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LAD, 1982–1998: the first ISIS diffractometer

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Abstract. The Liquids and Amorphous Diffractometer, LAD, was the main diffractometer for studying the structure of disordered materials at the ISIS pulsed neutron source, until its removal at the end of 1998. LAD was first installed at the Harwell Linac neutron source in 1982, making it the first ISIS diffractometer, and was subsequently moved to ISIS prior to the first production of neutrons at the end of 1984. During its lifetime LAD performed many experiments on a wide range of different structural problems, leading to about 280 scientific papers. The seventeen-year history of LAD is discussed, giving particular emphasis to the development of pulsed neutron diffraction, from its early days as a relatively new and untested technique, to its current status as a well-established major international project.

1. Introduction

In a way, the history of the Liquids and Amorphous Diffractometer, LAD, and its eventual transmogrification into GEM (the GEneral Materials Diffractometer), mirrors the history of pulsed neutron scattering itself. In its early days, the pulsed neutron technique was a small-scale national enterprise which was regarded as needing to prove itself, whereas now it is a subject of international collaboration with a bright future in the years ahead.

The LAD diffractometer was designed by Spencer Howells [1] for use on the ISIS spallation neutron source [2] at the Rutherford Appleton Laboratory (see figure 1). However, in order for experimenters to gain early operating experience, LAD was originally used on the Helios electron Linac neutron source [3] at AERE Harwell, and was then moved to ISIS prior to the first production of neutrons there.

In the early years there were doubts as to whether a non-crystalline diffraction experiment on a pulsed source could be successfully performed and analysed. These doubts may have been due in part to the great success of the competitor diffractometer, D4, at the Institut Laue–Langevin (ILL) in Grenoble, France [4]. However, since then LAD and pulsed neutron scattering have steadily proved themselves. About 280 scientific papers containing data from LAD have been published, including a significant number in high-impact journals (see, for example, references [5–8]). The development of the comprehensive ATLAS (Analysis of Time-of-Flight Diffraction Data from Liquid and Amorphous Samples) suite of data analysis software [9, 10] has also been of importance in establishing the viability of pulsed neutron diffraction for studying the structure of non-crystalline materials.

