CYCLOPS, A proposed high flux CCD neutron diffractometer

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Abstract

New single crystal diffractometers on spallation sources, with ultra-large area detectors, such as TOPAZ at SNS and SXD-II at ISIS, will allow real-time exploration of reciprocal space and rapid structure refinement. ILL pioneered similar techniques with image plate machines, and retains a potential advantage because of the high flux on the sample from a continuous white neutron source. Yet neutron image plates are not real-time, are only ~20% efficient, and the image decays even as the data is collected. These differences have been made startlingly clear by recent in-situ comparisons of LADI with electronic PSD's. We propose to build a new machine, the **CY**lindrical **C**cd **L**aue **O**ctagonal **P**hoto **S**cintillator, using methods similar to those used for SXD, but with much higher flux. A small prototype machine at ILL, OrientExpress has already proved the principle.

Keywords: CYCLOPS; CCD detectors; neutron diffractometers;

1. Neutron Image Plates and Electronic Detectors

Neutron image plates (NIMs) have proved important for increasing the efficiency of neutron diffractometers[1,2] just as they once were for X-ray crystallography. Although they are not very efficient, and do not provide real-time read-out, they are a cheap way of covering large angular ranges. However, it has always been clear that such film techniques will ultimately give way to electronic detectors for neutrons, just as they have for X-rays.

Recently Guerard and Wilkinson[3] compared *in-situ* diffraction patterns obtained with the original LADI neutron image-plate detector and a 2D electronic gas detector (fig.1). An 8mm^3 lysozyme crystal was exposed for 12 hours on LADI, and then the image plate was rotated to allow the diffraction pattern to fall instead on the prototype D19 ^{3}He multi-wire gas detector. Within 1 hour, the diffraction pattern from the gas detector was already better than that from the image plate. Not only was the gas detector much more efficient, but the peak-to-background ratio was also much better, due to γ-ray discrimination.

This comparison is a little unfair, because the original LADI is scanned from the back of the image plate; the new machine LADI-III will be scanned from the front, giving higher efficiency. However, the image plate used was the most efficient available, and identical to that for the new VIVALDI[4] and LADI-III machines.

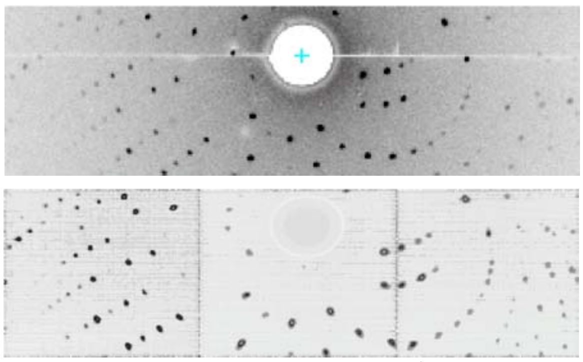

Fig. 1 (a) Diffraction pattern from an 8mm^3 lysozyme crystal exposed for 12 hours on the original LADI compared to (b) the same crystal exposed for 1 hour on the prototype D19 gas detector. Three exposures were needed with this small detector (lines).

These quasi-Laue machines use a white beam of neutrons, like TOF diffractometers on pulsed sources. However, since the reactor neutron source is continuous, the time-averaged flux on the sample is normally much higher than for a pulsed beam from a spallation source. For example, the time-averaged flux on the sample for SXD at ISIS[5] can be estimated at ~2x10^6 n.cm^2.sec^{-1} (value measured for GEM[6]), while the time-averaged flux predicted for the thermal super-mirror guide feeding the ANSTO quasi-Laue machine[7] exceeds 10^9 n.cm^2.sec^{-1} ! One might expect to do even better at ILL.

