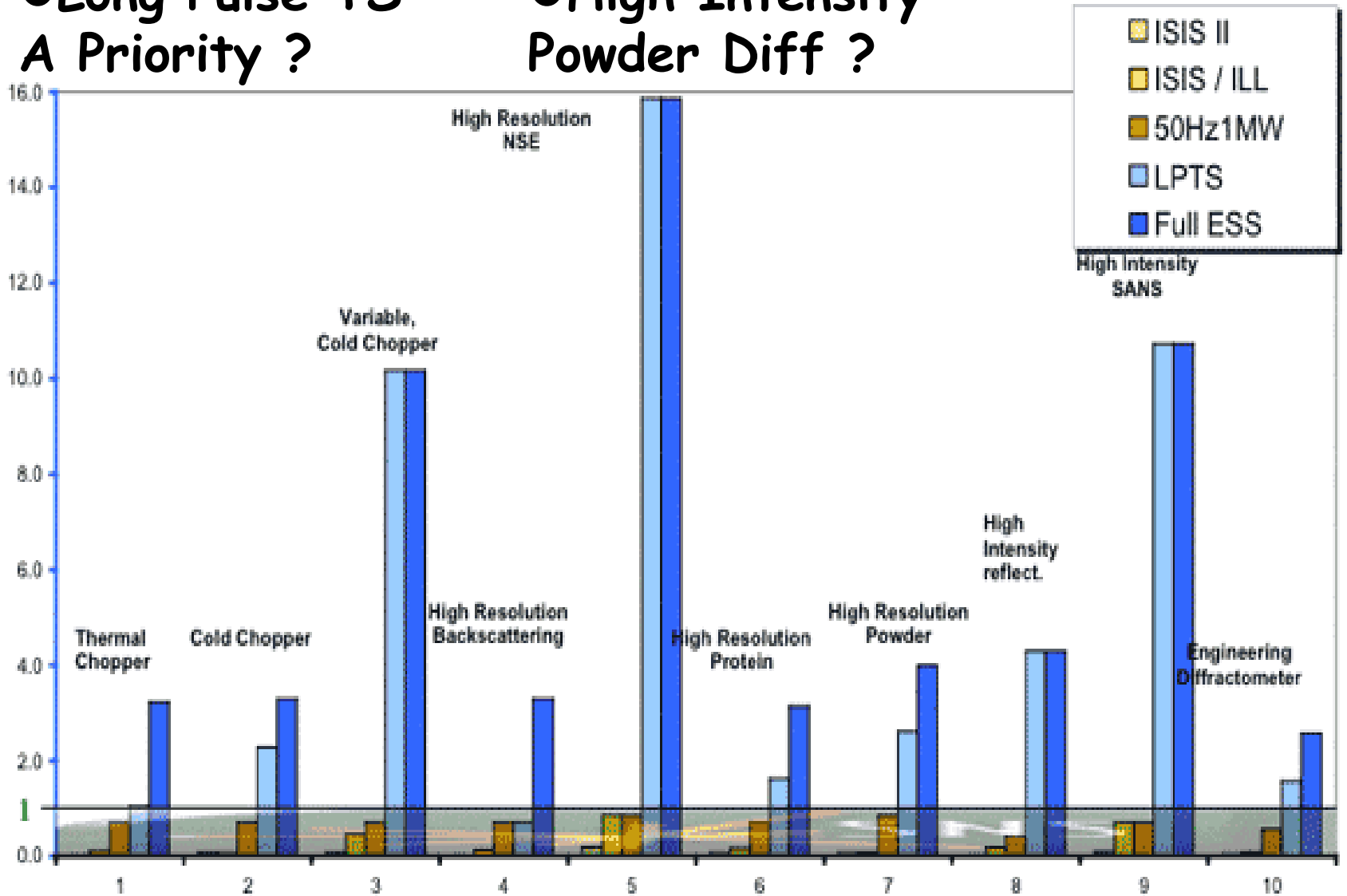
Can we compete with the Americans while waiting for ESS?

(Free Advice to ILL Directors)

- Use our natural advantage – time average flux on sample
- Use big detectors, as on pulsed neutron sources
- Do not assume that the SNS will be a long time coming
- Do not wait until the SNS is operational before reacting

While we are waiting for ES…

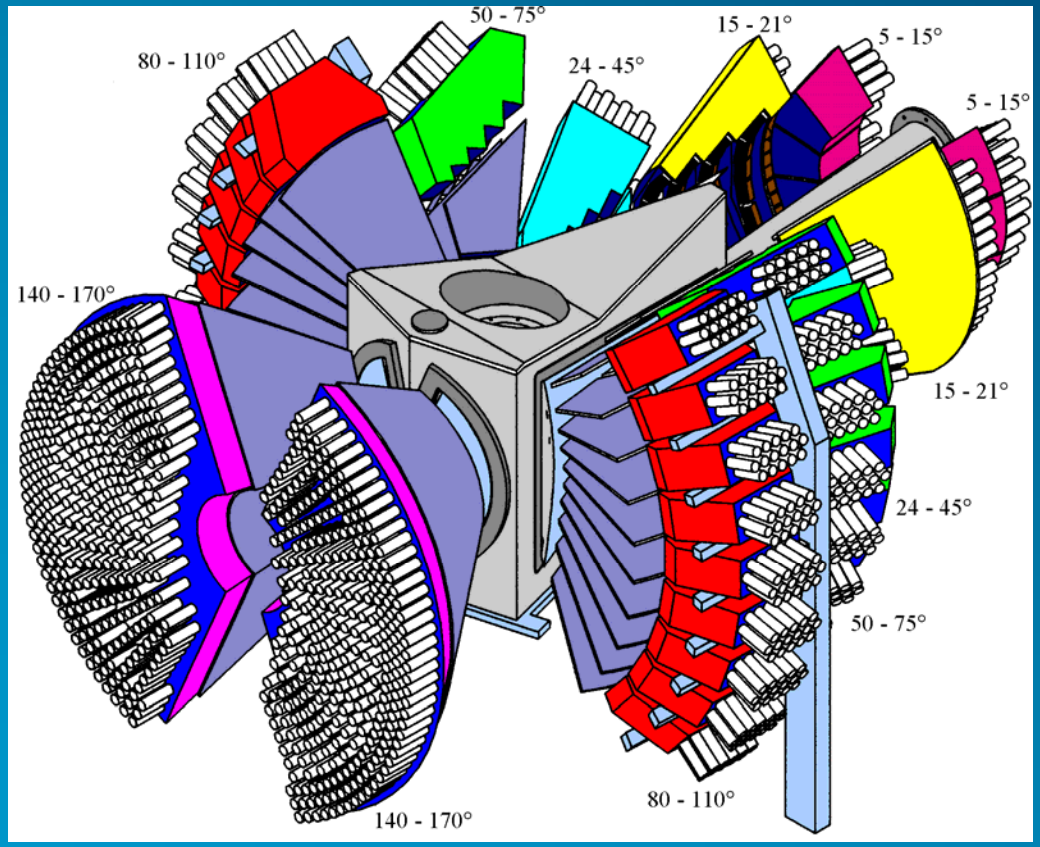
- Long Pulse TS A Priority ?
- High Intensity Powder Diff ?



