DRACULA and the H12 beam tube renewal project

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Abstract

Dracula will be the world's fastest Diffractometer for Rapid ACquisition over Ultra Large Angles, competing favourably with the machines being built on the new American and Japanese pulsed sources. Dracula will be a third generation ILL machine, following the success of D1A/D1B and D2B/D20. The project was rated as top priority by the ILL Instrument Committee in 2003, and approved by the ILL Science Council in 2004. The idea is to take advantage of the very high time-averaged flux that a continuous source can deliver to the sample with modern neutron optics, and then to match the large detectors that have been used so successfully on pulsed sources. Instruments like Dracula will be essential if Europe is to maintain it's lead in neutron diffraction until ESS can finally be built.

Keywords: DRACULA; high flux; powder diffraction; neutron diffractometers;

1. Strategy for Dracula

A third generation neutron powder diffractometer will be one of the baseline instruments on the US-SNS, with installation starting in 2006 and operation by September 2007.

"SNS without a world-class powder diffractometer on day one is unthinkable." SNS-EFAC, May 2001.

In making their case for a new high flux neutron source, the American Shelter Island[1] workshop compared the relative advantages of pulsed and continuous sources. It was found that in many cases the two sources were competitive, and Shelter Island finally recommended the reactor source as being more flexible. But because of difficulties in building a high flux reactor significantly better than ILL, it was finally decided to build the pulsed source SNS instead. The comparisons made at Shelter Island remain however, valid, and serve to show how instruments on the existing high flux ILL reactor might be further optimised.

Jorgensen et al.[1] concluded that because neutrons are scattered isotropically, the relative merit of a powder diffractometer with a given resolution can be estimated from the product of just three parameters – the time averaged flux on the sample, the solid angle of the detector and of course the volume of the sample, which may be limited by resolution and other requirements. This product rule appears to be supported by the world's best high flux diffractometers, GEM on the ISIS pulsed source and D20 on the ILL reactor (below), which are roughly of equal merit, even though the time-averaged flux on D20 from the ILL reactor is more than an order of magnitude higher than on GEM.

	D20	**GEM**	**Dracula**	**SNS**
Time averaged flux	$5x10^7$	$2x10^6$	10^8	$2.5x10^7$
Solid angle (sr)	0.27	4.0	1.5	3.0
Sample volume	1	1	1	1
Merit product	18	8	150	75

Table. 1 The product of flux.angle.volume as a measure of merit.

This table shows that GEM competes only because of its exceptionally large detector, much bigger than anything that could be built until recently for reactor-based diffractometers. The pulsed source machine is unique in that the detector is not restricted to high resolution backscattering, but covers all scattering angles, with however correspondingly lower resolution.

Clearly if we could construct a diffractometer with similar design objectives, the high flux reactor machine would easily outstrip the medium flux pulsed source machine and compete with the high flux SNS pulsed source diffractometer.

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Indeed, modern focussing monochromators using very large beam tubes on a high flux reactor can deliver more flux to the sample than even the SNS pulsed source; fluxes greater than 10^8 n.cm^{-2}.sec^{-1} have already been achieved on ILL 3-axis machines, and they approach that level for D20, which is almost 20 metres from the reactor! The clear conclusion is that a modern diffractometer (D20 was conceived 20 years ago) would be superior to anything on the American SNS until ESS could finally be built.

The situation is not very different for single crystal machines with large detectors, especially for reactor-based machines such as quasi-Laue image plate detectors, where a white beam of neutrons is used as on a pulsed source, with the important difference that the beam is not pulsed! White beam diffractometers are the top priority of SNS, but they will not match ILL's time-averaged flux on the sample.