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2. Neutron CCD detectors

A review[9] of the different techniques available 20 years ago for position-sensitive detectors (PSDs) for neutron diffraction included the “Anger” camera[7] based on neutron scintillator plates and photo-multiplier (PM) tubes. The first Anger cameras for neutron diffraction were constructed on pulsed neutron sources at IPNS[10], KENS[11], LANSCE[12], and ISIS[5]; Schultz[13] reviewed these early machines. The original SXD detector at ISIS consisted of a 300x300x2mm ^{6}Li-glass scintillator coupled via a light diffusing layer to an array of 45 PM tubes. The co-ordinates of scintillating points were determined on a 128x128 pixel grid from the relative outputs of adjacent tubes. Optical fibre coupling as originally used at KENS was later employed, and the solid angle extended to cover ~2π. Because of the desire for TOF analysis, these detectors require much faster electronics than for simple accumulation over time scales of seconds, as proposed by Arndt & Gilmore[14] using TV scanning.

Heidemann[15] and others constructed simple neutron area detectors consisting of a CCD TV camera scanning a scintillator plate reflected in a 45^{o} mirror, to take the camera out of the neutron beam. Such a detector was used for the first tests of neutron tomography at ILL, for which a more advanced system was later devised. It was proposed then to use the original Heidemann detector to replace the photographic film method used to align crystals at ILL.

This proposal eventually resulted in an entirely new detector for the OrientExpress[16] machine constructed by Ouladdiaf at ILL, and based on CCD technology developed for synchrotron X-ray sources. Instead of an array of photo-multipliers as in the Anger camera, Photonic Science[17] use a pair of thermo-electrically cooled image-intensified CCD cameras viewing a 0.4mm thick 252x198mm ^{6}LiF:ZnS:Ag neutron scintillator in transmission. Lenses focus images of the scintillator onto image intensifiers, which are coupled tightly to the cameras using fibre-optic tapers bonded directly onto the CCDs.

The output from the cameras is automatically scaled and stitched together to produce a single image of 1680x1320 pixels, corresponding to a resolution of 150 μm. Since this resolution is often much better than required for neutron diffraction, these pixels can be binned to increase intensity. This image data is collected using a firewire interface to a dedicated PC, where the LAUEGEN[18] program suite from CCLRC-Daresbury is used to index the pattern. The effective efficiency of this CCD detector is ~100 times that of the photographic neutron camera that it replaces, though in absolute terms it is estimated at ~25% for the 1-4Å wavelength band used. For comparison, the efficiency of the best “Niimura special” neutron image plates is ~20%.

The diffractometer itself[19] is composed of the CCD detector mounted on a 2θ arm, a two-stage tilt goniometer with ω rotation around a vertical axis, and a video system for the optical alignment of the crystal with respect to the incident neutron beam (fig.2).

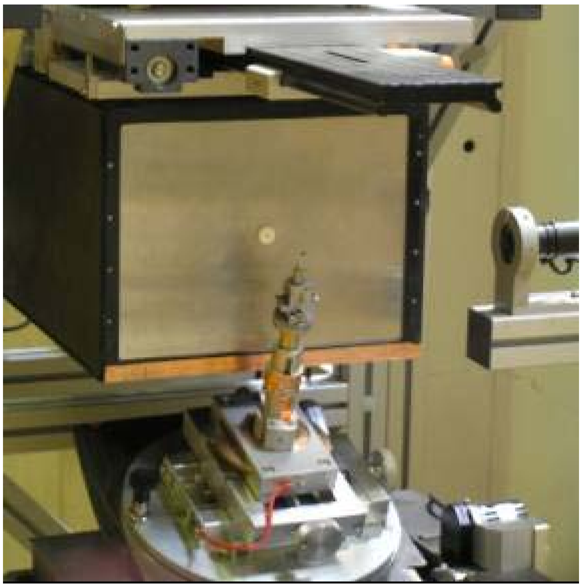

Fig. 2 OrientExpress at ILL is a CCD detector on a continuous white neutron beam from a thermal guide (Ouladdiaf et al[16]).

The detector-sample distance can be set between 32-220 mm. The detector is normally used in backscattering to maximise alignment precision, with the incident neutron beam arriving through a small evacuated tube through the center of the detector, but it can also be used in forward scattering. All motors of the diffractometer are controlled through the standard ILL instrument control program MAD running under LINUX.

