A new D11 Detector and Tank: the Scientific Case

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Executive Summary

Following the renewal of the collimation system between 2002 and 2004, it is now the detection unit of D11 that constitutes the limiting part of the instrument, in particular with respect to dynamic range, range of momentum transfer, spatial resolution, efficiency and dead time.

The project of developing a new SANS detector for D11 started in 2005, in the framework of the ILL's Millennium Program. The new detector will have a larger counting area of ≈ 100x100 cm^2, thus increasing the dynamic range from 7 to 15. The device will be installed in a larger tank (diameter ≥ 2m), with an extended length of ≈ 42m, thus allowing the extension of the available range of momentum transfer from 0.82 $Å^{-1}$ down to values as low as 3 10^{-4} $Å^{-1}$. In addition to the capability for faster counting, the new detector will have an improved spatial resolution and an increased efficiency.

1. The instrument D11: low Q for soft matter and biology

D11 (Figure 1) is the archetype of a long pinhole geometry instrument, designed for the small angle neutron scattering studies of large scale structures in chemistry, biology, material science and solid state physics. With its unique range of momentum transfer at lowest background and highest flux, it is the world's leading instrument of this kind.

D11 is in operation since 1972. With the continuing revolution in major components such as the detector & tank, the velocity selector, the collimating system together with the development of specialized sample environment, the instrument has maintained its foremost position in the field over more than 3 decades.

Recent statistics of use between 1999 and 2004 show that 55% of the experiments are in the field of soft matter research and 17% in the field of biology. About 75 different experiments are performed each year, with an average duration of about 2 days per user group. The "overload factor", i.e. the ratio between the number of requested beam time to available beam time is about 2. The scientific output of the instrument is on average 30 publications per year.

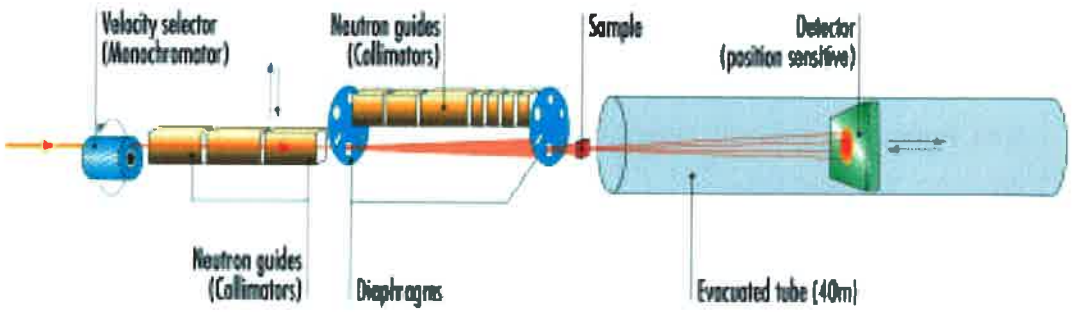

Fig. 1: Schematic drawing of D11. It shows the general setup and is not meant to render all single components.

2. Small angle scattering at D11: versatility for our users

A SANS experiment at D11 reveals the structure of the sample from analyzing the normalized intensity I, scattered from the sample at small angles and recorded on the position-sensitive area detector (which is placed perpendicular with respect to the beam axis), as a function of the momentum transfer Q. I=f(Q) is called the "scattering curve" and contains the complete information about the sample size and shape, i.e. the structure on a nanometre scale. The momentum transfer Q has the unit [$Å^{-1}$] and is defined as:

$$Q = 4\pi/\lambda \sin(\theta/2) = 4\pi/\lambda \sin[0.5 \arctan(r/L)]$$

λ is the neutron wavelength, r is the modulus of the radius vector starting from the beam centre on the detector and L is the sample-to-detector distance.

For a given detector size (minimum radius r_{min}, maximum radius r_{max}) the 2 tunable parameters of an experiment are the neutron wavelength λ and the sample-to-detector distance L. λ and L therefore define the window of momentum transfer $Q_{min} - Q_{max}$ in which the structure of the sample can be investigated. The ratio Q_{max} / Q_{min} for a given setting of λ and L is defined as the dynamic range of the instrument; for D11 with its present detector (r_{max}=36cm) it is 7. Q is the relevant size parameter in reciprocal space and roughly speaking inversely proportional to a characteristic distance, for instance the size of a polymer molecule or the distance between two colloidal particles. In other words, a SANS instrument is to be considered as a "microscope in reciprocal space", with the focal length of the lens being adjusted with the choice of Q.

