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D4c: A very high precision diffractometer for disordered materials

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Abstract. The disordered-materials diffractometer D4 at the Institut Laue-Langevin (ILL) has been thoroughly upgraded through improvements in detectors, collimation and shielding to become the D4c instrument. A larger solid angle of detection has increased the total counting rate by a factor of 5, thereby reducing random error in the diffraction measurement, and a corresponding factor of 5 improvement in detector stability has reduced the principal cause of systematic error. The overall precision of the instrument has therefore been increased by a factor of 5 as compared to its previous version, D4b. We present an overview of the D4c instrument's design as well as some results of the successful very high precision experiments performed at D4c since its commissioning in May/June 2000.

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To increase detection solid angle and stability, the D4c project replaced the 2 multiwire detectors of the D4b liquids/glasses diffractometer with 9 microstrip detectors in a fan-like array (Fig. 1). The usual configuration for D4b placed its two detectors at radii of about 1.5 m and 0.75 m (giving 0.1° and 0.2° per cell in scattering angle 2θ, respectively). The 9 D4c detectors at a radius of 1.146 m (0.125° per cell) have the same width (160 mm) but increased height (100 mm as compared to 70 mm) and provide a factor of 4 increase in total solid angle of detection as compared to D4b's usual configuration, amounting to a factor of 5 increase in total counting rate for typical scans due to reduced detector movement time for D4c. For $2\theta \lesssim 50°$, the increase in counting rate over D4b is in fact a factor of about 10.

A D4 sample of average dimensions ($\phi = 7$ mm, height = 50 mm), density (0.1 atom/Å^3) and cross-section (2 barns/atom) is a 10% scatterer and produces 200 cps per D4c detector cell for an incident neutron wavelength of $\lambda = 0.7$ Å (using the hot source), leading to a diffractogramme with an average statistical precision of 0.1% (per 0.125°) after only 3 h of counting.

Each of the 9 microstrip detectors has an angular range of 8° in 2θ, which is slightly larger than half the 15° fixed angular separation between detectors, allowing the execution of a complete 135.5° scan with sufficient overlap via a 7.5° displacement of the 9-detector ensemble on an air-suspension (Tanzboden) surface. This reasonably quick two-step scan technique allows real-time diffraction studies to be performed at D4c. The minimum and maximum scattering angles for typical scans are about 1.5° and 137°. A special scan on a vanadium sample is used to determine the efficiencies of all $9 \times 64 = 576$ detector cells with respect to each other.

1 Detectors

The active element of each D4c microstrip detector is an electrically conducting ''black glass" (Schott S8900) substrate supporting the chromium metallization of the 64 microstrip detection cells, each 2.5 mm wide and consisting of 2 anodes and 2 cathodes. A depth of 30 mm for the ^{3}He detection gas at 15 bar provides a detection efficiency of about 90% at 0.7 Å and 69% at 0.35 Å neutron wavelength. Electrically polarized guard plates at the four sides of the microstrip plate assure a uniform drift field and thus an undistorted diffractogramme.

The mechanical stability and other advantages of the microstrip technology [1, 2] permit a greatly enhanced counting stability for D4c's detectors as compared to the multiwire technology of D4b. Thorough tests first verified the proper functioning of the D4c prototype detector and its associated electronics (Nov 1997 [3]), and then finalized the designs for detector collimation, beamstops and neutron shielding (February/March 1998 [4]). A detector counting rate stability of 2×10^{-4} over nearly 3 d was measured for the D4c prototype detector as compared to only 10^{-3} for a D4b detector [5, 6]. The same stability was later measured for the 9 D4c detectors during the May/June 2000 commissioning tests. The factor of 5 increase in total counting rate for D4c is thus matched by a factor of 5 increase in detector stability, as compared to D4b.