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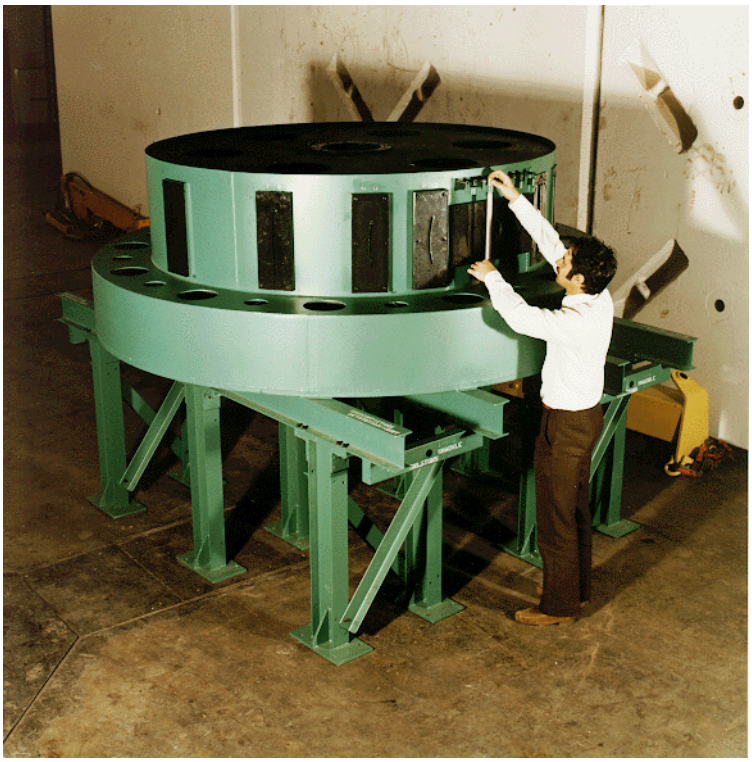
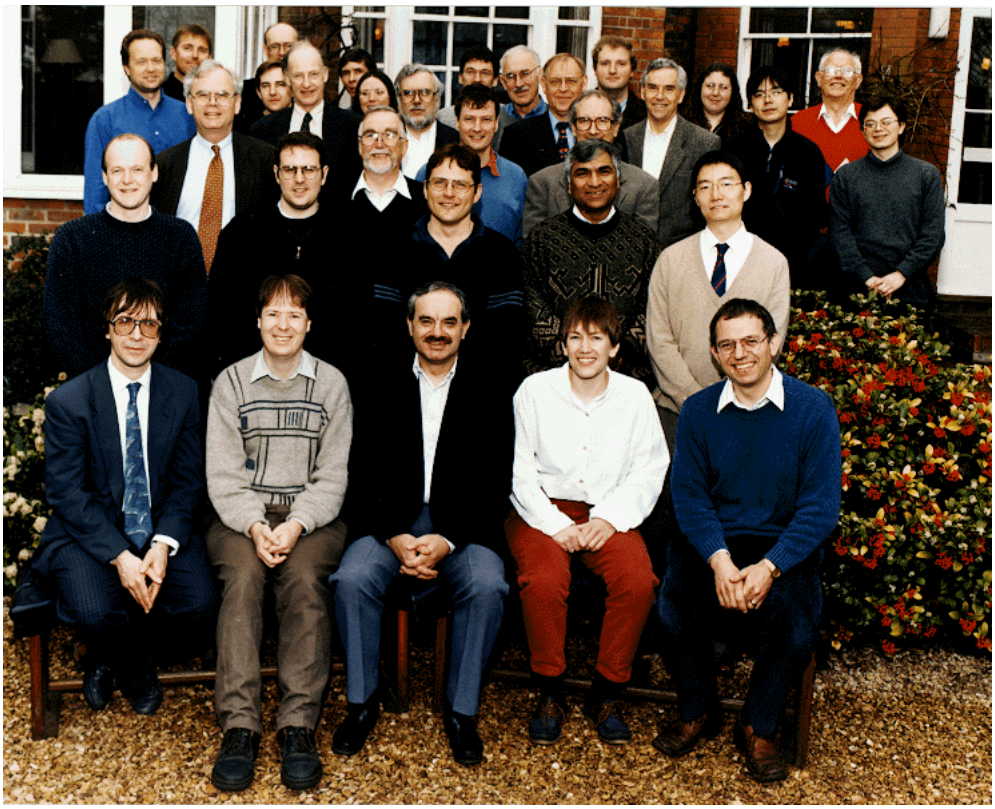


Figure 1. A youthful Spencer Howells checks a ^{3}He gas detector on LAD, prior to installation at ISIS in 1984.

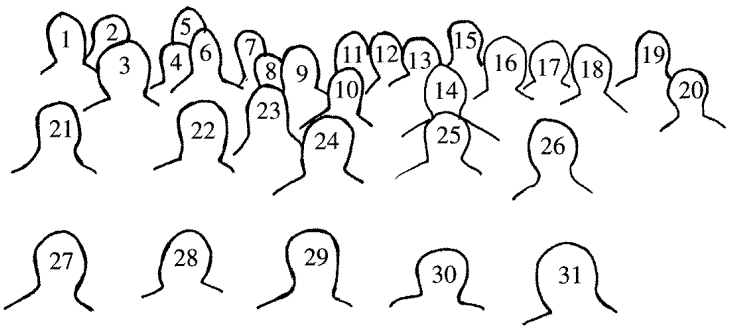
Finally, after a long, 17-year career, LAD is now being replaced by a new GEneral Materials Diffractometer, GEM [4, 11], the design of which draws greatly on the lessons learnt on LAD. In order to celebrate the contribution made to the study of the structure of disordered materials by LAD, a scientific meeting was held in March 1999 at the Cosener's House, Abingdon (see figure 2). Twelve talks and eight posters were presented at the meeting, showing the wide range of materials covered by LAD, including glasses, liquids, disordered crystals, fluids in clays and polymers. The papers from the meeting are published in this issue of *Journal of Physics: Condensed Matter* [12].