SNS

Partially ordered Materials (super-D9)
Low resolution with short wavelengths

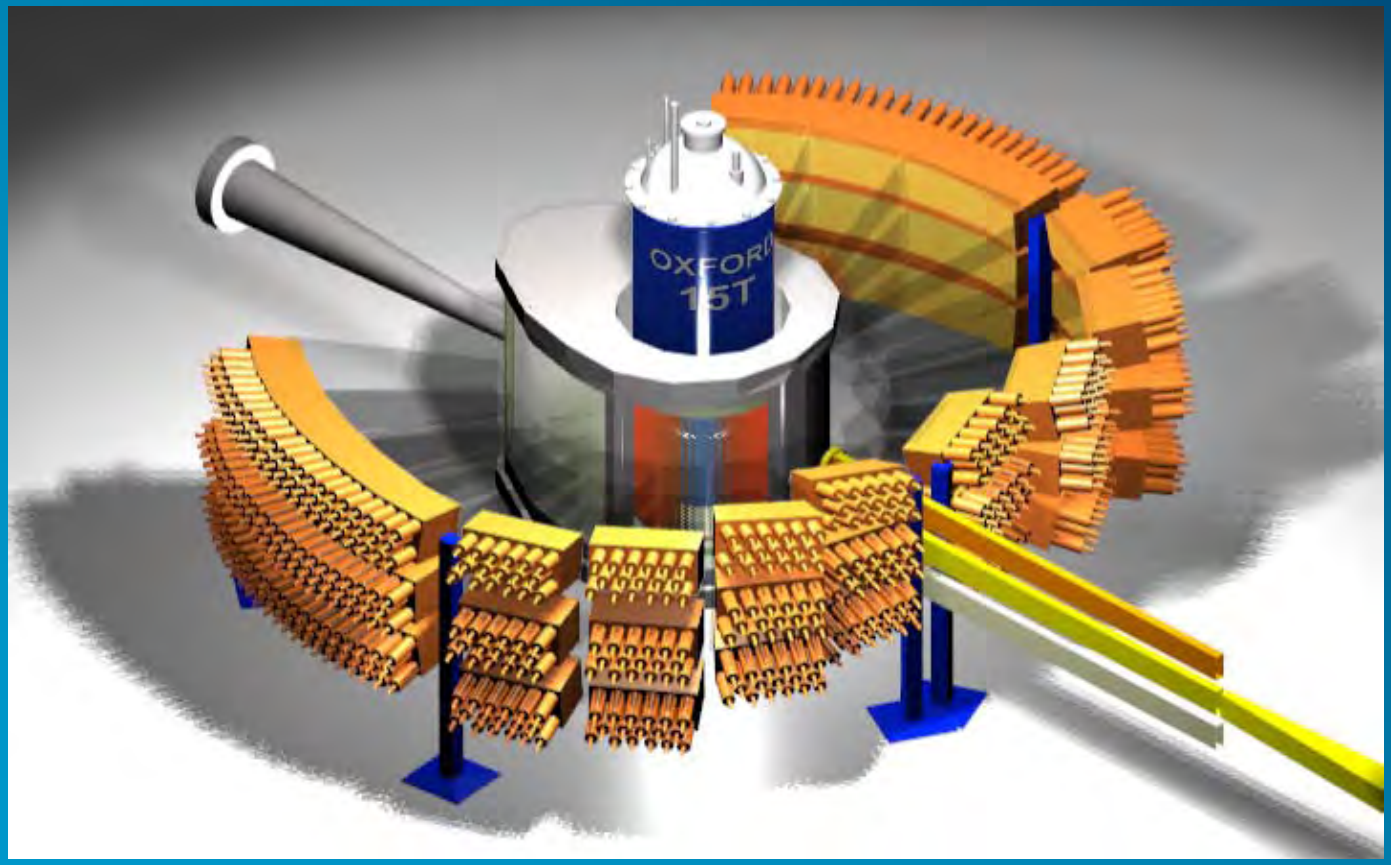
- ILL lacks a partially ordered materials PDF machine like ISIS-GEM
- GEM can use shorter wavelengths than D20, & D4 is only 50% available
- Replace current small "bidim" on D9 by large Dracula-type banana ?



GEM at ISIS

Magnetism and Biology Dracula (super-D16)
High resolution with long wavelengths

- ILL lacks a wide-angle, cold neutron machine like ISIS-WISH
- WISH is designed for long-period structures – magnetism & biology
- Replace current small "bidim" on D16 by large Dracula-type banana ?



WISH at ISIS

Powder Diffractometer Efficiency

ESS Instrumentation Group Reports, May 2001

Powder Diffraction Instruments

P.G. Radaelli, S. Hull , H.J. Bleif & A. M. Balagurov

The average count rate is the product of:

- the flux at the sample
- the detector solid angle and efficiency
- the sample volume

Powder Diffractometer Efficiency

ESS Instrumentation Group Reports, May 2001

Powder Diffraction Instruments

P.G. Radaelli, S. Hull , H.J. Bleif & A. M. Balagurov

- Reactor-based instruments tend to maximise the source solid angle, by exploiting focusing monochromators (flux)
- Diffractometers at pulsed sources tend to have much larger detector solid angles.