For practical reasons, the appropriate Q-window $Q_{min} - Q_{max}$ for the sample is tuned with the sample-to-detector distance L and the neutron wavelength λ is kept fixed during the experiment. The collimation distance (i.e. the distance between the exit of the collimating guide and the sample position) is normally "symmetric", i.e. set equal to the sample-to-detector distance. Because of the restriction in dynamic range Q_{max} / Q_{min} for a given setting with the present detector, and in order to cover a Q window as largely as possible, almost all experiments at D11 use more than one sample-to-detector distance.

3. What do our users need in future?

A new trend in soft matter science and biology is the increasing interest in so-called "smart materials", i.e. nano-structures with possible innovative applications. Examples are for instance polyelectrolyte-based microcapsules, which form monodisperse nano-containers. These structures are gaining importance in applications like encapsulation/ release of agents in fields as diverse as medicine, food design, cosmetics, printing. Other examples are block-copolymer based thermosensitive core-shell microgels. Because of their ability to react on a temperature change with swelling or collapse, they find potential applications in a broad range of areas such as carrier systems for targeted drug delivery and controlled drug release, separation, filters, nano-actuators etc.

In this growing field, more knowledge is necessary in understanding the mechanisms involved in the formation of nano-particles of well defined geometry and the parameters that control their size and shape. SANS at D11 is an important tool for the study of these complex materials on the mesoscopic scale. The method complements other scattering techniques, direct imaging (electron microscopy), NMR and other spectroscopic methods. However, despite recent upgrades, the instrument is today limited with regard to several aspects. In particular the dynamic range, the dead time of the detector, the range of accessible momentum transfer and the spatial resolution of the detector need improvement.

3.a. a higher dynamic range Q_{max} / Q_{min}

About one third of all experiments at D11 require the maximum possible detector distance of at present 36.7m, thus the lowest possible values of momentum transfer Q.

In order to explore the totally available range of momentum transfer of the instrument, down to the minimum detector distance (1.1m), it is currently necessary to perform measurements at 4 different detector distances (1.1m, 2.5m, 10m and 36.7m) for obtaining a sufficient overlap of the "scattering curves" I=f(Q) in each window. There is increasing scientific interest to explore the structure of the sample on the broadest possible length scale and to lower values of the momentum transfer. With an increase in dynamic range of the instrument and together with an improved detector

resolution, it will in future be possible to achieve small angle scattering data with considerably improved quality, with less experimental configurations (2 instead of 4 detector distances) and consequently an enormous time gain.

At present, typical experiments at D11 focus on the elucidation of structures on the nano-metre scale. For instance, when block-copolymers form micellar-type complexes, SANS is a most convenient tool for analyzing the structure of the complex or the behaviour of one component in the complex. This is called a static (or equilibrium) experiment as the final complex is investigated and not the formation process itself. In future, however, it will become more and more important to directly investigate the formation process of nano-structures by real-time (kinetic) experiments. This will reveal new insight in the building mechanisms of matter, and this information will then help, in conjunction with complementary methods, to develop new materials with better characteristics.

One example is water-soluble polymers, such as polyelectrolytes, which can form fascinating shapes like cigars or pearl necklaces in water. The study of the complex phase behaviour allows for a selective shape formation, thus making the tailoring of distinct polymer shapes feasible. The shape of course largely influences the properties in technical processes. It is noteworthy that polyelectrolytes are one of the industrially most often used polymers: e.g. in nappies, sanitary napkins, as waste water treatment agent or as paint disperser just to name a few applications. Interestingly, if a polyelectrolyte forms a pearl necklace structure, the latter fluctuates with respect to the number of pearls. At present, time-resolved experiments with the aim to follow up the dynamics of this process are not feasible because of the limited dynamic range at a given instrument setting. The approach to reveal the dynamics of formation processes implies that one instrument setting must be chosen, i.e. the sample-detector distance cannot be changed. Accordingly, a large Q-window within one setting is of utmost importance.

3.b. a faster detector with a lower characteristic dead time

About 25% of all experiments at D11 measure at short detector distances L (corresponding to high momentum transfer, $Q \geq 0.03$ Å^{-1}), under conditions where the sample scattering is often strongly dominated by a high incoherent background originating from the hydrogen (H) containing solvent of a deuterated (D) solute. The detector consists of a single gas volume, and when a neutron is hitting the detector, the whole detector is blocked for the moment of signal treatment (characteristic dead time). Under the conditions described above, the present D11 detector cannot be used in an optimal way with a short collimation distance setting: the countrate is often exceeding the present limit of 50 kHz and is too high to allow for dead time corrections. In order to reduce the detector countrate in this range of momentum transfer, the collimation distance is usually increased; the aim is here to reduce the neutron flux at the sample position. Thus neutrons are "thrown away" in a range of momentum transfer where the signal-to-noise ratio between sample and solvent is usually bad and where a higher intensity could improve the statistical quality of the data in the time given for the experiment.

