Progress with DRACULA?

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

Diffractometer for Rapid Acquisition
Ultra Large Areas
Pilgrims Progress...

Pulsed Sources 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)

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
Big monochromators give very high flux
D2B, D20, IN8…

Very high flux on the sample
- D2B $1.0 \times 10^7$ n.cm$^{-2}$.sec$^{-1}$
- D20 $9.8 \times 10^7$ n.cm$^{-2}$.sec$^{-1}$
- IN8 $6.5 \times 10^8$ 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

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
High flux compatible with good Resolution
DRACULA resolution matches peak density

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

DRACULA resolution
(minimum Δd/d~3*10^{-3})

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

Cagliotti

Required

Real

0.0° 0 90° 2θ
0 0.3° 0.6° 0.9° Δθ

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
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...

“Scientific opportunities with advanced facilities for neutron scattering”
Shelter Island Workshop, 1984

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


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.

<table>
<thead>
<tr>
<th></th>
<th>D20</th>
<th>GEM</th>
<th>DRACULA</th>
<th>SNS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flux average on sample</td>
<td>5x10⁷</td>
<td>~2x10⁶</td>
<td>~10⁸</td>
<td>~2.5x10⁷</td>
</tr>
<tr>
<td>Detector solid angle</td>
<td>0.27 sr</td>
<td>4.0 sr</td>
<td>1.5 sr*</td>
<td>3.0 sr</td>
</tr>
<tr>
<td>Efficiency=Flux*Detector</td>
<td>1.7</td>
<td>1</td>
<td>18</td>
<td>9</td>
</tr>
</tbody>
</table>

* Based on new D19 detector: R=760 mm, h=400 mm, 800 linear resistive wires 30°x160°
A High Resolution SNS Powder Diffractometer

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

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

\textbf{Constructed as HRPD at ISIS}

http://www.ill.fr/dif/AlanHewat.htm

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
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 \(x10\)

\[
\frac{\Delta d/d}{d} \sim 0.1\% \quad \text{for} \quad \frac{\Delta \lambda/\lambda}{\lambda} \sim 1\%
\]
2 minute D20 data for a ~700 mm$^3$ sample of Na$_2$Ca$_3$Al$_2$F$_{14}$

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
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
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
Very fast chemical and electrochemical kinetics

- 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

High-T Microwave Furnace Super-D2B (Boysen et al.)
...with Carsten Korte from Giessen (2004)
- DRACULA needs a high flux thermal beam tube
- The H9 beam tube has ILL’s highest flux
- Similar requirements for TOMOGRAPHY & DRACULA
DRACULA on High Flux Beam Tube H9
H9 similar dimensions to H11 (D20)

200mm
DRACULA on High Flux Beam Tube H9
H9 similar dimensions to H11 (D20)

DRACULA on H9 - Vertical beam from Lohengrin

- 200mm
- -> 175mm
Aufbau einer dynamischen Radiografie- und Tomografiestation mit thermischen Neutronen

DRACULA on High Flux Beam Tube H9
H9 similar dimensions to H11 (D20)
DRACULA+TOMOGRAPH on Beam Tube H9
Level C Floor Plan
DRACULA+TOMOGRAPH on Beam Tube H9
Level B Support
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**
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

Number of Papers (1999-2004)

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

Total 178

Source: ILL library

Most ILL Beam Tubes are Thermal

H11
Publications on ILL Thermal Beam-Tubes from ILL WWW pages


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

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
Large number of citations for ILL neutron powder work


- **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...

Undermines ESS?

If we want ESS we have to...

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

<table>
<thead>
<tr>
<th>Plane</th>
<th>d-spacing</th>
<th>Filter</th>
<th>d-range</th>
</tr>
</thead>
<tbody>
<tr>
<td>[115]</td>
<td>1.54 Å</td>
<td></td>
<td>0.89 Å - 1.54 Å</td>
</tr>
<tr>
<td>[113]</td>
<td>2.44 Å</td>
<td>(graphite filter)</td>
<td>1.39 Å - 2.44 Å</td>
</tr>
<tr>
<td>[111]</td>
<td>4.61 Å</td>
<td>(beryllium filter)</td>
<td>2.66 Å - 4.61 Å</td>
</tr>
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A High Resolution SNS Powder Diffractometer
B.E.F. Fender & A.W. Hewat (1978)