Powder Diffractometer Efficiency

ESS Instrumentation Group Reports, May 2001

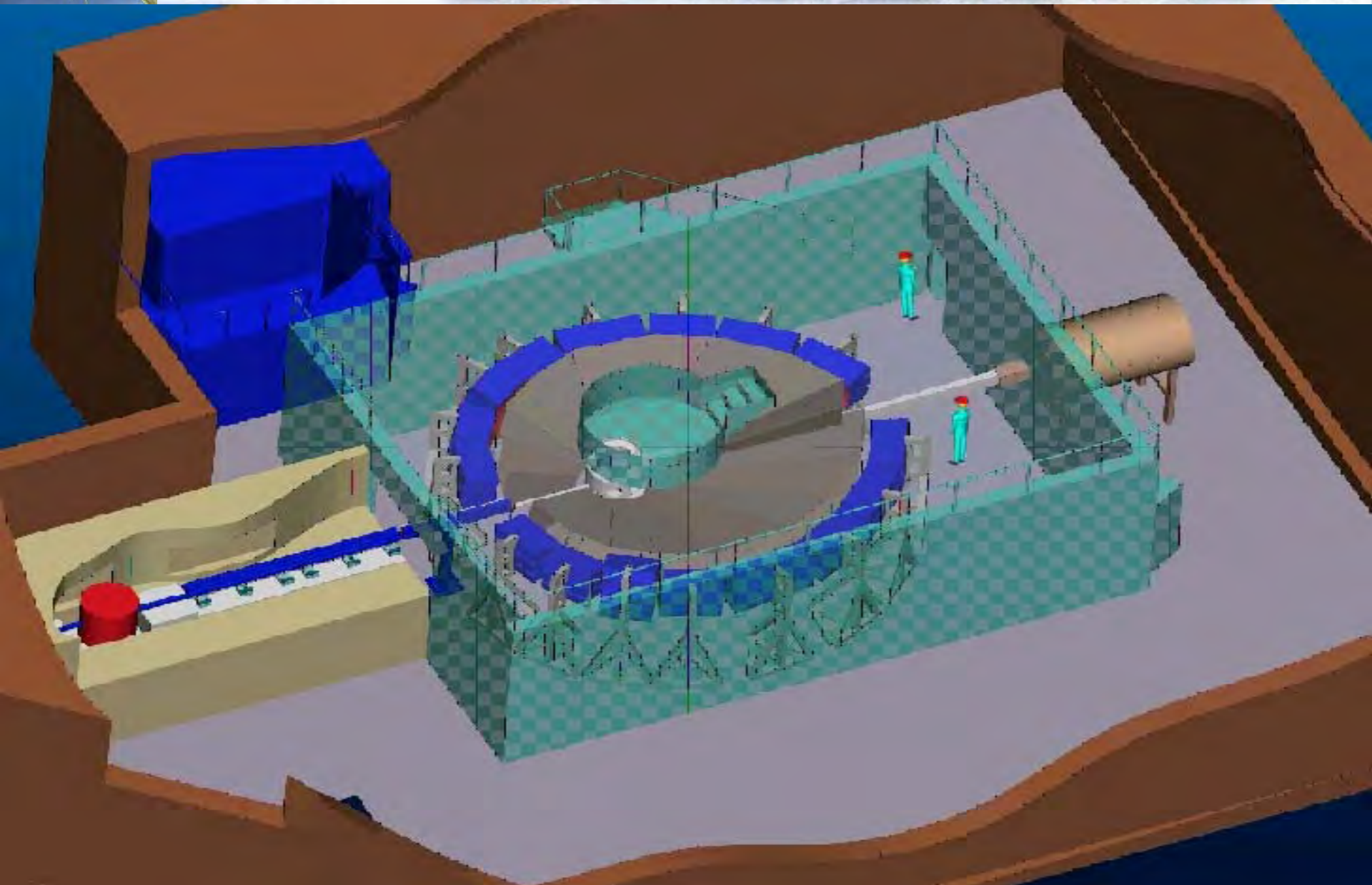
Powder Diffraction Instruments

P.G. Radaelli, S. Hull , H.J. Bleif & A. M. Balagurov

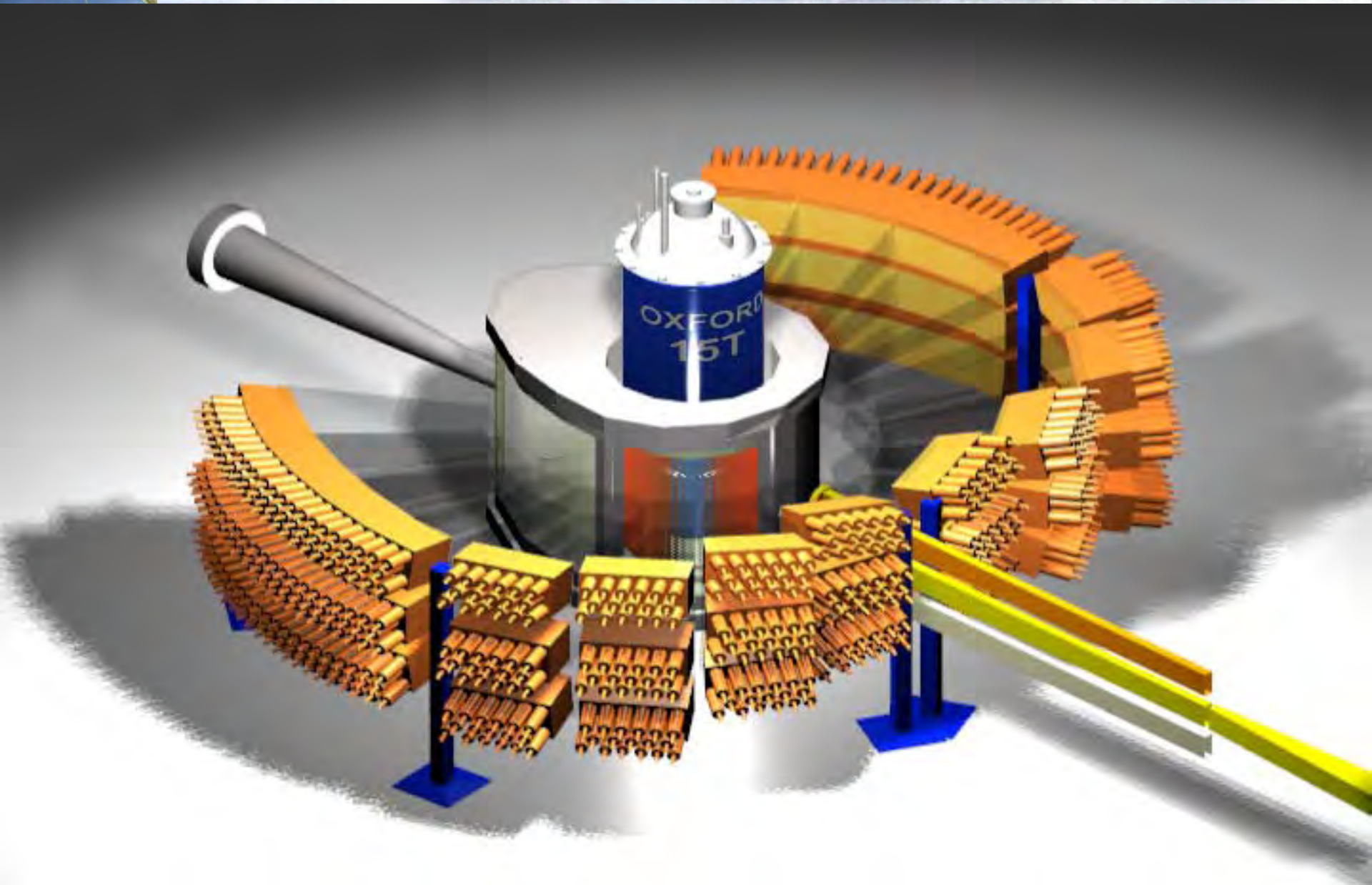
Conclusions:

- TOF machines need a higher flux source (SNS,ESS)
- CW machines need a larger detector

POWGEN3 at SNS – a BIG detector



WISH at TS2-ISIS – a BIG detector



While we are waiting for the ESS dream…

Proposed ESS Powder Diffractometers

- ST05 High-Q Powder Diffractometer HQP
- ST06 Liquids & Amorphous Diffractometer LAD
- SM10 Single Pulse Diffractometer SPD
- SD17 Magnetic Powder Diffractometer MagP
- SD18 High Resolution Powder Diffractometer HRPD
- LM05 Ultra-high Resolution Powder Diffractometer URPD
- LM06 High Pressure Powder Diffractometer HiPD

Comparison of TOF & CW Diffractometers

The time-averaged **Flux*Detector** criterium

D20 has high flux, GEM has a big detector

	D20	GEM
time averaged sample flux	5×10^7	$\sim 2 \times 10^6$
detector solid angle	0.27 sr	4.0 sr
efficiency	1.7	1

Comparison of TOF & CW Diffractometers

The time-averaged **Flux*Detector** criterium

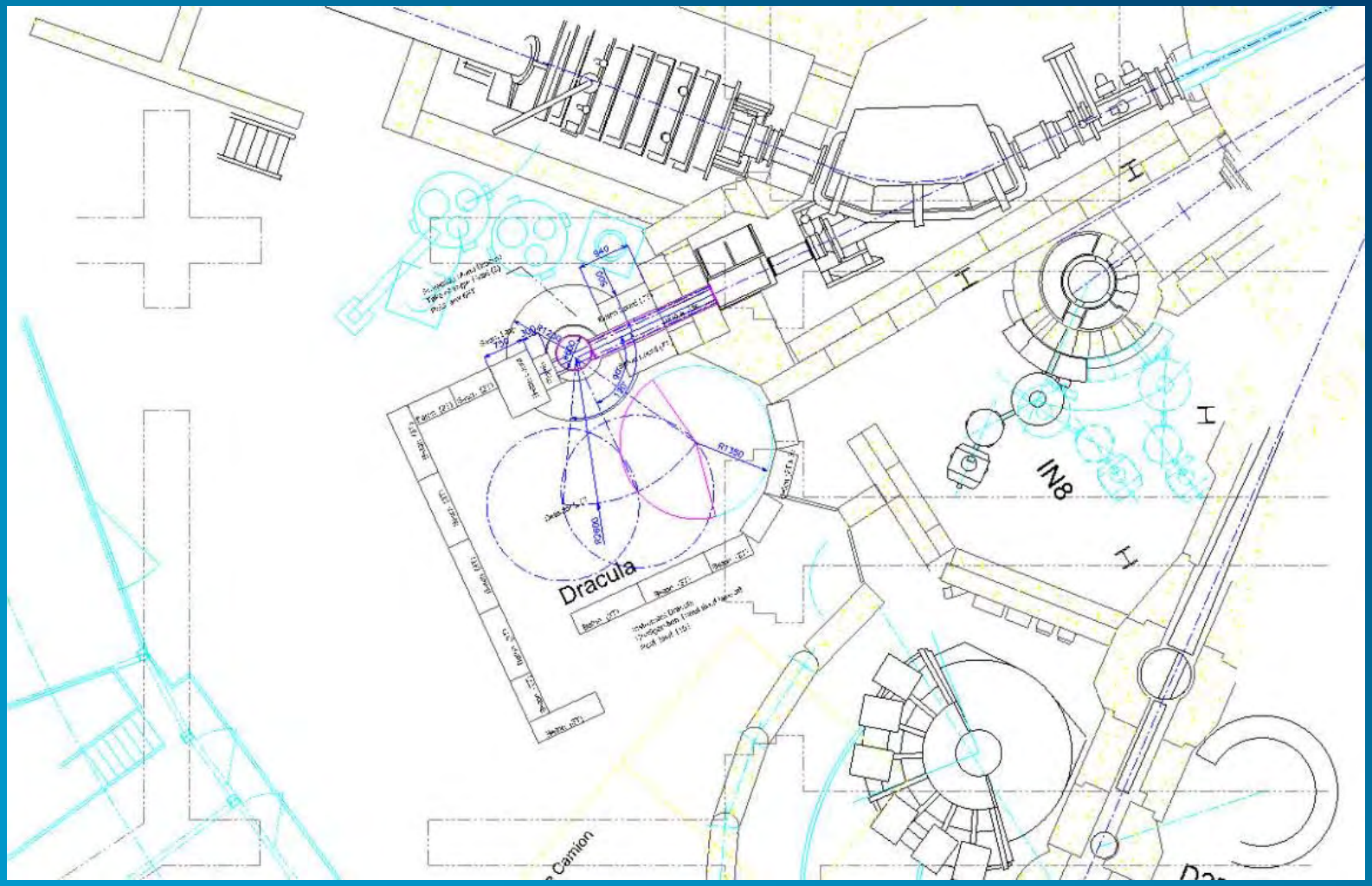
So, let's use a big detector too !