2. The Liquids and Amorphous Diffractometer, LAD

Figure 3 shows the final configuration of the Liquids and Amorphous Diffractometer, LAD, used at the ISIS spallation neutron source. The incident neutron beam was obtained from a moderator containing liquid methane at a temperature of 110 K. The sample position was located at a distance of 10 m from the moderator, with a beam at the sample position which was 20 mm wide and 40 mm high. Detector banks were placed in the horizontal plane, on both sides of the instrument, at the scattering angles given in table 1. Originally all of the detectors were 10 atm ^{3}He gas tubes. However, as is described in section 5 below, the 20°, 35°, 60° and 90° detector banks were replaced with lithium-glass scintillator detectors. The neutron wavelength range covered by LAD extended in principle from 0.05 Å to 7.0 Å, although the data collected at the higher wavelengths are not of ultimate use in extracting a structure factor



(a)



1	Lars Börjesson	Chalmers, Sweden	12	George Neilson	Bristol	22	Jonathan Wasse	UCL, London
2	Dennis Engberg	ISIS Facility	13	Neil Cowlam	Sheffield	23	Alan Leadbetter	ILL, Grenoble
3	Gavin Williams	ISIS Facility	14	Bob Newport	Kent	24	Phil Salmon	Bath
4	Matthew Tucker	Cambridge	15	Steve Cooper	Reading	25	Ashok Adya	Dundee
5	Neal Skipper	UCL, London	16	Brian Boland	ISIS	26	Haruaki Matsuura	Dundee
6	John Dore	Kent	17	Cora Stone	Reading	27	Simon Hibble	Reading
7	Robert McGreevy	Studsvik, Sweden	18	Shu Hayama	UCL, London	28	Alex Hannon	ISIS Facility
8	Kathy Johnson	Liverpool	19	Roger Sinclair	Reading	29	Spencer Howells	ISIS Facility
9	Geoff Mitchell	Reading	20	Maxim Shcherbakov	Dundee	30	Jacqui Cole	Kent
10	Adrian Barnes	Bristol	21	Chris Benmore	ISIS Facility	31	Alan Soper	ISIS Facility
11	Dave Keen	ISIS Facility						

(b)

Figure 2. (a) The happy attendees at the meeting *'LAD, 1982–1998: the First ISIS Diffractometer'*. (b) The key to (a).

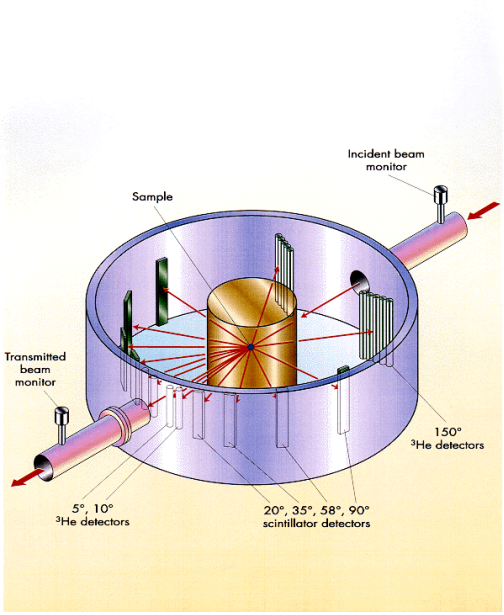


Figure 3. The final configuration of the Liquids and Amorphous Diffractometer, LAD.

