Liquid Diffraction

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1 Introduction

Neutron diffraction as a technique for structural studies of liquid and amorphous materials is now well established and one of the major advances in recent years has been the availability of instruments using short wavelengths whether on a reactor hot source, such as D4 at ILL, or a pulsed source, such as the Total Scattering Spectrometer (TSS) at the Harwell linac. For the SNS, two instruments for liquid studies have been proposed. The first, the Liquid and Amorphous Diffractometer (LAD) is a standard general purpose diffractometer based on the experience of TSS on the linac and so concentrates on the high Q region. The existing short wavelength instruments have extended the Q range to higher Q but there has been no corresponding improvement in the low Q region - D4 has a minimum Q of $\sim$ 0.2 $Å^{-1}$. This low Q region is accessible using existing low Q or small angle instruments, but it requires the use of long wavelengths (λ > 6 Å) neutrons. Furthermore, the small angle instrument may not be able to measure in the Q range corresponding to the main peak in the structure factor and so several instruments need to be used to obtain the full Q range. The second proposed instrument, the Small Angle Diffractometer for Amorphous and Liquid Studies (SANDALS) aims to fill this gap in the available Q range.

The LAD spectrometer will be built during 1981 and installed on the Harwell linac. Operating experience will then be gained before it is transferred to the SNS.

2 Inelastic Corrections

The accurate determination of the structure of liquids and amorphous solids depends on the understanding of the inelasticity or Placzek corrections. The corrections for time-of-flight diffraction have been discussed by several authors[2 - 5] and the conclusion has been that the corrections are easier to calculate and the useful range of neutron wavelengths is increased if the moderator is cooled rather than at ambient temperature.

Mildner et al[6] have measured the spectrum of three types of moderator - ambient temperature (300K), poisoned ambient, cold (77K) - and have fitted simple functions to the spectra. These results have now been combined with the formalism of Powles[5] to give a more versatile method of calculating the Placzek corrections[7].

The magnitude of the correction depends on the flight path ratio R, defined as $\ell_2/(\ell_1 + \ell_2)$ where ℓ_1 and ℓ_2 are the primary and secondary flight paths respectively. Powles showed that for the self-scattering term for a molecular fluid a small ($\sim$ 0.1) value of R is necessary whereas for the interference scattering a value of R = 1 is required. Other criteria, such as resolution, intensity and space available for the instrument will also influence the choice of R.

Using a cold moderator, the 1/E region is extended and the useful range of Q (ie the region where the correction is almost linear) is increased. Figure 1 shows the self-scattering correction for nitrogen gas at 77K for 2 types of moderator and the gain in Q for the cold moderator is obvious. The overlap of the useful Q range between angles is also improved and, as an extra bonus, the neutron pulse wddth on a cold moderator is narrower, thus improving the Q resolution.

3 LAD

The proposed instrument[8] (Figure 2) has path lengths ℓ_1 = 9 m and ℓ_2 = 1 m, thus giving R = 0.1. A cold (eg 77K) moderator has been specified, so for most use an energy range of 0.082 eV < E < 2.0 eV corresponding to 0.2 Å < λ < 1.0 Å is presently considered suitable. The maximum wavelength that can be used is determined by the frame overlap condition and is $\sim$ 8 Å. A maximum of 1.0 Å is considered more suitable because of the corrections. The minimum wavelength depends on the efficiency of the detectors; for example a 4 atm He^3 counter has an efficiency of only 15% at 0.2 Å compared with 50% at 1 Å. Thus while we refer to the range 0.2 Å < λ < 1 Å, the instrument could use wavelength less than 0.2 Å and up to 8 Å.

In order to obtain a large range of Q, a series of angles is chosen so that there is overlap in Q-range between adjacent detectors. The values chosen are 5°, 10°, 20°, 25°, 58°, 90° and 150°. At 150° there is a bank of detectors in the Q-focussing position[9], while all other angles have single detectors. The detector aperture for the 90° and 150° angles is 0.025 m wide by 0.3 m high whereas all the other angles have apertures 0.005 m wide lying on the Debye Scherrer cone.

The area of moderator viewed will be 0.1 m x 0.1 m and the maximum sample size 0.05 m x 0.05 m. The Q range and resolution for the various angles are given in table 1, where the resolution has been calculated for a sample 0.005 m in diameter and 0.025 m high. The detectors are 5 atm, 25 mm diameter He^3 counters and the count rates have been estimated as $7 \times 10^4 s^{-1}$ at $\theta = 150^o$ decreasing to $5 \times 10^3 s^{-1}$ at $\theta = 5^o$.