2. The Dracula Proposal

We proposed[2] to capitalise on recent ILL advances in the construction of large gas detectors and focussing monochromators to build a very high flux third generation powder diffractometer. This Dracula machine would be the world's most powerful neutron diffractometer, especially for the study of chemical kinetics, or small samples in special environments such as pressure cells. It would help maintain Europe's lead in neutron diffraction. The objectives are to:

- Construct a more compact machine than D20 to maximise intensity on the sample.
- Use big doubly focussing composite monochromators to deliver a large wavelength band.
- Provide sufficient space for extreme sample environments such as pressure cells to 100 kbar, magnets to 15 Tesla, very high temperature furnaces and neutron polarisers.
- Ensure $\Delta d/d \sim 10^{-3}$ resolution with low background by working at take-off angles around 90^o with radial collimation to eliminate scattering from special sample environments such as pressure cells.
- Use several different wavelengths in this focussing geometry to cover a wide range of d-spacings.
- Adapt a new D19-type 2D position-sensitive detector to cover an exceptionally large solid angle, integrating the diffraction cones as on super-D2B.

3. The Proposed H12 Beam Tube for Dracula

Stage 1 of the 2006 ILL Millennium proposal calls for a rebuild of the H12 beam tube, replacing IN4 with a new polarised neutron spectrometer (PASTIS) on a high-flux, low-background super-mirror thermal guide, together with a new thermal guide for diffraction. In particular, space will be made for DRACULA between IN20 and the guide protection, and as close to the reactor is possible.

This proposal will provide high-flux, low-background positions for at least three new instruments, and as such represents a much more cost-effective use of H12 than is possible at present. Indeed, the H12 proposal resembles that implemented 20 years ago on H11, where three world class instruments (D2B, D19, D20) share a single beam tube independently. Even if projects DRACULA and PASTIS did not exist,the H12 proposal would still be a good idea.

To obtain the highest flux for all three instruments, and to ensure their independence, it will be necessary to convert H12 to a 200mm diameter beam tube, as has been done for H11 and also more recently for IN8. Fortunately this is relatively easy, but of course calculations of possible reductions in reactor reactivity will be necessary. Even if the reactor cycle is shortened by a few hours, the interest of providing three new high flux beam positions is outstanding. (Future plans to increase the reactor fuel load would in any case increase the reactor cycle much more).

4. The Dracula Monochromator

As for D2B and D20, large focusing monochromators will be used, with even larger mosaic spread. As well as focusing in real space, focusing in reciprocal space means that a wide band of wavelengths $\Delta\lambda/\lambda$ can be used (fig.1). When this beam is scattered by the sample back into the direction of the beam from the source, all of the wavelengths enter the detector with the same incident angle; this means that the width of the Bragg peaks depends only on the size of the sample and detector elements, and the divergence of the beam from the reactor.

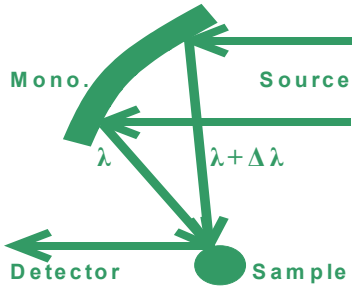

Fig. 1 Focusing of a large band of wavelengths still permits high resolution at high monochromator take-off angles.

Since very small samples would normally be used, and since a choice of Soller collimators can fix the horizontal divergence from the reactor, high resolution $\Delta d/d \sim 10^{-3}$ would be obtained even with a wavelength spread of ~1%. For angles near 90^o with a radial collimator, scattering would be limited to a small volume of diameter D (fig.2).

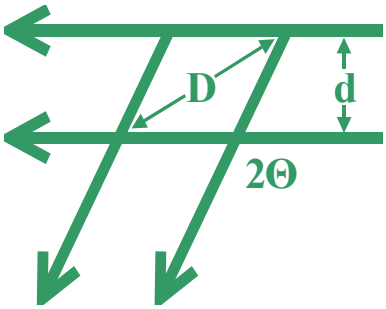

Fig. 2 For angles near 90° with a radial collimator, scattering would be limited to a small volume of diameter D.

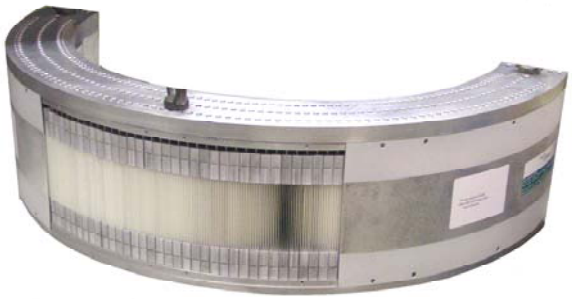

Fig. 3 The Dracula radial collimator will be similar to the new D20 radial collimator above (EuroCollimators, Cheltenham 2005)[3].