Table 1. The final LAD detector bank parameters (incident flight path $L_1 = 10.0$ m).

Bank	Scattering angle 2θ (deg)	Total flight path	Resolution $\Delta Q/Q$ (%)	Detector type
1	5	11.047 m	14	^{3}He gas tube
2	10	11.033 m	7	^{3}He gas tube
3	20	11.040 m	3.5	Li-glass scintillator
4	35	11.043 m	1.8	Li-glass scintillator
5	60	11.046 m	1.2	Li-glass scintillator
6	90	11.039 m	0.8	Li-glass scintillator
7	150	11.128 m	0.6	^{3}He gas tube

due to the effects of the experimental corrections, especially the inelasticity correction.

Spencer Howells acted as instrument scientist for LAD throughout its life, assisting users with experiments. In 1988 he was joined in this role by Alex Hannon (see figure 4).

The design of LAD was similar in its layout to the Total Scattering Spectrometer [3, 13], TSS, at the Harwell Linac, with large quantities of shielding material (not shown in figure 3) between the detector banks. The angular width of the channels in the shielding for the 20°, 35°, 60° and 90° detector banks was less than 2.5°. Thus the LAD detectors only covered a very small solid angle, 0.016 steradians, with the consequence that the count rate of LAD was low and long counting times were required to achieve adequate statistical accuracy. At the time that TSS and LAD were designed, it was believed that it was necessary for pulsed neutron diffractometers to be very well shielded. However, subsequent experience has shown that this belief is not correct, and hence the new GEM diffractometer [4] will have a much more open design with minimal collimation between the detector banks and with the detector banks extending ±45° out of the horizontal plane. In this way, the total solid angle of GEM

Figure 4. Alex Hannon and Spencer Howells with the LAD sample-changer.

will be about 3.4 steradians, and hence a major increase in count rate will be achieved, despite the loss in intensity (by a factor of 3) due to the increase of the incident flight path from 10 m to 17 m.

3. The early days

The ideas for the LAD instrument first appeared in the technical specification for the Spallation Neutron Source (SNS) project [14], as ISIS was then called. In this report there is a design specification for a Total Scattering Spectrometer, but because an instrument called TSS [3, 13] already existed on the Harwell Linac, the instrument was later renamed as LAD. A design group, comprising members of the UK neutron scattering community, was set up and first met in March 1978. As well as discussing the design of LAD, the group were also requested to

look into the possible use of LAD for powder diffraction. Even at this early stage a second non-crystalline diffractometer concentrating on small scattering angles was under discussion.

In July 1977, the Science Planning Group (SPG) of the SNS was established to advise the Neutron Beam Research Committee (NBRC) of the then Science Research Council (SRC) and Rutherford Appleton Laboratory (RAL) on all aspects of exploitation. In October 1978, the RAL reported back to the SPG that the LAD design group had recommended that combining LAD with a high-intensity powder diffractometer was not desirable.

In April 1979, the SPG submitted a paper to the NBRC seeking approval for the construction of seven instruments, as detailed in table 2. There was also a proposal to construct LAD early and install it on the Harwell Linac in order to relieve the high demand for the use of TSS.

Table 2. The original plan for the initial ISIS instrument suite.

Acronym	Instrument
LAD	Liquids and amorphous materials diffractometer
HIPD	High-intensity powder diffractometer
HRPD	High-resolution powder diffractometer
LOQ	Low-Q diffractometer
HET	High-energy-transfer spectrometer
HTIS	High-throughput inelastic spectrometer
IRIS	High-resolution quasielastic spectrometer

By October 1978, the early ideas of a second non-crystalline diffractometer had become firmer with a proposal for the Small-Angle Neutron Diffractometer and the outline design for this was approved. In January 1980, a modified design, now referred to as SANDALS (Small-Angle Neutron Diffractometer for Amorphous and Liquid Samples), was presented and the emphasis was now on low scattering angle and high-energy neutrons to reduce inelastic corrections.