4 SANDALS

The proposed instrument is designed for structural studies in the Q range from 0.05 $Å^{-1}$ to 10 $Å^{-1}$ with the possibility of limited data up to 50 $Å^{-1}$. The instrument characteristics are similar to those of a conventional small angle instrument but with a different wavelength range. The maximum wavelength is restricted to 2 Å because inelasticity is difficult to correct for at longer wavelengths. The longer wavelengths will be removed with a gadolinium foil filter. With this maximum wavelength, the maximum possible length of the instrument, defined by the frame overlap condition, is 40 m.

The layout of the proposed instrument is shown in Figure 3. The main detector is a one metre diameter scintillation detector positioned 7 m from the sample. A series of rings 1 cm wide allow scattering angles in the range 0.5^o to 4^o. Two more sets of rings are positioned at 4 m and 1.6 m from the sample to give scattering angles of 5^o and 10^o respectively. All these detectors are contained in an evacuated vessel and it is proposed to utilise vacuum tanks from the Nimrod particle physics programme. A further set of detectors, 5 mm wide by 30 cm high, is situated at 1 m from the sample at angles of 30^o, 60^o and 90^o. The flight paths for these detectors and the sample area are in one vacuum tank which is connected to the main detector tank via a large vacuum valve so that the sample tank can be let up to air independently of the detector tank. The detectors will be of Li^6 glass - a 2 mm thick scintillator has an efficiency comparable to that of an 8 mm diam 40 atm He counter. The rings will be segmented - the lowest angles with four segments, higher angles in mulitiples of four with 64 at 4^o to enable measurements of antisotropic scattering.

The sample position will be at 14 m from a cooled (eg 77 K) moderator and collimation of the incident beam will be by two variable apertures, the first in the bulk shielding after the beam shutter and the second at the entrance to the sample tank. Sample sizes will depend on the resolution required, but will be of the order of 1 cm square.

The resolution has been chosen so that it is comparable with liquids diffractometers such as LAD in the region of the main peak of the structure factor; it becomes progressively worse at lower Q values. The Q ranges and resolutions for the detectors are given in Table 2. Estimates of the count rate for a whole ring range from $\sim 10^3 s^{-1}$ at $\theta = 4^o$ to $\sim 10^2 s^{-1}$ at $\theta = 0.5^o$. The count rate for this 4^o ring is comparable to that of the 5^o detector on LAD.

References

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TABLE 1

The Q ranges and resolution (0.2 < λ < 1.0 Å) for LAD

θ (o)	ΔQ/Q	Q_{min} ($Å^{-1}$)	Q_{max} ($Å^{-1}$)	Detector Aperture
5	0.12	0.5	2.7	) 5 mm
10	0.06	1.1	5	) on
20	0.03	2.2	11	) Debye-Scherrer
35	0.017	3.8	19	) cone
58	0.010	6.1	30	
90	0.006	8.9	44	) 25 mm x 300 mm
150	0.004	12.1	61	

TABLE 2

The Q ranges and resolution for SANDALS

θ	Q_{min} (λ = 2.0)	Q_{max} (λ = 0.1)	ΔQ/Q
0.5	0.025	0.55	0.11
4	0.22	4.4	0.015
5	0.28	5.5	0.015
10	0.55	11.1	0.015
30	1.6	32	0.012
60	3.1	63	0.010
90	4.4	89	0.006