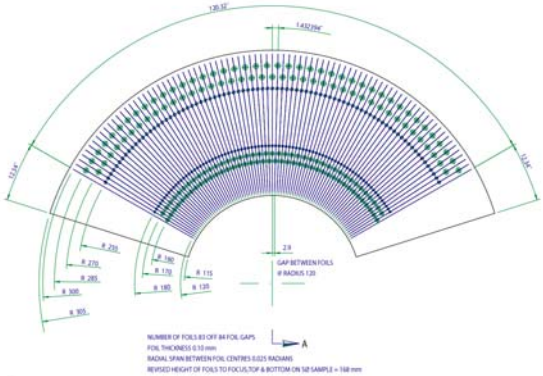

Fig.4. The Dracula radial collimator will cover a very large solid angle (160° x 30° vertical) yet restrict scattering to a very small volume of diameter ~5mm around the sample. (EuroCollimators)

A choice of fixed monochromator take-off angles would be available, probably 44°, 60°, 90° and 120°. The standard take-off would be 90°, but a low angle take-off to provide very high flux from a graphite monochromator would also be available, together with a high take-off of 120° for very high resolution. The main monochromators would be germanium [hhl] giving, for a take-off angle of 90°, a large choice of wavelengths and a range of d-spacings (table 2) for scattering within the restricted range 2Θ=60°-120°:

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

Table 2 The large range of d-spacings available within the limited scattering range 2Θ=60°-120° using just three wavelengths from a focusing Ge[hhl] monochromator.

Even if the scattering range is limited to 2Θ=60°-120°, which is attractive for reducing scattering from the sample environment with a radial collimator, a large range of d-spacings could be covered with just three different wavelengths, which would be obtained by simply rotating the Ge[hhl] monochromator about its vertical axis as on D1A/D2B. The most useful wavelength of 1.54Å would not require any filter, while a graphite filter would be used for 2.44Å and a beryllium filter for 4.61Å neutrons.

5. The DRACULA Sample Environment

Because of its very high flux, Dracula will be particularly powerful for measuring very small samples, as in high pressure cells. A new 100 kbar Paris-Edinburgh cell design, recently developed for single-crystal studies at ISIS, has been adapted for ILL to fit within a special cryostat (N. Kernavanois). This new pressure cell will be particularly suited to Dracula, with its high flux for small samples and radial collimator to eliminate background scattering from the sample environment.

Fig. 5 The new 100 kbar pressure cell with its He cryostat[4].

One of the problems with the present ILL diffractometers D2B and D19 is that they are situated under the reactor transfer channel, which limits the height of the sample environment. Dracula will not be limited in this way, and will allow even large cryostats to be used, such as our 15 Tesla magnet, the new 7 Tesla diffraction group cryomagnet, and cryostats equipped with dilution refrigerator inserts. Very high temperatures can already be obtained with mirror furnaces and microwave furnaces, and will also be available on Dracula.

6. The DRACULA Detector

This is the key element, and will provide Dracula with a detector x5 times as large as that of D20, which is already the fastest detector on a fixed-wavelength neutron powder diffractometer. Together with the extra flux on the sample, Dracula will then be an order of magnitude faster than D20 at the cost of slightly less resolution than available in D20's high resolution mode. Indeed, the solid angle of the Dracula detector will approach 50% of that of the fastest TOF machines, which can also measure at negative scattering angles without loss of resolution since monochromator focussing is not needed. The big difference will remain the very much higher flux on the sample due to the relatively wide wavelength band from the reactor.