In October 1980, the SPG defined the first group of instruments to be installed at ISIS as LAD, HRPD, LOQ, HET, HTIS, IRIS, and a polarized neutron development beamline, POLARIS. It was also decided to construct both LAD and HTIS early and install them on the Harwell Linac in order to gain experience of pulsed source neutrons. LAD was then installed on the Linac during 1982.

4. Experience on the Harwell Linac

LAD performed its first scheduled experiment in June 1982 and remained operational on the Harwell Linac until March 1984. A wide range of samples was investigated, including glasses, metallic glasses, molten salts and aqueous solutions. These provided much valuable experience, especially in data analysis and sample environment.

Two of the main concerns at that time were the effects of nuclear absorption resonances and reciprocal-space resolution. One of the advantages of a pulsed source diffractometer is the use of high neutron energies (in the eV range) to provide high values of the momentum transfer, Q; unfortunately this is the energy region where many of the resonances occur. The data in the region of a resonance cannot be corrected sufficiently well and must be removed, thus leaving gaps in the useful Q-range. The occurrence of nuclear resonances becomes more common with increasing atomic number, so the concern at the time was that experiments using the heavier elements (which also have lower inelastic corrections) would be compromised. Fortunately, over the succeeding years, experience has shown this concern to have been over-emphasized.

The design of LAD (and TSS) is such that the Q-resolution varies with angle and deteriorates with decreasing angle. Thus it is difficult to merge the data from the different angles if the resolution is so poor that the peak widths are markedly different. This was particularly true of TSS and a method of correcting for resolution was proposed by Howells [15] and first used by Gardner [16]. The design of LAD is such that the resolution is much better than that of TSS, so again these concerns have been shown to be exaggerated.

During these early years the first steps were taken to provide a suite of data analysis programs. Existing programs for analysing data from reactor-based instruments were modified for use on instruments on the Intense Pulsed Neutron Source (IPNS) at the Argonne National Laboratory (ANL), USA, and then brought to the RAL for conversion for use on LAD. This early package was then further modified and improved, ultimately to become the ATLAS package [9, 10].

5. ISIS operation

LAD was one of six instruments installed at SNS for the first neutrons delivered in December 1984. For the short time of running, the sample chosen was a high-density nickel powder standard so that the resolution characteristics could be determined. Neutron absorption foils were also placed in the beam for calibration [17, 18].

Over the next few years, the available neutron beam was used first of all to run calibrations. Then, well-known samples, which had already been studied on reactor instruments, were measured in order to compare results. New experiments were initially chosen by the design group and the first round of the formal ISIS experimental selection procedure took place in 1988.

During the early 1990s, LAD was used for testing scintillator detectors. This had a dual purpose: firstly, to increase the detector area and hence to improve the counting statistics; and secondly, as part of a development programme for detectors suitable for SANDALS. The first change involved the installation of one scintillator module at 90° which could be directly compared with the gas detector diametrically opposite. This proved successful, and so banks of scintillators were installed to cover the 20°, 35°, 60° and 90° scattering angles on one side of the instrument to be compared with the gas detectors on the other side. When these were shown to be satisfactory, a similar bank was installed on the other side. The experience was that the scintillators had poorer stability than gas detectors and that this particular form, using Li-doped glass, is too sensitive to gamma rays. This latter problem is particularly acute when the sample contains a neutron-absorbing isotope which emits a gamma ray when a neutron is absorbed [19]. In some cases the gamma ray is prompt, leading to the observation in the scintillator detector data of a peak which occurs before the trough due to the neutron absorption resonance. In this case the gamma sensitivity of the detector leads to an extension of the range in the data which is compromised by the nuclear resonance. In other cases the gamma ray is delayed, leading to an approximately time-independent background in the scintillator detector data which results in a large background in the structure factor data at low Q.

The shutter was closed on LAD for the last time in December 1998—fourteen years and one day after its first ISIS neutrons (see figures 5 and 6).