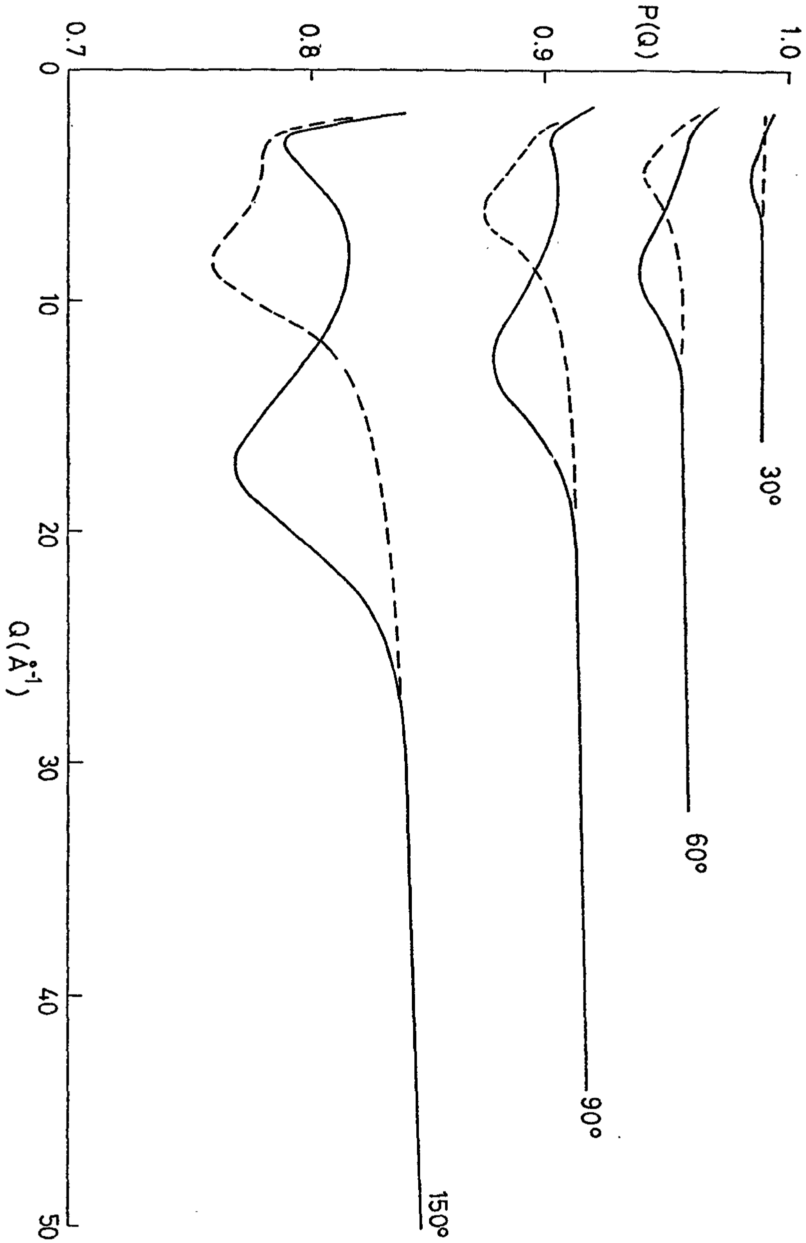

FIGURE 1 Placzek corrections for various detector angles and 2 types of moderator. (— ambient (300K), --- cold (77K)).