In 10 seconds OrientExpress can collect a diffraction pattern from a small ruby crystral that previously took at least 10 minutes with photographic film ! Figure 3 shows such a pattern, consisting of a large number of Bragg peaks perfectly indexed by the LAUEGEN software.

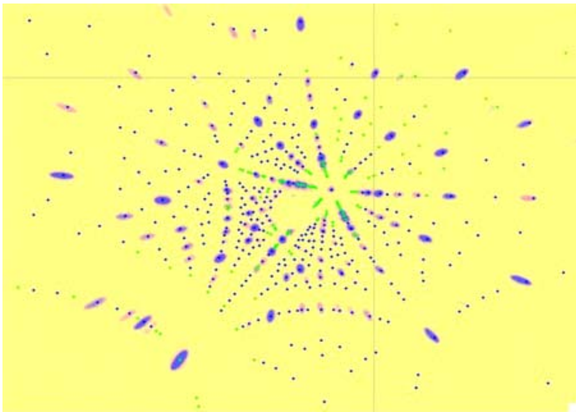

Fig. 3 Neutron diffraction pattern from a standard ruby crystal obtained in 10 seconds on OrientExpress, and automatically indexed with the LAUEGEN software (small points).

3. CYCLOPS, a high-flux CCD detector on a reactor

CYCLOPS[20] would be a much larger version of OrientExpress covering a >2π solid angle like SXD-II, but on a focusing super-mirror thermal guide. With an almost white beam from the continuous neutron source, the flux on the sample would be much higher than possible on a pulsed source. Although the ability to use TOF to sort neutrons according to their energy would be given up in return for this higher flux, the machine would retain the advantages of neutron image-plate machines such as VIVALDI while providing real-time read-out and greater efficiency.

While VIVALDI is unequalled for the refinement of structures from very small crystals, and SXD-II is unique for background discrimination based on TOF, CYCLOPS would be the ideal machine for the rapid scanning of reciprocal space in real time, as a function of temperature, pressure etc. As such it would represent a unique facility for the study of structural and magnetic transitions, including modulations, incommensurable structures, and transient phenomena.

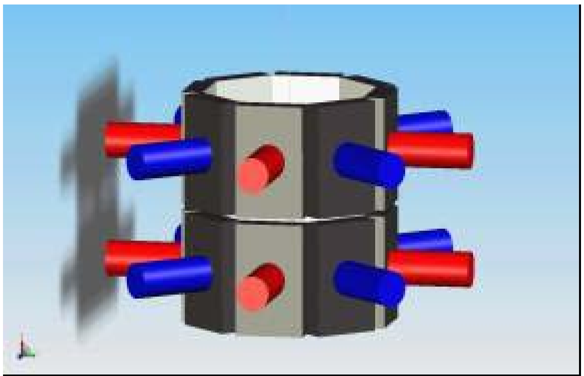

Fig. 3 Schematic drawing of the octagonal CCD detector array for CYCLOPS covering a cylinder 400mm high by 400mm diameter.

The CYCLOPS detector (fig.3) would consist of an octagon of CCD plates each 153mm wide x 400mm high. These plates would form a hollow vertical cylinder of 400mm diameter (between octagonal flats) and 400mm high, covering 70% of 4π. The solid angle subtended by an octagonal cylinder is almost identical to that of a true cylinder of height h and diameter d, being a fraction $\sin\phi$ of 4π where $\phi = \arctan(h/d)$ is the angle of elevation of the top of the cylinder. For h=d=400, $\phi = 45^o$ and this fraction is $1/\sqrt{2}$. Even doubling the detector height would only increase this fraction from 70% to 89%.

The neutron beam would enter through a 20mm hole in the equatorial plane in the middle of the detector, and exit through a second hole opposite. Each of the eight segments would be scanned by two cameras aligned vertically giving a resolution of ~160 microns. These 16 camera images would be collected in rapid sequence via firewire to a Windows computer, where they would be geometrically corrected and automatically stitched together to form a single panorama of diffraction space containing a total of ~20M 16-bit pixels. This technology is very similar to that being developed by Photonic Science and partners in the European OPTAG project for real-time panoramic video security monitoring at 320M colour pixels/second !