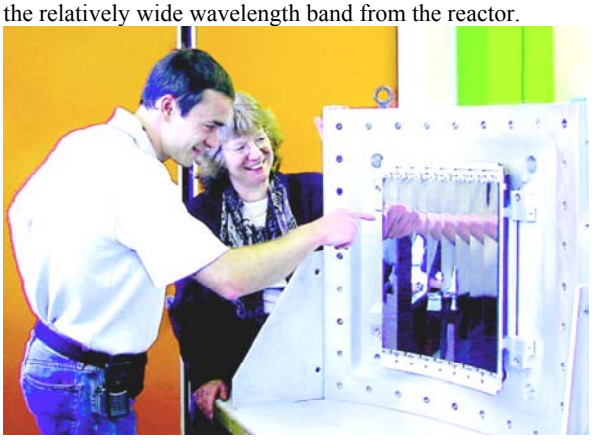

Fig. 6 A very large 2D PSD for D19, showing arrays of linear sensitive wires in front of electrodes deposited on glass (inset)[5].

The Dracula detector will be almost identical to the new D19 detector, except that extra modules will extend coverage to 160° as on D20, instead of 120° as on D19. The vertical acceptance angle will remain at 30° . The principle of using a large 2D detector for powders has already been proven on super-D2B, where it was shown that the diffraction cones can be integrated around their curved sections without substantial loss of resolution.

With a horizontal definition of at least 2.5mm, the Dracula detector will be resolution-matched to samples as small as 2.5 mm diameter. For even better definition of the powder profile, it will be possible to fine-step the Dracula detector on its tanzboden floor, as on D2B.

Fig. 7 The very large 2D PSD for D19, showing arrays of linear sensitive wires in front of electrodes deposited on glass (inset) [5].

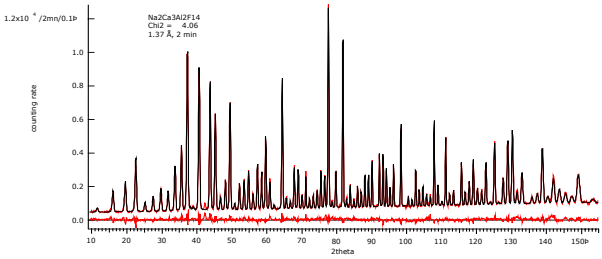

Fig. 8 The complete refined diffraction pattern obtained from $Na_2Ca_3Al_2F_{14}$ in only 2 minutes on D20 in high resolution mode[6].

The complete neutron pattern in fig.5, which was obtained in just 2 minutes on D20 can be refined to give a high precision structure, as indicated by the red obs-calc difference pattern in the profile plot. On Dracula we might expect to obtain similar results in a matter of seconds, or a few 10's of milliseconds for large samples when studying chemical kinetics.

For very small samples, such as those in high-pressure cells, we might expect to collect good quality high-resolution data from samples of a few 10's of milligrams. The sample volume in the new 100 kbar cell, at ~4 mm^3, is large enough to allow very short counting times, and this would mean that data could be collected as a function of temperature even at very high pressure!

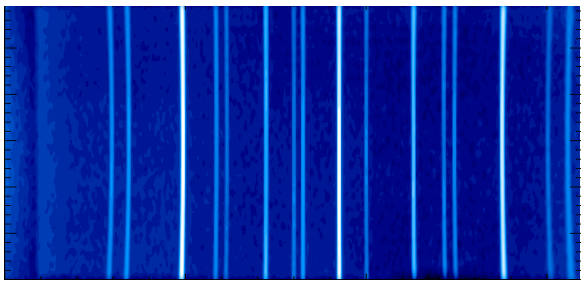

Fig. 9 A high resolution 2D detector array is needed to correct for the curvature of the diffraction cones on the new super-D2B[7].

7. New Science on DRACULA

7.1. Magnetism and Polarised Neutrons

DRACULA would be the first thermal neutron powder diffractometer with polarised neutron capability. The interest of using polarised neutrons for powder diffraction has been demonstrated by Wills et al.[8] using a ^{3}He spin filter between the monochromator and the sample. Eventually a second ^{3}He filter between the sample and detector could provide polarisation analysis.