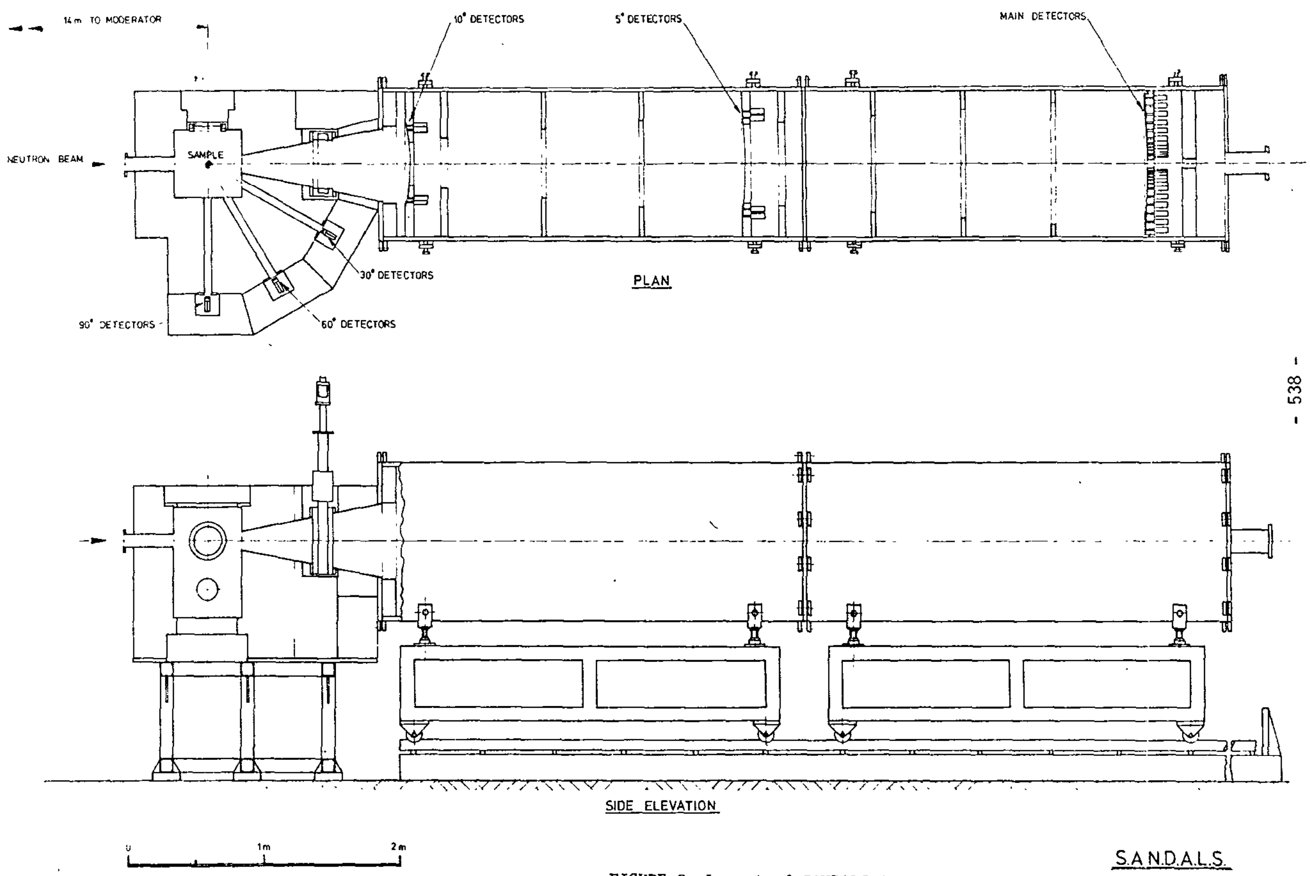

FIGURE 3 Layout of SANDALS instrument.

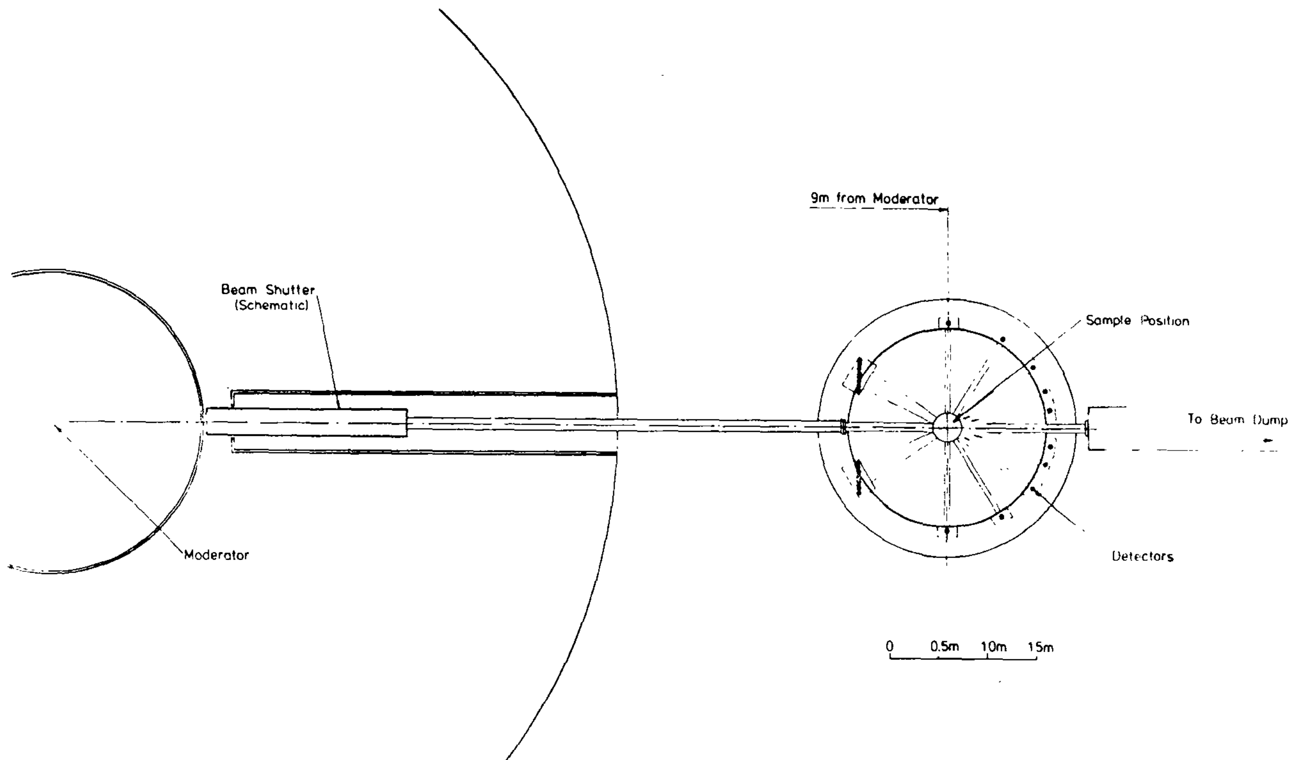

FIGURE 2 Layout of LAD instrument.