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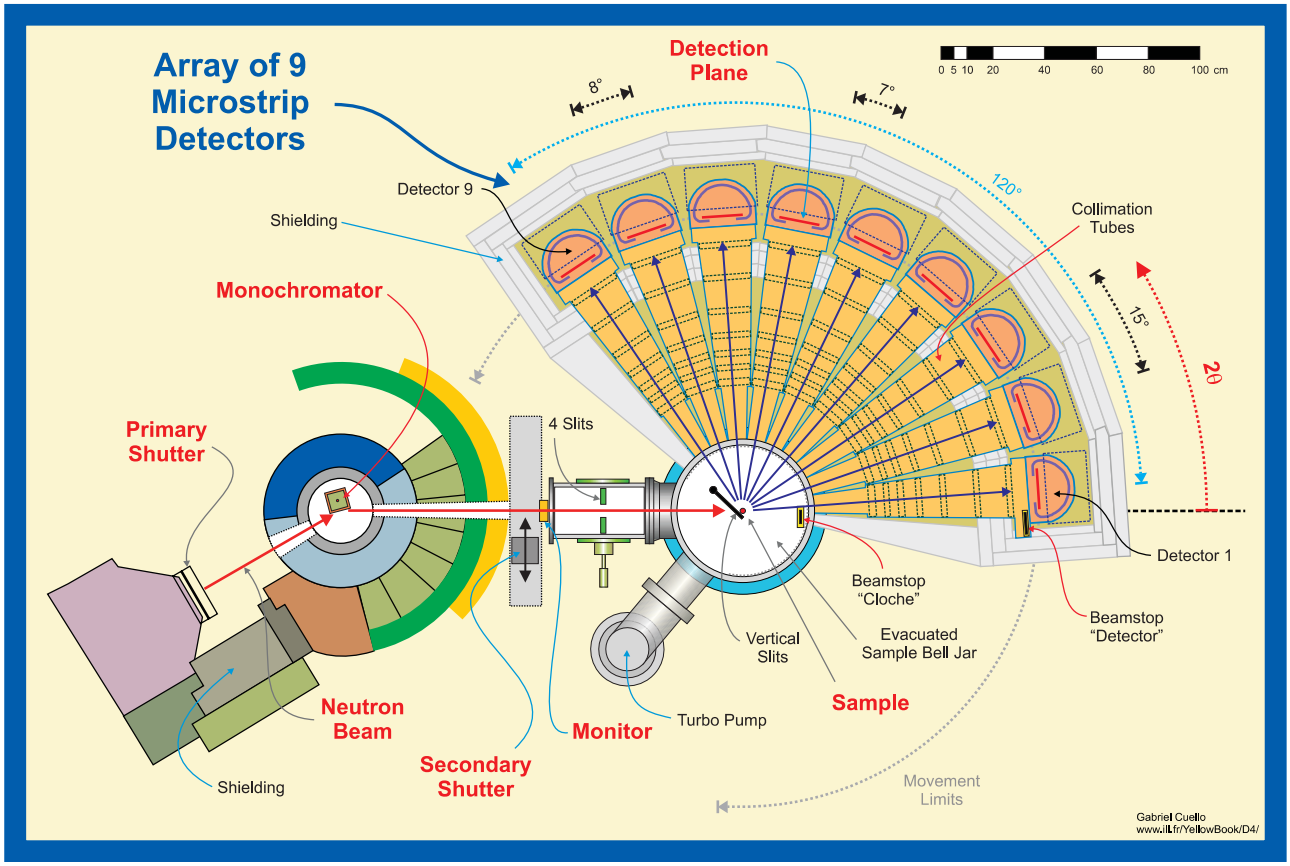

Fig. 1. Top-view schematic of D4c showing the arrangement of the 9 microstrip detectors and their collimation tubes

At the time of writing, the 9 D4c detectors have been under high tension (anode-cathode) for about 2 yr and exposed to 0.7 Å neutrons over a period of more than 1 yr (although not continuously). There are as yet no signs of intrinsic long-term instability in the D4c detector cell efficiencies.

2 Collimation and shielding

The D4c detector collimation design assures that any point on the detection surface sees the solid angle subtended by the sample volume to be surrounded by neutron-absorbing B_4C plates placed ring-like within the evacuated collimation tubes of each detector. A series of collimation rings is the most effective way to minimize the (partially neutron-diffusing) collimation surface area that is visible both to the detectors and to the sample or direct beam, thereby reducing background counts to a minimum. To further reduce the background from direct-beam scattering at the lowest angles, the first detector's collimation uses isotopically enriched $^{10}B_4C$ (neutron absorption constant of 160 cm^{-1} for $\lambda = 0.7$ Å) and includes a mobile beamstop ('detector') of $^{10}B_4C$ just before the detector. A second mobile $^{10}B_4C$ beamstop ('cloche') is placed further upstream in the sample bell jar, cutting off about half of the direct beam. This collimation+beamstops geometry prevents direct beam and parasitic peaks (e.g. from the aluminium bell jar window) from striking any of the 9 detectors at both scan positions and for all three standard wavelengths used at D4 (0.7, 0.5 and 0.35 Å).

The neutron shielding is effected by 100 mm of thermalising polyethylene, followed by 10 mm of borated polymer, surrounding the 9-detector ensemble. The addition of a Cd sheet would be superfluous, since tests [4] showed the remaining very small background counting rate to result principally from non-thermalized neutrons.