6. Novel techniques

This section highlights some of the unique features of LAD and some of the novel techniques employed in experiments on LAD.

Figure 5. Spencer Howells closes the LAD shutter for the final time, watched closely by Alex Hannon.

6.1. High Q-resolution

Although designated as a diffractometer for non-crystalline materials where a resolution of $\Delta Q/Q$ $(=\Delta d/d)$ of about 2% is typical, the backward-angle (scattering angle $2\theta = 150^\circ$) detectors on LAD had a resolution of about 0.6% which is comparable to what is achieved by good powder diffractometers. Indeed, for several periods LAD was scheduled as a powder diffractometer—usually when the other ISIS medium-resolution diffractometer, POLARIS, was not available due to upgrading.

In addition, this advantage of high resolution was used in a variety of experiments for disordered materials. A routine, but important, use was in checking the quality of amorphous and glassy materials—here the good resolution was sufficient to clearly reveal Bragg peaks, which would immediately show that the sample was not fully amorphous. Such problems occurred, for example, in amorphous ice and metallic glasses. In the latter case, Bragg peaks would be visible at high resolution, but at poorer resolution the diffraction pattern would look typically amorphous. A service was provided for users where test samples could be checked in advance of a scheduled experiment, so that such problems could be identified and rectified, thus making better use of the neutron beamtime.

Yet another case for good Q-resolution is a system in which the diffraction pattern is a combination of an amorphous-like structure factor, $S(Q)$, and Bragg peaks: an example is the pattern from solid sulphur, where there are Bragg peaks at low Q due to the crystal structure, and oscillations at high Q from the nearest-neighbour distance [20]. More recently a trend has

Figure 6. A mature Spencer Howells resting after removing LAD from ISIS in 1999.

developed of analysing powder diffraction by calculating the structure factor, $S(Q)$, and the pair correlation function, $g(r)$, as well as using the traditional method of profile refinement of the Bragg peaks (see for example references [21] and [22]).

6.2. Low-angle diffraction

Low scattering angles are essential for studying the structure of disordered materials because they provide the low-Q region of $S(Q)$, albeit at poor resolution—the lowest two angles on LAD had values of $\Delta Q/Q$ of 7% and 14% respectively. In certain circumstances this was also of use in powder diffraction: for example, during the early days of high-T_c superconductors, samples would arrive for characterization on HRPD and sometimes on LAD where Bragg peaks with d-spacings of up to 50 Å could be measured. The low-angle, low-Q region is also where the magnetic form factor is largest, so these detectors were used to study magnetic structures.

6.3. Aligned samples

With its fixed, well-shielded angles, LAD was ideal for investigating aligned samples. The best case occurs when the detectors at $\pm 90^\circ$ are used to simultaneously measure the structure factor perpendicular and parallel to the plane of a flat sample. The first such experiment on LAD was on a thin-film polymer [23] and the technique was later used on metallic glass ribbons [24].

Lower angles can also be used for this technique, but then the sample has to be rotated to obtain the two alignments. This version of the technique has been used extensively on layered structures such as clays [25]. These experiments on clays, at high temperature and pressure to mimic conditions underground, have brought a new unit of measurement into neutron scattering—km deep!

6.4. Sample environment

The detector shielding on LAD also minimized the background scattering from sample environment equipment, especially at scattering angles of 90° and 60°. This led to the use of novel equipment, such as an autoclave to measure molten alkali metal alloys at high temperature and pressure—not only technically difficult but subject to considerable safety assessment! The first such experiments performed on LAD were on KPb up to 1873 K and 60 bar and NaSn up to 1673 K and 50 bar [26].

Using standard furnaces, more recent experiments have looked at the recrystallization of metallic glasses *in situ*—this involved many short runs of the order of a few minutes [27].