The 20M pixel diffraction panorama would be rebinned if necessary to increase signal, and then indexed and displayed with the same LAUEGEN[18] suite used on Vivaldi and OrientExpress. Eventually software to fit overlapping peaks (2D Rietveld refinement) would be developed to enable intensity to be increased by relaxing the divergence of the incoming beam, and thereby the angular resolution.

In order to maximise the flux on the sample, CYCLOPS would be positioned at the end of a small m=2 thermal super-mirror guide. An optional focusing trumpet could be inserted at the end of this guide to further increase the flux on the sample in the "higher-flux, lower-resolution" configuration using longer wavelengths and/or smaller unit cells. A choice of multi-wafer filters would be used to select a wavelength band in the range $0.8<\lambda<3.0$Å, depending on unit-cell dimensions. The white neutron flux on the sample would then easily exceed 10^9 n.cm^{-2}.sec^{-1}, greater than possible on any other neutron instrument.

The mechanics would remain rather simple, as is possible for quasi-Laue geometry, with a large two-stage tilting table to support heavy sample environments, and provision for rapid rotation of the sample around a single near-vertical axis.

The standard sample environment would include a cold-hot gas stream to allow rapid scans with temperature and easy sample changing; sample alignment need not be precise, since it would be determined anyway from the complete pattern at the end of each measurement. Other sample environments, in particular a cryo-refrigerator with furnace insert, would be available, and indeed the 400mm inside diameter of CYCLOPS would be such that most of the standard ILL diffraction environments, including cryo-dilution inserts and a superconducting cryomagnet might be used.

An optional oscillating radial collimator, as on D20, could be used to remove background from the sample environment, which would make CYCLOPS particularly attractive for very high pressure measurements on small single crystals.

4. Complementarity of CYCLOPS and SXD-II

Although both CYCLOPS and SXD-II[21] use a 2π array of ^{6}Li-glass scintillators, they are otherwise very different machines with rather different applications. With its TOF energy resolution, SXD has the advantage of lower background, especially for hydrogenous samples, but also very much lower flux - a factor of ~10^3. SXD is then ideal for the detailed structural study of small molecules when crystals are sufficiently large, such as simple drugs, as pioneered by Wilson[22]. In particular, crystals of the actual drug can be studied, without the need for deuteration, which may change the hydrogen bond geometry. The TOF geometry also allows greater relaxation of the incident beam divergence, necessary to gain intensity; the energy resolution of the machine allows peaks to be more easily resolved, since they are distributed over a 3D space instead of the 2D space of the integrating detector.

CYCLOPS would however make use of its higher integrated flux, especially with inorganic materials, to allow experiments on much smaller samples. More importantly, it would provide real-time surveys of large volumes of reciprocal space under various conditions of temperature, pressure and magnetic field. Crystals of various stoichiometry could be compared due to the simplicity of sample changing and speed of data collection. While SXD would excel for "classical" structure determination and refinement of hydrogeous materials, CYCLOPS would instead be a kind of panoramic reciprocal space explorer.

5. Complementarity of CYCLOPS and VIVALDI

The CYCLOPS geometry is of course very similar to that of VIVALDI[23], although the detector is very different. VIVALDI's flexible neutron image plates mean that a large surface can be covered without the need for other than simple geometrical corrections out of the equatorial plane. Neutron image plates are also very cheap, although at present we are limited to a single manufacturer.

The geometrical corrections needed for CYCLOPS are however well understood and tested on OrientExpress, and are only a matter of computing power, which is no longer a problem. The total cost of the instrument is quite modest compared to that of most other neutron instruments, and in particular very much less than the more complex scintillator array on SXD-II.

Like SXD, VIVALDI would remain important for the precise structure refinement of small crystals with rather long counting times (hours). CYCLOPS would instead combine some of the advantages of both machines – real-time data collection together with very high flux on the sample, meaning fast surveys of small samples. It will be possible on CYCLOPS to rapidly scan a very large region of reciprocal space, and then watch on a TV screen what happens in real time within various smaller regions as the sample environment changes.

This would be a truly unique new way of studying the dynamic structure of new materials with neutrons.

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