	D20	GEM	DRACULA
time averaged sample flux	5×10^7	$\sim 2 \times 10^6$	$\sim 10^8$
detector solid angle	0.27 sr	4.0 sr	1.5 sr*
efficiency	1.7	1	18

* Based on new D19 detector: R=760 mm, h=400 mm, 800 linear resistive wires $30^o \times 160^o$

Thermal DRACULA on a High Flux Beam

DRACULA on H9 (replacing the Tomography station)

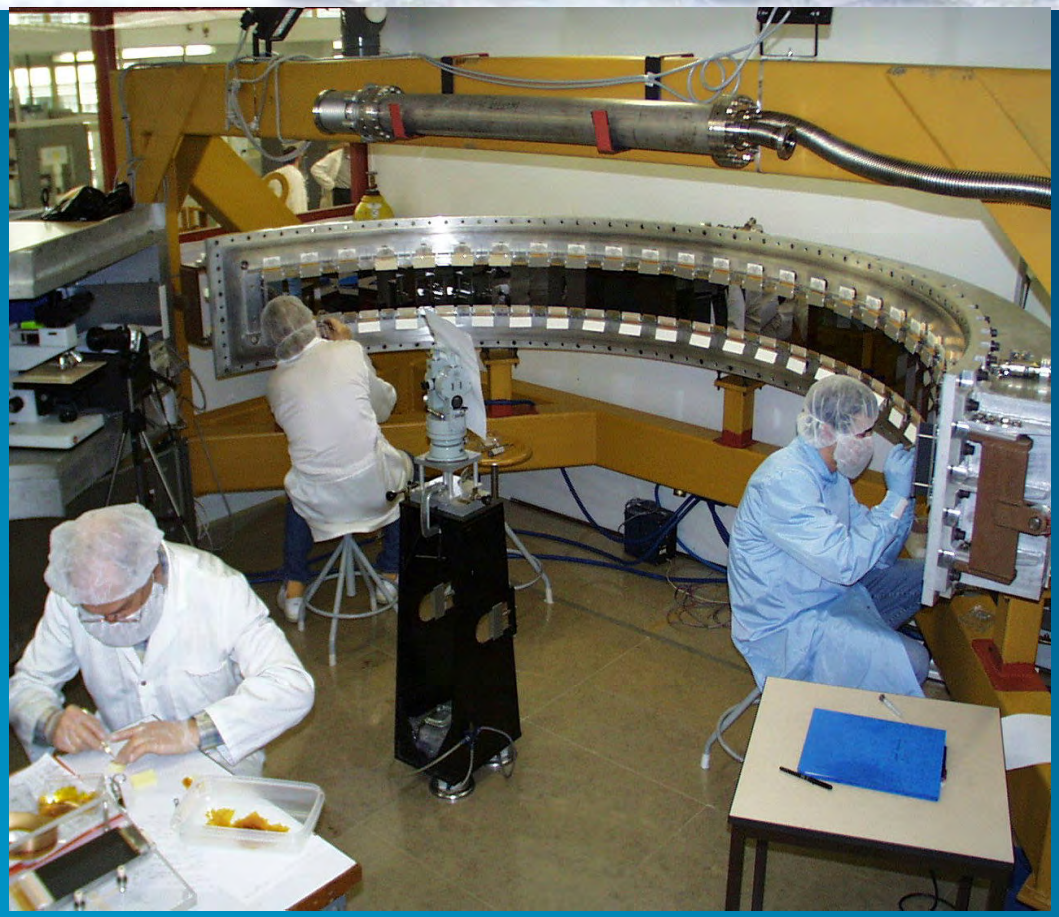


Thermal DRACULA on a High Flux Beam

DRACULA on H9 (replacing the Tomography station)

- Dracula would weigh about the same as Tomography
- Dracula slightly restricted by Lohengrin chariot/magnet
- But the big intensity gain comes from wavelength focussing
- For that, we need a wide monochromator, not so tall
- But it would be nice to find a place for tomography…

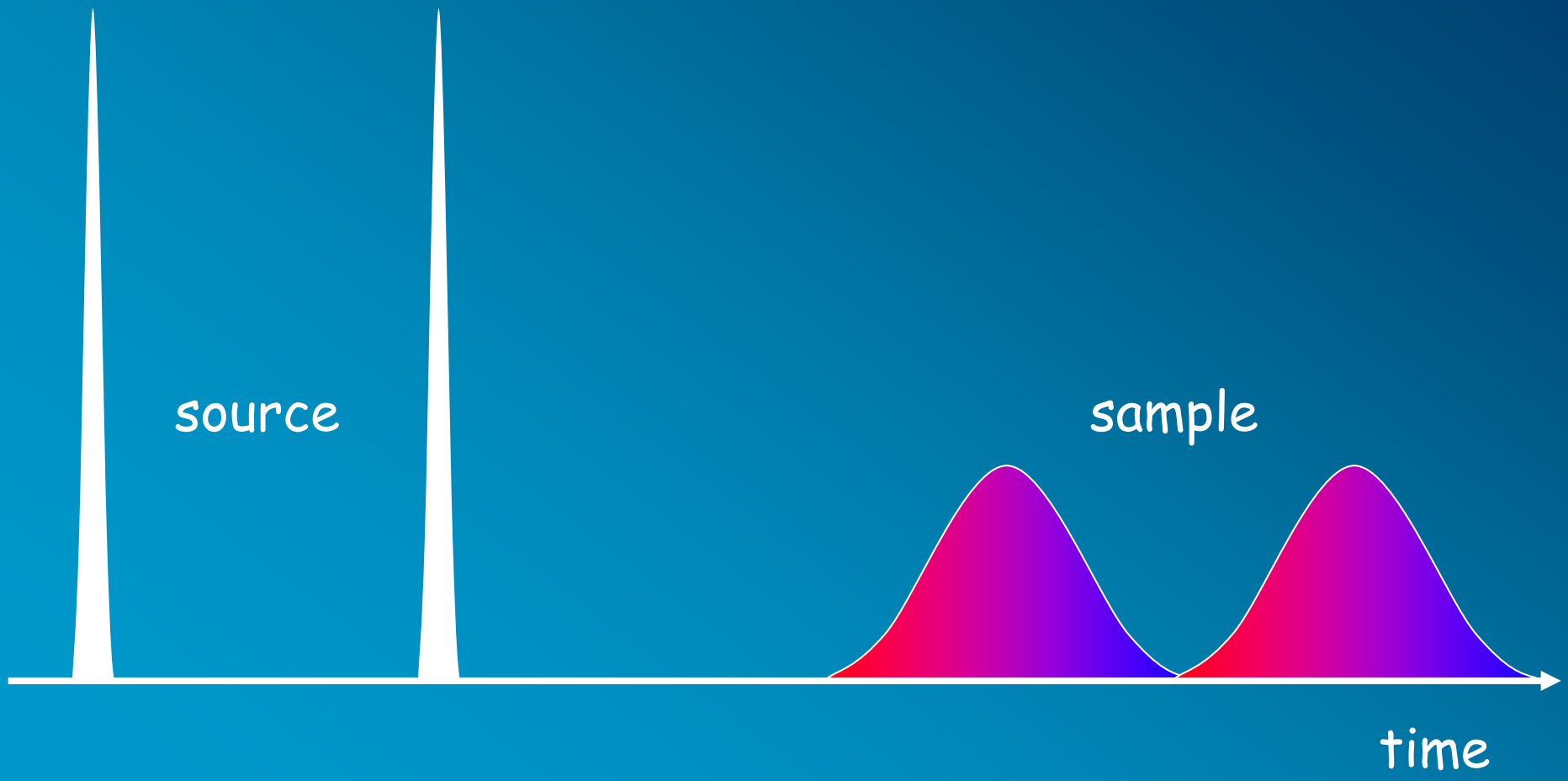
Convert D20 to DRACULA ?
Need for a High Flux Thermal Beam Tube



- D20 has only recently been finished & is now working well
- D20 is the ILL's most requested machine (57 proposals)
- Only 2 modern powder machines for 22% of all proposals

Why is sample flux low on a pulsed source ?