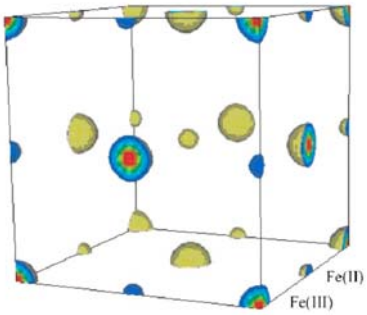

Fig. 10 Maximum Entropy reconstruction of the magnetisation density in Prussian blue determined using polarised neutron difference powder diffraction on D1B at ILL.

7.2. Magnetism under exotic conditions

The study of materials that undergo magnetic transitions at high pressure usually involves very small samples. Rochette et al[9]. have recently shown that pyrrhotite undergoes a ferrimagnetic to paramagnetic transition at about 2.8 Gpa, and this has been used to explain the magnetisation distribution around impact craters on Mars, where impact has generated high pressures. One might even imagine using periodic shock waves to generate very high pressure on a fast machine like Dracula.

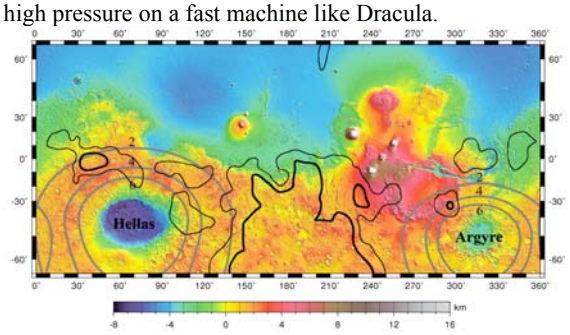

Fig. 11 magnetisation density around Martian imapact craters from D20 measurements by Rochette et al[9].

7.3. High Pressure and Extreme Sample Environments

High pressure is easier with X-ray diffraction because high fluxes, especially from synchrotrons, mean that very small samples can be used. Yet there are many problems where it would be valuable to be able to use neutrons as well – for the same reasons that neutrons are needed for materials at zero pressure. Many electronic and magnetic properties depend on pressure, we need to understand why, and these are just the kind of problems where neutrons are most useful. Clearly, the maximum pressure that can be obtained with a cell small enough to fit in a cryostat scales with the size of the sample, so high pressure necessarily means small samples and high flux diffractometers like Dracula. TOF diffractometers have some advantages for high pressure, but Dracula will also collect all d-spacings with different wavelengths over a limited scattering range.

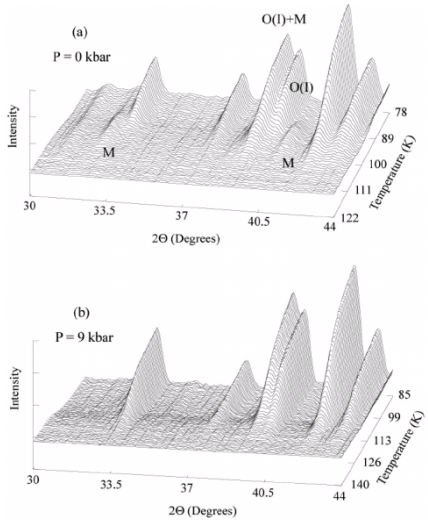

Fig. 12 Pressure Enhancement of the Giant Magneto-Caloric Effect in $Tb_5Si_2Ge_2$ on D20 on cooling (a) 0 kbar (b) 9 kbar hydrostatic according to Morellon et al.[10].

7.4. New Materials and High Pressure Synthesis

New materials are usually only available as very small polycrystalline samples, and X-ray powder diffraction is often not sufficient to understand their structure. Examples are the high Tc superconductors[11], GMR and other magnetic materials. This is particularly true when new materials are synthesised under high pressure; yield volumes are small, and several samples must often be combined for neutron measurements, with the corresponding risk of inhomo-geneity. A diffractometer with an order of magnitude increase in efficiency would be of great benefit in the search for new materials synthesised at high pressure.