Figure 1

http://www.ill.fr/dif/AlanHewat.htm
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 λ < 2.2 Å, giving $\frac{\Delta t}{t} = 2.8 \times 10^{-4}$ at λ = 100 metres. (Steichele has 100 μsec at 143 metres giving $\frac{\Delta t}{t} \sim 3 \times 10^{-3}$).

ii) **Constant resolution** for λ < 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 ~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 ~10³), eliminating the need for the very large samples (~100 cm³) used at Munich, and making possible experiments on quite small (~1 cm³) hydrogenous samples of complex structure, dilute precipitates 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.

http://www.ill.fr/dif/AlanHewat.htm

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
Does large $\Delta \lambda/\lambda$ mean low resolution?

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

D20: Resolution $\Delta d/d \sim 0.2\%$

at high angles for $\Delta \lambda/\lambda \sim 1\%$
1) Diffraction
   a) aerogel
   b) crystalline

2) PDF
   a) aerogel
   b) crystalline

BOTH PDF functions have strong peaks to large r

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
Hydrated Vanadium Oxide Crystal
Y. Oka, T. Yao, and N. Yamamoto (1997)

$V_2O_5 \cdot nH_2O$ Xerogel

PDF and “Beyond Crystallography”
Partially Ordered Materials - Super-D9?
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 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 ?
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?
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
• 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?
The average count rate is the product of:

- the flux at the sample
- the detector solid angle and efficiency
- the sample volume
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.
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
Proposed ESS Powder Diffractometers

<table>
<thead>
<tr>
<th>Instrument Code</th>
<th>Instrument Description</th>
<th>Abbreviation</th>
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<tbody>
<tr>
<td>ST05</td>
<td>High-Q Powder Diffractometer</td>
<td>HQP</td>
</tr>
<tr>
<td>ST06</td>
<td>Liquids &amp; Amorphous Diffractometer</td>
<td>LAD</td>
</tr>
<tr>
<td>SM10</td>
<td>Single Pulse Diffractometer</td>
<td>SPD</td>
</tr>
<tr>
<td>SD17</td>
<td>Magnetic Powder Diffractometer</td>
<td>MagP</td>
</tr>
<tr>
<td>SD18</td>
<td>High Resolution Powder Diffractometer</td>
<td>HRPD</td>
</tr>
<tr>
<td>LM05</td>
<td>Ultra-high Resolution Powder Diffractometer</td>
<td>URPD</td>
</tr>
<tr>
<td>LM06</td>
<td>High Pressure Powder Diffractometer</td>
<td>HiPD</td>
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Comparison of TOF & CW Diffractometers

The time-averaged \textbf{Flux*Detector} criterium

D20 has high flux, GEM has a big detector

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<td>$5 \times 10^7$</td>
<td>$\sim 2 \times 10^6$</td>
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<td>detector solid angle</td>
<td>0.27 sr</td>
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<td>efficiency</td>
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The time-averaged **Flux*Detector** criterium

So, let’s use a big detector too!

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* Based on new D19 detector: R=760 mm, h=400 mm, 800 linear resistive wires 30°x160°
Thermal DRACULA on a High Flux Beam

DRACULA on H9 (replacing the Tomography station)
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...
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.
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.
Large detector & radial collimator near 90° scattering

±15° vertical as for the new D19 detector cf ±7° for new Paris-Edinburgh cell
±30° horizontal i.e., $2\Theta = 60° - 120°$ (range of scattering angles for pressure cell)

$D = 5\text{mm} - 7\text{mm}$ for $2\Theta = 60° - 120°$

$\Theta = \frac{d}{\sin(\Theta)}$

$= d\sqrt{2}$ (minimum at $2\Theta = 90°$)

$= 2d$ (maximum at $2\Theta = 60°$ & $120°$)

Scattering limited to a very small sample volume
DRACULA on High Flux Beam Tube H9

DRACULA on H9 - Horizontal beam from Lohengrin

200mm

Magnet

Shutter
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 \(\frac{1}{4}\) of all proposals

- SAS-Reflectometry (4 ILL)
- Single Crystals (5 ILL)
- Powders-Liquids (3 ILL)
- HR-TOF (7 ILL)
- 3-Axis (4.5 ILL)
- NP (5 ILL)

Alan Hewat, DRACULA, ILL Instrument Committee 4 Oct 2004
Helimagnetic order in ferric arsenate, FeAsO$_4$


~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


- 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 ?