A: The sharp white peaks spread out in wavelength with time
TOF machines have rather constant (low) flux at sample



High Resolution SNS Powder Diffractometer

B.E.F.Fender & A.W.Hewat (1978)

In the standard high resolution geometry, a counter of diameter 1 m and 1 m from the sample will subtend a solid angle of 6.25 % of 4Π , almost an order of magnitude better than on a conventional diffractometer such as D1B.

ii) Low angle counters

The most important low angle counter will be the low efficiency monitor directly behind the sample. This counter is needed, not only to measure

iii) 90° counters

The main interest of detectors at 2θ=90° would be for avoiding scattering from the sample surroundings –air, cryostats, furnaces etc. The incident and diffracted beams, if orthogonal, may define a very small scattering volume. However, backscattering would still be superior in at least some situations ; for example, a pressure cell or cryostat might be made with only two small single crystal windows.

Why is sample flux so high from a reactor?

A: Large vertically focusing monochromators ?

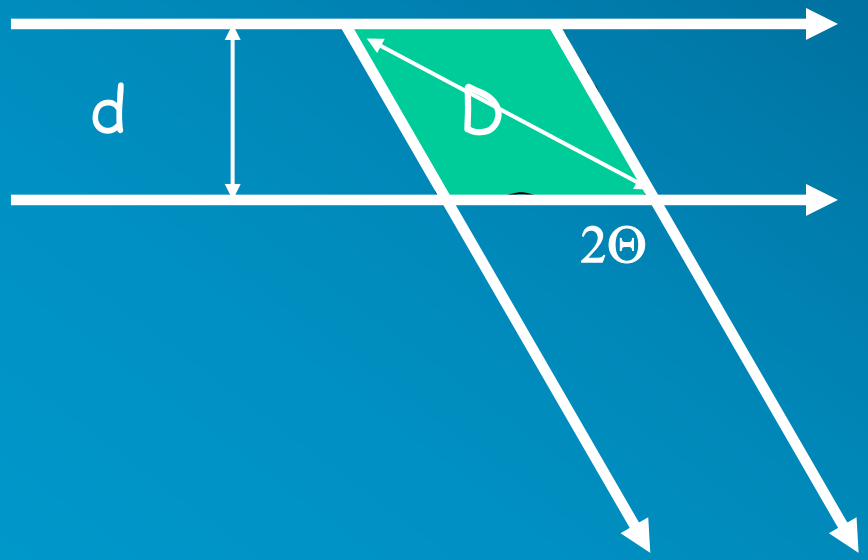
Slight smearing of diffraction cones due to monochromator vertical divergence (esp. at very low and very high angles).

Not a big problem for medium resolution

DRACULA – Small Samples, Low Background

Large detector & radial collimator near 90° scattering

± 15° vertical as for the new D19 detector cf ±7° for new Paris-Edinburgh cell
± 30° horizontal ie 2Θ = 60° - 120° (range of scattering angles for pressure cell)

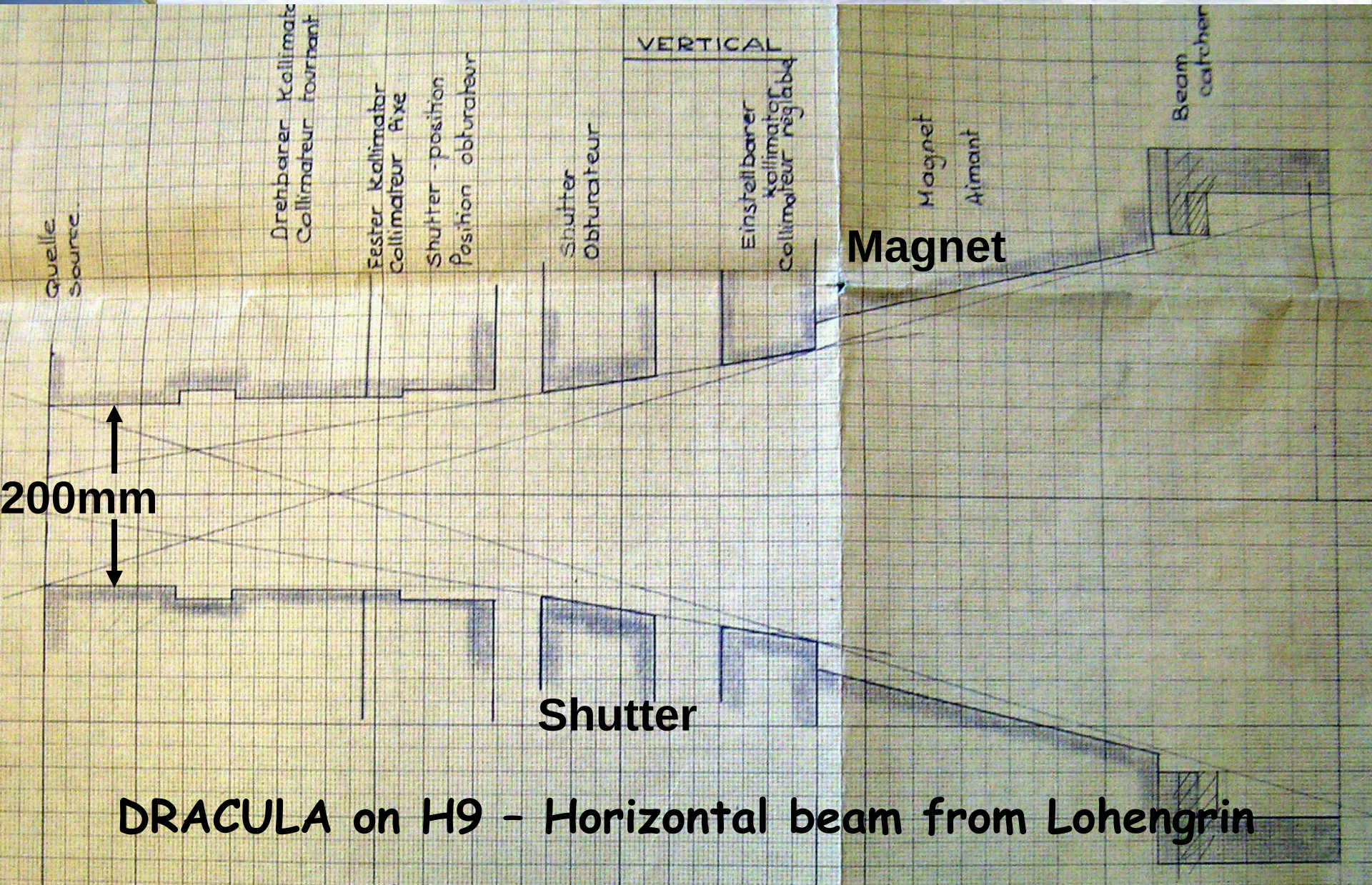


d = diameter of the incident beam
D = diameter of scattering volume
= d/sinΘ
= d√2 (minimum at 2Θ = 90°)
= 2d (maximum at 2Θ = 60° & 120°)