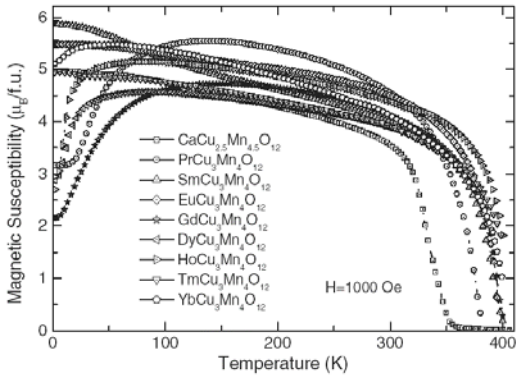

Fig.13 Temperature dependence of the DC magnetic susceptibility of new $RCu_3Mn_4O_{12}$ compounds prepared for the first time under 2 GPA pressure. Neutron structure measurements were on D2B by Sánchez-Benítez et al[12].

7.5. Isotope replacement

Isotope replacement is a potentially powerful method for investigating the role of particular atoms in a structure, yet because of the cost of producing large samples, this technique is often restricted to deuteration. Yet isotope replacement has already demonstrated its interest for superconducting materials, where both the isotope effect predicted by BCS theory, and the effect of magnetism on specific atoms, has been investigated. Dracula would make isotope replacement feasible for a wider range of problems, since much smaller samples would be required.

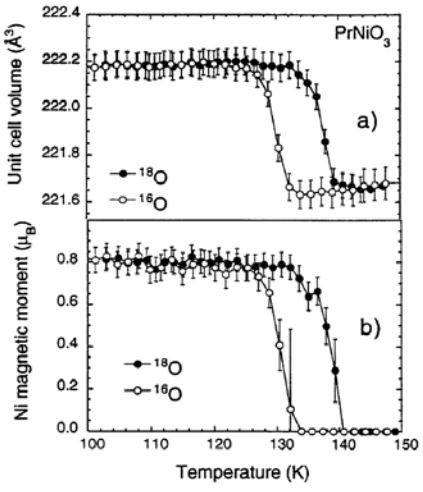

Fig.14 Temperature dependence of (a) unit cell volume (b) Ni^{3+} magnetic moment across the metal-insulator T/N for isotopically substituted $PrNi^{16}O_3$ measured on D1A by Medarde et al[13].

7.6. Strongly aborbing elements

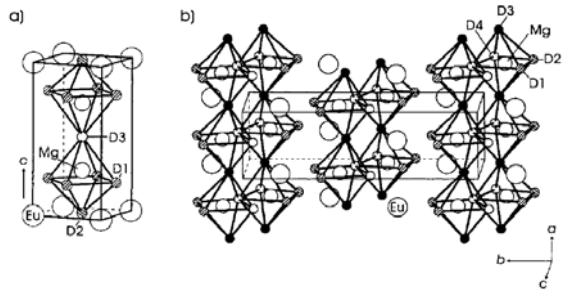

Fig 15. Location of hydrogen in heavily absorbing Europium compounds on D20 by Kohlmann et al[14] a) $EuMg_2D_6$ b) $EuMgD_4$

One of the big advantages of neutrons over X-rays is their low absorbtion for most materials. Yet neutrons are strongly absorbed by a small number of rare earth elements whose compounds are increasingly interesting. For example, it is difficult to study magnetism in these materials, or absorbtion of hydrogen etc., and it is not always feasible to use sufficiently energetic neutrons. This problem can be addressed by using very thin samples, but again a highly efficient neutron diffractometer is needed.

7.7. In-situ chemical kinetics

Because they can penetrate relatively large chemical and electro-chemical cells, neutrons have long been of interest for the in-situ study of chemical reactions. For example, the chemical reactions that occur in a full-size nickel oxyhydroxide battery can be studied as the cell is discharged and recharged. Intercalation experiments, especially those involving hydrogen or other light elements, are ideally studied with neutron diffraction. Classical examples include hydration of minerals, hydrogen storage materials, zeolites, clathrates etc.

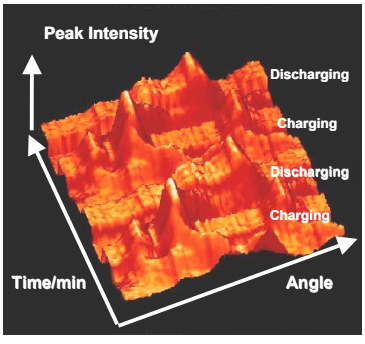

Fig. 16 Electro-chemical reactions in a nickel oxyhydroxide electrode on D20 by Barde et al[15].