3 Results of a recent D4c experiment

As only 0.7-Å wavelength neutrons were available when D4c was commissioned in May/June 2000, further tests at 0.5 and 0.35 Å will be made once the new ILL hot source is installed. On-line software at D4c now provides for data normalization and regrouping into diffractogrammes, the calibration of detector cell efficiencies, the monitor's deadtime correction, and most other aspects of data reduction and analysis.

A recent D4c experiment on the solvation structure of aqueous $CaCl_2$ (6.4 molal) at ambient temperature used the technique of second-difference isotopic substitution ($^{44}Ca/^{nat}Ca$ and H/D). Figure 2 shows the obtained partial structure factors, $F(Q)$, and corresponding partial pair-distribution functions, $G(r)$, for Ca–H and Ca–X (where X represents an atom other than H or D). The smooth counting statistics of the second-difference $F(Q)$ data is impressive considering that these curves amount to less than 1% of the intensity of the total diffractogrammes, and since the incident neutron flux at 0.7 Å is only 30% of what it would be with the hot source. The $G(r)$ functions for Ca–H and Ca–X are pro-

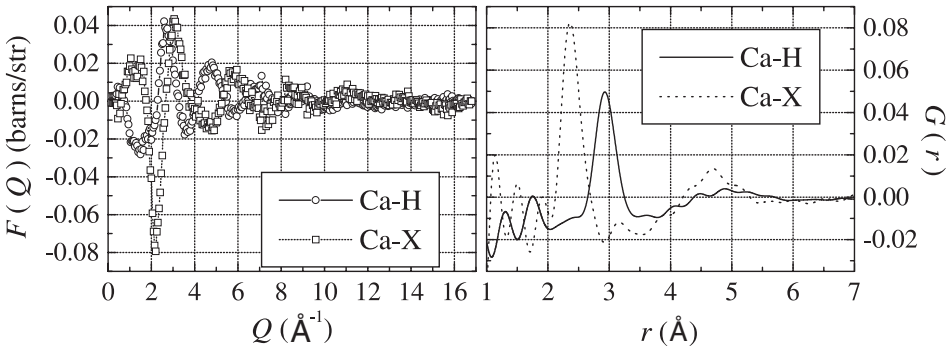

Fig. 2. Partial structure factors, $F(Q)$, and partial pair-distribution functions, $G(r)$, for 6.4 molal aqueous $CaCl_2$

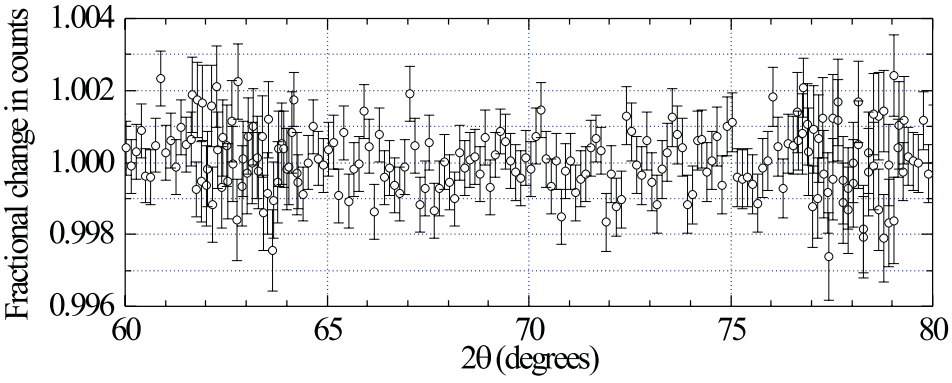

Fig. 3. Stability of successive 5-h scans when counting on a 6.4 molal solution of $^{44}CaCl_2$ in H_2O at ambient temperature

portional to the probability of finding a hydrogen or X atom at a distance, r, from a Ca^{+2} ion, respectively. Contributions to not only the first (between 2.0 and 3.5 Å) but also the second (between 4.0 and 5.5 Å) hydration shells of Ca^{+2} are clearly resolved in the second difference $G(r)$ results.

Figure 3 attests to the stability of the D4c detectors as well as to that of the sample environment for this experiment: For a total counting time of 10 h on a sample of $^{44}CaCl_2$ in H_2O, the second 5 h of the acquisition are divided by the first (here showing $60° < 2\theta < 80°$). It is clear that there is no systematic deviation of the data points (separated by 0.125°) from a value of unity, i.e. the scatter in the data is simply that given by the counting-statistics error bars of less than 10^{-3} (the variation in error-bar size resulting from the particular scan used in the experiment).

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