D = 5mm – 7mm for 2Θ = 60° - 120°

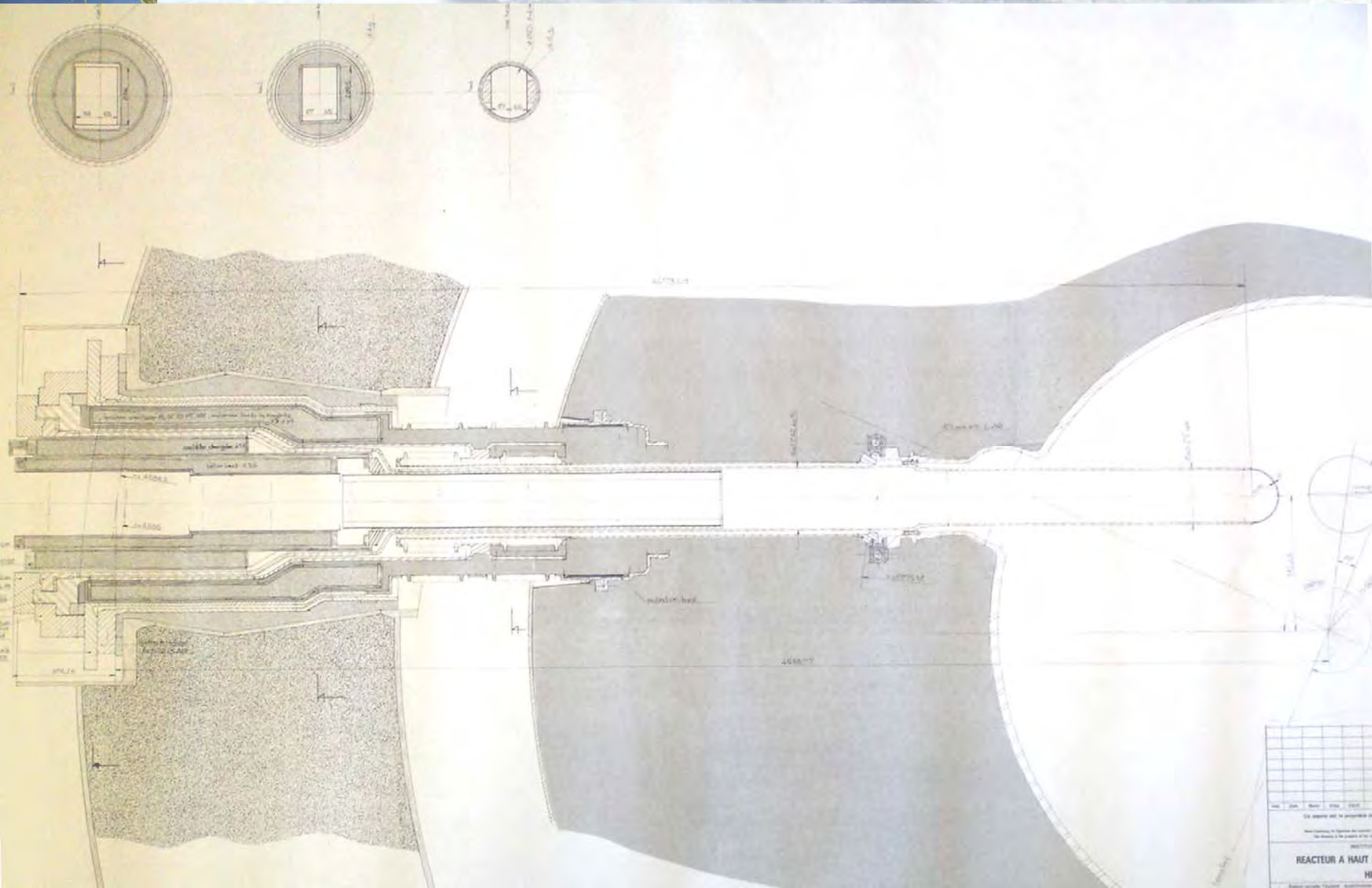
Scattering limited to a very small sample volume

DRACULA on High Flux Beam Tube H9



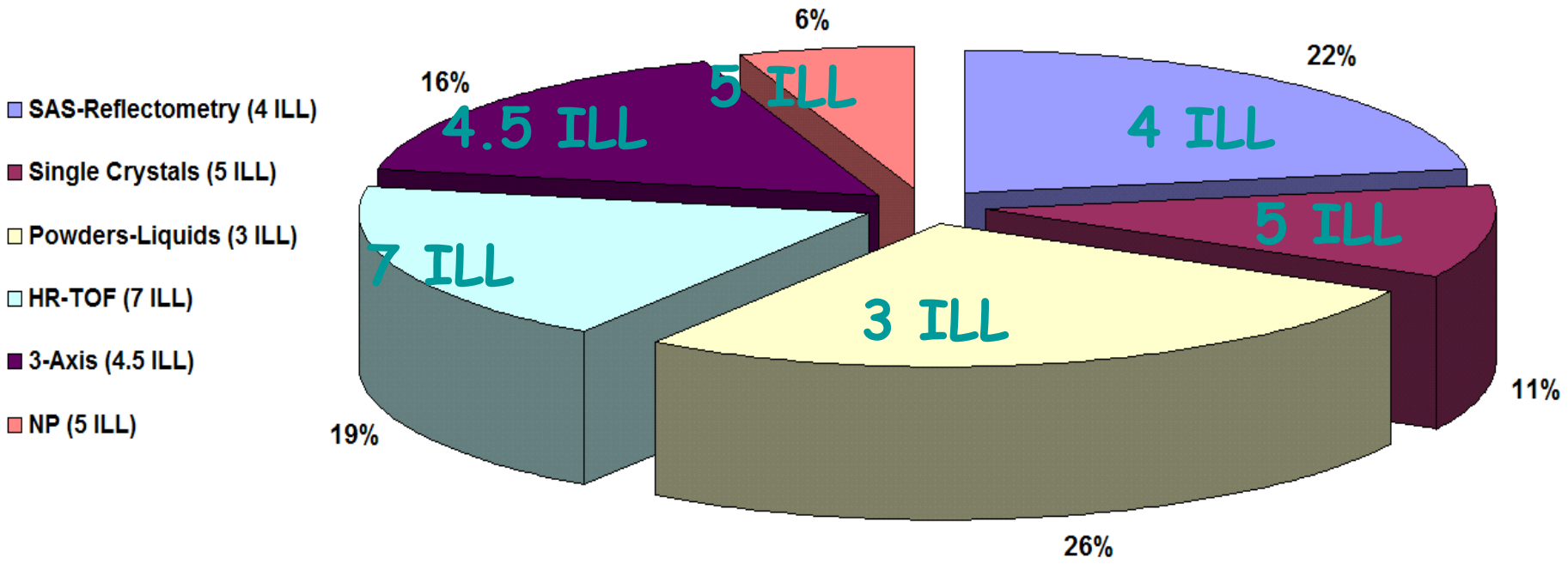
DRACULA on H9 – Horizontal beam from Lohengrin

DRACULA on High Flux Beam Tube H9



Convert D20 to DRACULA ?
Need for a High Flux Thermal Beam Tube

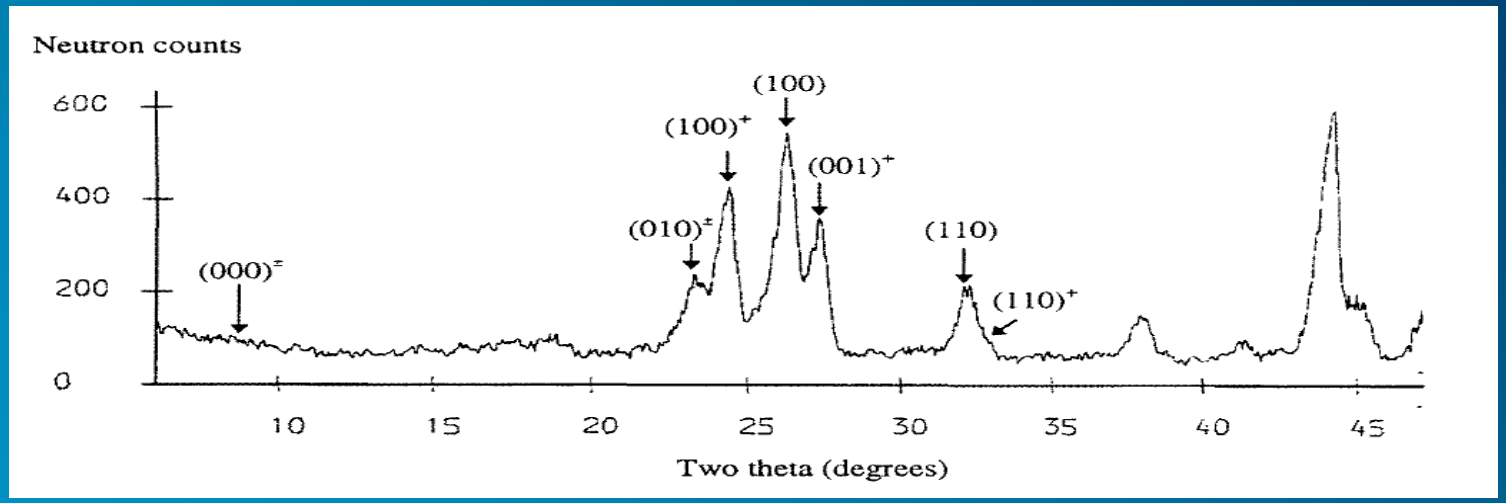
Only 3 ILL machines for more than ¼ of all proposals