7.8. Hydrates, Clathrates and Ice Structures

Neutron diffraction is of course ideal for the location of light atoms, and in particular hydrogen; hydrogen bonding is fundamental to the structure of most hydrogenous materials, many of which cannot be obtained as crystals. The ice clathrates are an example where relatively little is known, even about their density, yet these materials will be important for the deep sea "mining" of hydrocarbon fuel (methane) and the storage of carbon dioxide waste at the bottom of the ocean. Kuhs et al.[16] have used neutron diffraction on D2B/D20 for the structure of these materials.

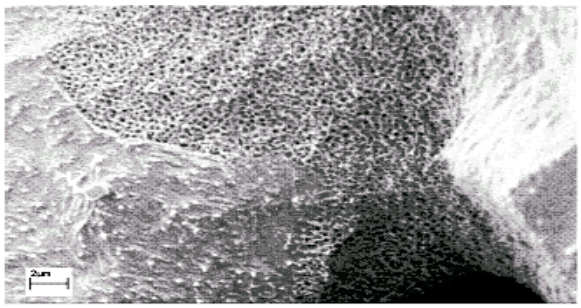

Fig. 17 Hydrate spreading into the open pore space of a porous ice sample[16].

7.9. Very fast reactions

Many other chemical reactions are simply too fast for neutron diffraction. The order of magnitude increase in speed that would be possible with Dracula would open up many new areas of chemical kinetics that cannot adequately be studied with X-rays. The explosive SHS reaction[17], occuring on a time scale of 100's of ms is a good example.

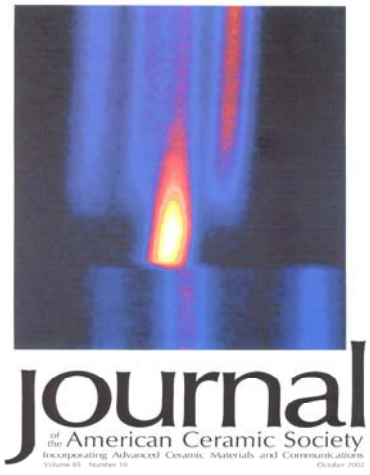

Fig. 17 The explosive SHS reaction[16] in Ti_3SiC_2 on D20. The intermediate phase (bright spot) lasts only ~1 second.

7.10. Non-equilibrium structures with very small samples

Usually we study the structure of rather large volumes of material under equilibrium conditions, but short-range order under non-equilibrium conditions offers fascinating insight into structural stability. For example, neutron diffraction by Schenk et al.[11] on small electro-magnetically levitated liquid drops has shown how supercooled metals can form short-range icosohedral clusters.

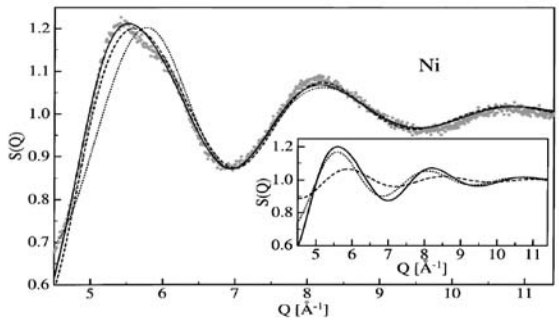

Fig. 18 Measured structure factor S(Q) of an electromagnetically levitated Ni drop at T=1435K on D20 showing evidence for icosohedral clustering (shoulders on the S(Q) peaks)[17].