6.5. Data analysis

During the years, the increasing complexity of the samples studied on LAD has required the development of new methods of analysing and interpreting the data. One such technique is the combined use of neutron scattering and molecular dynamics to unravel the liquid structure of dichlorodifluoromethane [28]. Another tool increasingly used is the method referred to as reverse Monte Carlo (RMC) modelling [29]. Both of these techniques are useful for multi-component samples where there is only one stable isotope or there are insufficient isotopes to use isotope substitution as a method for determining the partial structure factors. In these systems, the best method then is to use a combination of several techniques—such as, for example, neutron diffraction, x-ray diffraction and EXAFS [7].

7. International participation

The design specification for LAD was produced by the UK neutron scattering community, and reflected the considerable activity in this field already present in the UK. From its early days, however, ISIS encouraged participation from overseas scientists and from the outset there were non-UK users of LAD. Many overseas users started either their first ISIS experiment or even their first ever neutron experiment at LAD before moving on to more general use of ISIS and other neutron facilities.

France already had a thriving neutron scattering community, through its use of ILL and its own reactor at CEA-Saclay, and scientists from Saclay took an early interest in using LAD and in the SANDALS specification, with their first publication appearing in 1989 [30].

Sweden, in contrast, had a small neutron community based at the Studsvik reactor, and they too became early users of LAD. As a result of that use—the first Swedish publication appeared in 1988 [31]—they were successful in obtaining a grant from the Swedish Research Council to finance the detectors for SANDALS. Wider use of ISIS then increased, with Sweden becoming an International Partner, and more recently becoming involved in the construction of the OSIRIS spectrometer.

Spain had no history of home-based neutron scattering and the first links were with CSIC in Madrid, partly financed by British Council grants. Again the first steps on LAD—with the first Spanish publication in 1990 [32]—led to a wider use of ISIS and other Spanish users were then mobilized to formalize involvement with the new OSIRIS instrument.

The Netherlands has a background of neutron scattering, primarily based at the Delft reactor, and they were soon users of LAD. The first Dutch LAD publication appeared in 1989 [33] and the Netherlands then became an International Partner of ISIS.

Italy too had some neutron scattering facilities and quickly become involved with ISIS. The first LAD publications from Italy appeared in 1989 [34, 35]. Italy is also an International

Partner, and more recently became involved in upgrades of the TFXA/TOSCA and eVS spectrometers.

Neutron facilities in the USA have had long-standing links with ISIS. The US experience of pulsed sources, such as IPNS at Argonne and LANSCE at Los Alamos, was extremely valuable at ISIS in the design phase. ISIS staff gained experience at these sources before ISIS started operation and there has been a continuous interchange of information. Scientists from ANL have been regular users of LAD, and LAD and UK scientists were involved in the design of GLAD, the Glass, Liquid and Amorphous Materials Diffractometer, at IPNS. More recently ISIS staff and LAD users have participated in the group designing the Liquids and Amorphous Diffractometer for the planned new US Spallation Neutron Source. For the first author this was definitely a situation of *déjà vu*, in that he started off his career on pulsed neutron sources designing LAD for SNS (as ISIS was then called), and some 20 years later became involved in designing a 'LAD' for SNS (but now the USA version).

8. Statistics

Finally, this section gives a brief resumé of the statistics of LAD usage. From the first scheduling round in 1988 to the last in 1998, 536 proposals were submitted, of which 388 were successful. The number of days of neutron beamtime requested was 2729 and 1483 days were allocated. All of these scattered neutrons were converted into about 280 publications.

9. The future

LAD may have gone, but the future for diffraction from disordered materials at ISIS is assured. For many years now, SANDALS [36] has operated in parallel with LAD and has specialized in hydrogenous materials. The LAD beamline will soon be occupied by the GEM diffractometer. This is a combined disordered materials and powder diffractometer—so after over 20 years the requested merging of the two instruments has finally happened! GEM has all the advantages of LAD but with a count rate which will be raised by a factor of 70, so we can look forward with eager anticipation to lots of new scientific results.

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