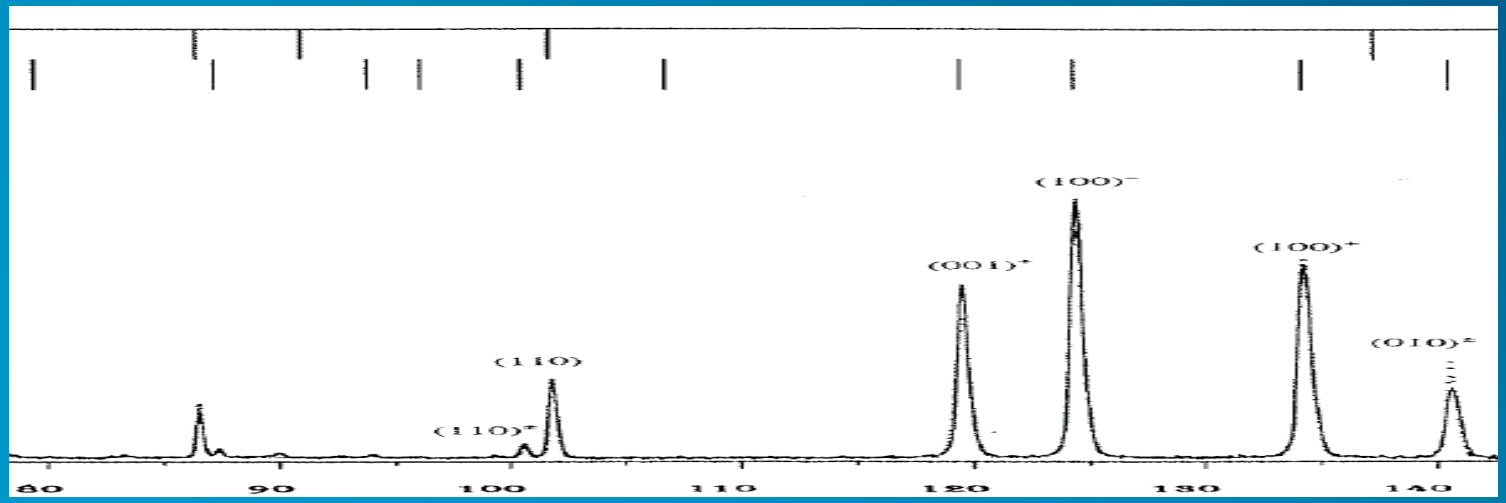
Magnetism and Biology Dracula (super-D16)
High resolution with long wavelengths

Helimagnetic order in ferric arsenate, FeAsO4

J B Forsyth, J P Wright, M D Marcos, J P Attfield & C Wilkinson (1999)



D1A



IRIS

GEM at ISIS, comparable to D20

Radaelli, Hammon & Chapon (2003) Neutroni e Luce di Sincrotrone

~700 minute GEM data for a 2mm^3 sample of $Y_3Al_5O_{12}$

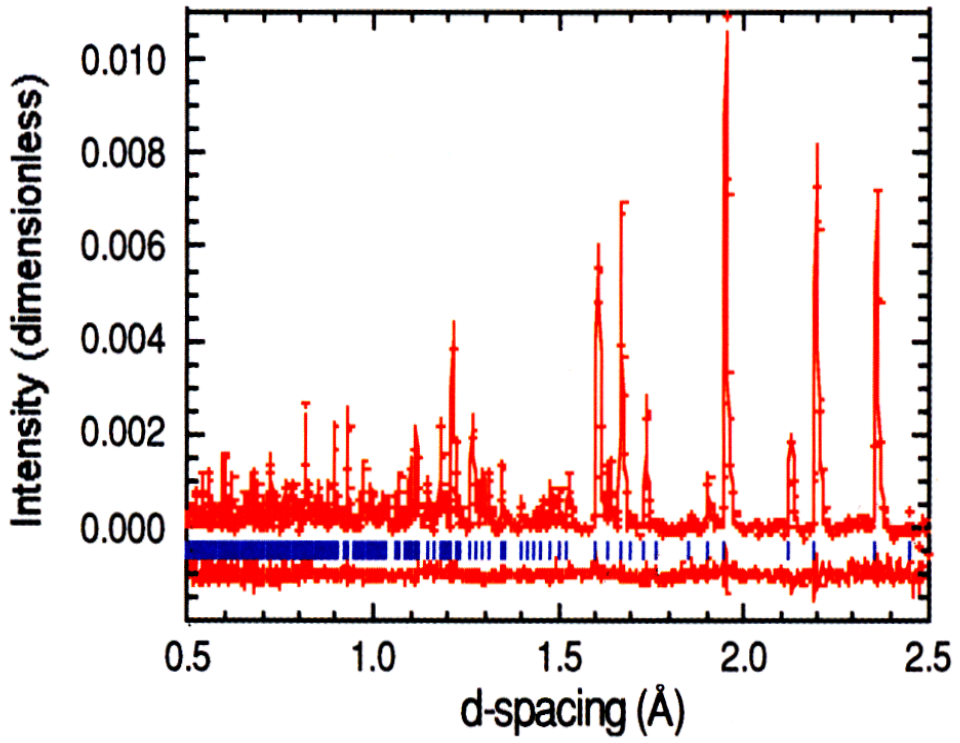
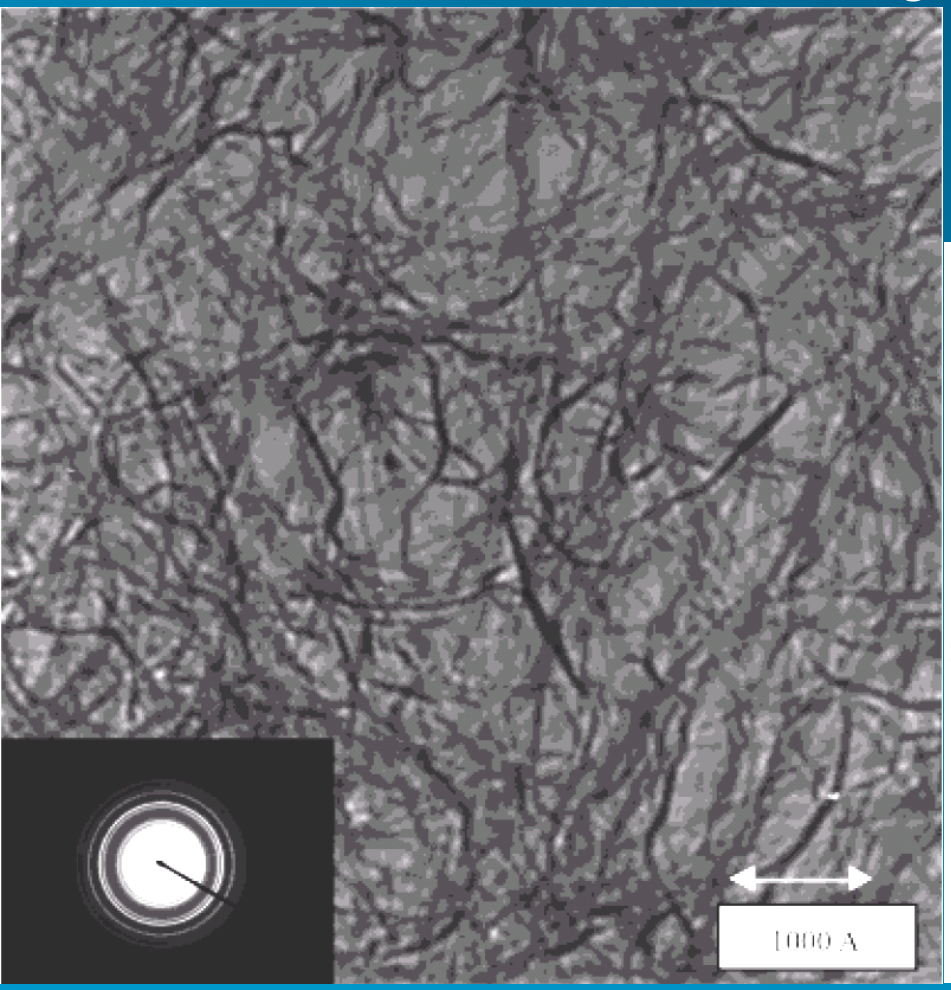


Fig. 7. Rietveld Refinement plot for a 2 mm^2 sample of Yttrium Iron Garnet (YAG), after an overnight data collection.