7.10. Magnetic structural transitions

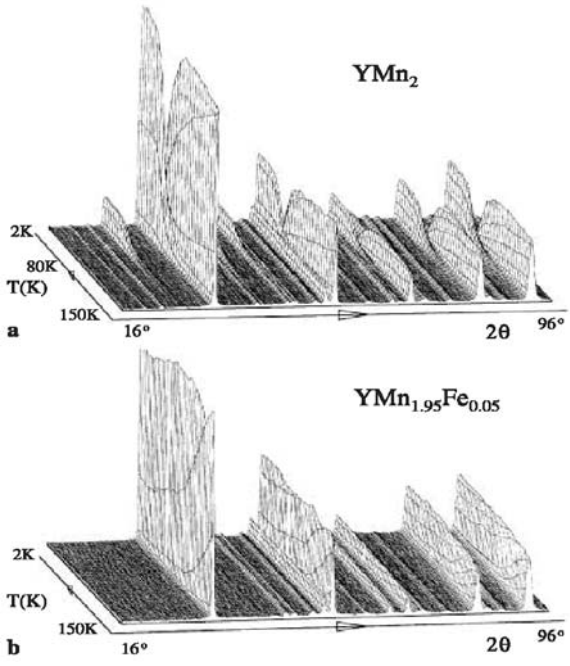

Fig 19 Neutron thermogram of a) YMn2 and b) $YMn_{1.95}Fe_{0.05}$ from D1B showing antiferromagnetism at 80K is supressed by 2.5% of Fe on the Mn-site (Cywinski et al[18]).

Magnetic structural transitions are a classical example of high flux neutron powder diffractometers. Often the magnetic peaks are very weak, requiring long counting times, especially if we want to study the temperature/field dependence, or the effect of changes in stoichiometry.

7.11. Texture

A large 2D detector is ideal for the study of the texture and preferred orientation of materials, such as mineral and geological samples, as well as industrial components. For example in High-Tc superconducting materials such as Bi2223, Guilmeau et al[19] have shown how critical current densities are strongly dependent on texture, crystallite size and phase ratios.

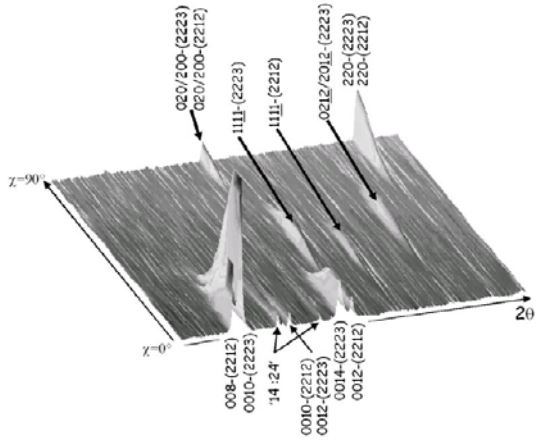

Fig. 20 Neutron diffraction pattern obtained for Bi2223 samples for a 0–90º χ-scan on D1B by Guilmeau et al[19]. The preferred orientation (texture) is clearly visible.

7.12. Single crystal diffraction

Finally, the large 2D detector of Dracula will be equally powerful for single crystal studies, where many reflections might be obtained simply by spinning the crystal. This is foreseen for the D19 large structure diffractometer; a machine like DRACULA is needed for smaller structures.

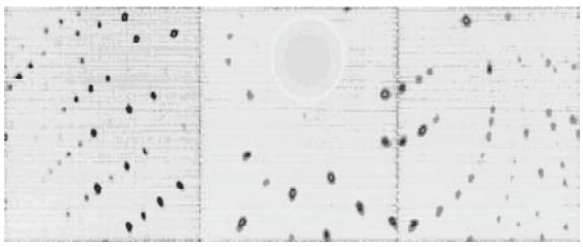

Fig. 20 Single crystal diffraction pattern from the small 2D-PSD for D19, which is also the prototype for the Dracula detector[20].

8. Conclusions

The main disadvantage of neutron scattering is the low flux compared to X-ray diffraction. An order of magnitude gain would have a big impact on many problems in chemistry and physics, and open up whole new areas for neutron diffraction. Eventually we must build a very high flux neutron source like ESS. But it is clear that we could already do more with the neutron sources we now have, which in the case of reactors, provide high time averaged flux to the sample. Advanced detector technology has made it possible to make far better use of already available neutrons for a relatively modest cost.

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