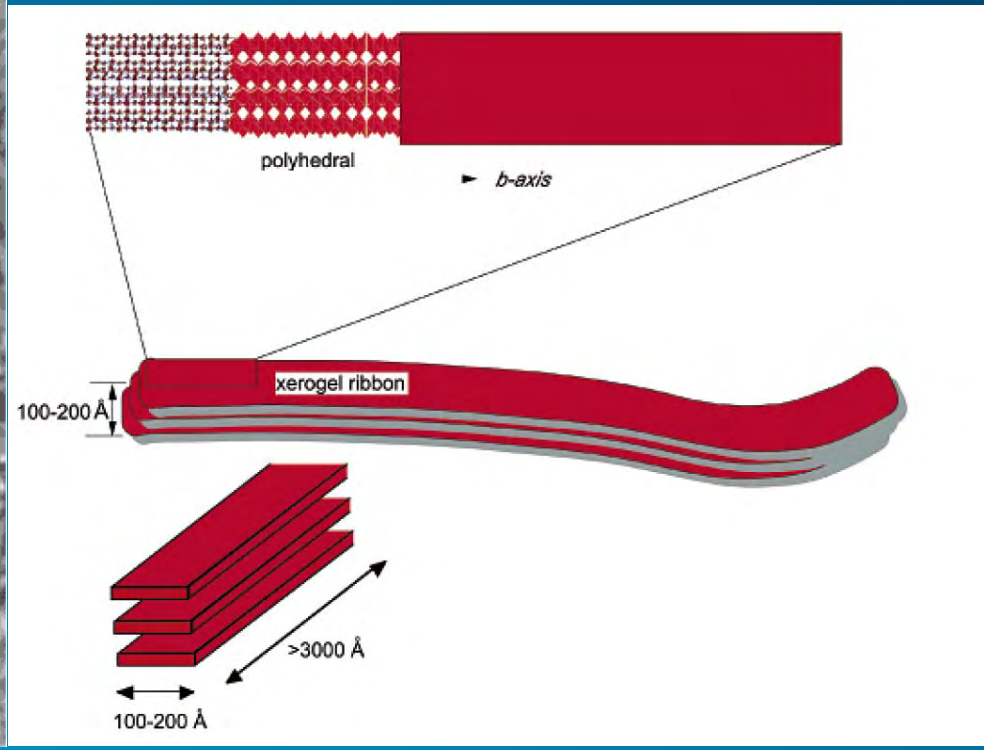
PDF and "Beyond Crystallography" Partially Ordered Materials - Super-D9 ?

$V_2O_5.nH_2O$ Xerogel by the Atomic Pair Distribution Function Technique

V.Petkov, P.Trikalitis, E.Bozin, S.Billinge, T.Vogt, and M.Kanatzidis (2002) JACS

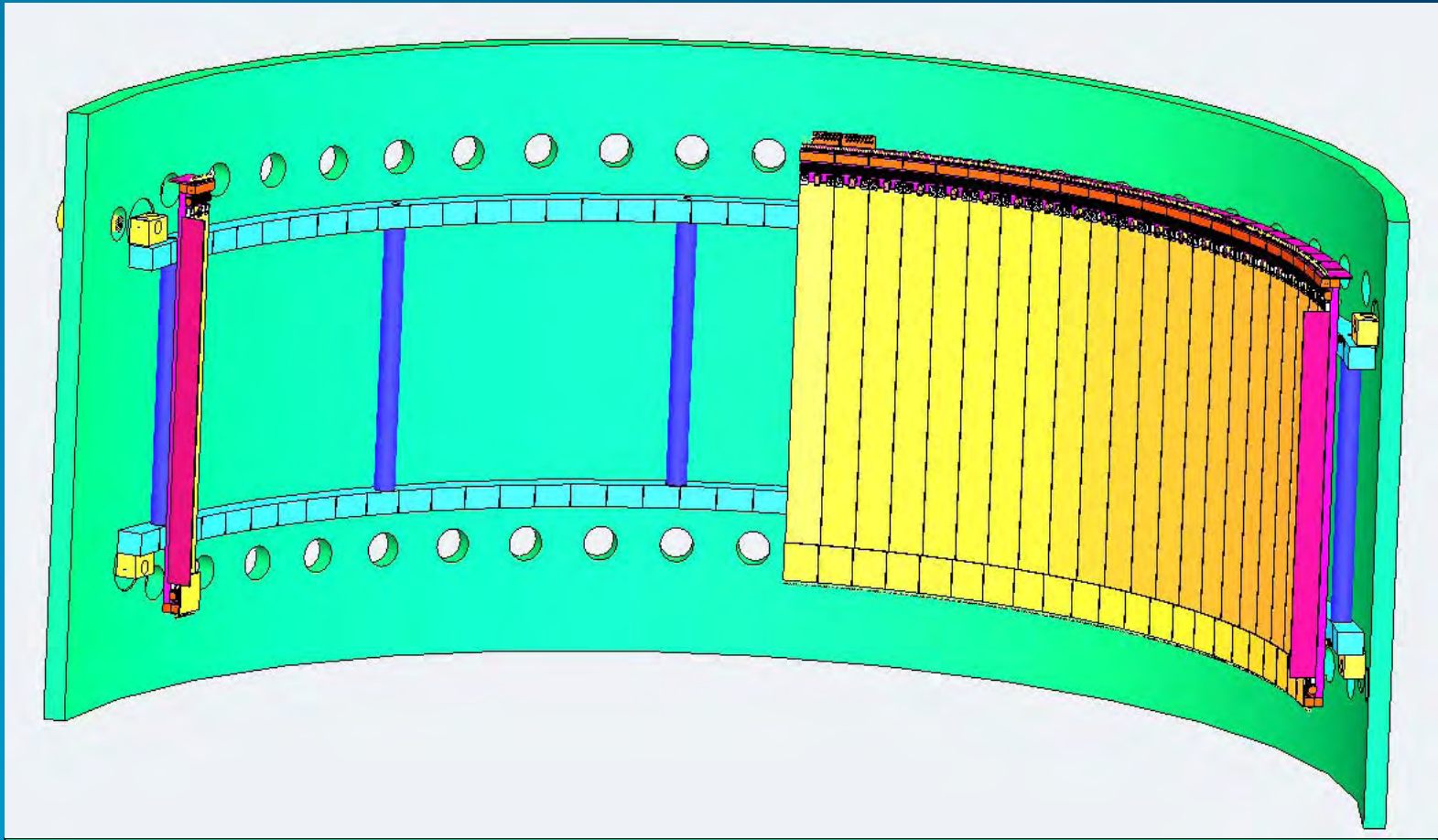


- Hydrogen-bonded sheets
- X-ray work only so far



Can we have a big detector too please?

D19 Millennium - A Revolution in large 2D Gas Detectors



2D with Solid Angle 1.5 steradian c.f. 0.27 on D20

Applications – What more can we do ?

ILL 2-axis machines that might use large 2D detectors

- High intensity, small sample powder diffractometer — DRACULA
- Partially Ordered Materials PDF Diffractometer — super-D9 ?
- Long Wavelength Magnetism/Biology Diffractometer — super-D16 ?