As a consequence, long measurement times are required so that the comparably small coherent signal is recorded with an acceptable signal-noise ratio. For poor data, the comparison of experimental data with theoretical fit models for the structure is becoming difficult. It must be stressed that the net intensity of the sample remains unchanged and is not depending on the fact whether the solvent or the sample contains H. It is only the background that changes.

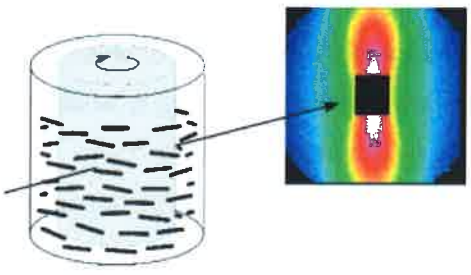

Fig. 2:

Shear thinning and orientation ordering of wormlike micelles
(Förster et al., Phys. Rev. Lett. 94, 017803 (2005))

Non-equilibrium (rheo-SANS) experiments at D11 yield pronounced anisotropic scattering patterns with high countrates on a small detector area.

A very recent example, which illustrates this problem, is the study of polyelectrolyte (PE) mixtures. The behaviour of single PE is quite well understood, but their interactions with other kinds of PE or with proteins (like occurring in food processing) still remain mysterious. The work performed so far was to build up a complex consisting of a hydrogenated PE together with a deuterated PE. It was looked at the complex in a deuterated solvent, where consequently only the hydrogenated PE is visible. The other one is matched and gives no excess scattering. Now, as a second step, it is necessary to look at the same system in a hydrogenated solvent, thus investigating how the PE behaviour of the second deuterated PE changes when a complex is formed. At present, this experiment is not feasible within normal beam time allocation (although contrast matching is the most significant advantage of neutrons in comparison to the increasingly popular x-ray techniques).

Another important aspect, which illustrates the need for a faster detector, are strongly anisotropic scattering patterns on the two dimensional detector, as obtained for instance with non-equilibrium experiments using the D11 shear apparatus ("rheo-SANS"). The study of the shear thinning and orientational ordering of wormlike micelles (see Figure 2) has revealed an anisotropic intensity distribution with a very pronounced high intensity in vertical direction on the detector, due to the macroscopic shear-alignment of the block copolymer micelles in flow direction. As the consequences of detector dead time losses are usually treated as an integral effect, using the total countrate, corrections are problematic when the intensity is not uniformly distributed.

3.c. a larger range of momentum transfer towards lower Q

There is considerable interest to extend the accessible Q window towards lower Q. At present, the limiting Q-value is reached at a sample-detector distance of 36.7 m with a neutron wavelength of 18 A. This yields $Q_{min} = 5\ 10^{-4}$ Å^{-1}. Those experiments, which require measurements at lower Q, usually use either light scattering (LS) or Ultra-SANS (USANS).

It is evident that the combination of scattering curves from SANS, LS or USANS requires a sufficient overlap of the different methods in terms of Q. The so obtained complete form factor can be interpreted in terms of particle size, molecular weight

and shape. At the moment, an overlap between SANS and USANS e.g. is hardly possible. An illustrating example is presented in Figure 3. The exact structure of cellulose fibres, which are used in clothing industry, could not yet be completely revealed. As many decades of particle sizes (from microfibrils down to single fibres and their sub-units) must be taken into account in order to understand material properties like crystallinity and deterioration tendencies, SANS and USANS must be combined.

Fig.3a:
Structure of Lyocell Cellulose Fibres
(E. Jericha et al., D11 experiment 9-11-964)

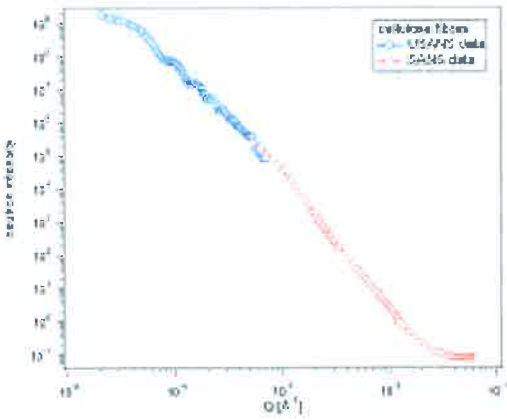

Fig.3b:

For characterizing the fibre structure experiments are done

with SANS at D11 (ILL), in order to see microfibrils
(size range 10nm – 100nm)

with USANS at S18 (ILL), in order to see the macrofibrils (size range 100nm – 10μm)

This is also the only way to interpret the data by comparison with reliable fit models. The presented example shows the actual overlap limits: while the USANS data hint to a further decrease of intensity with increasing Q, the SANS data indicate a plateau in the overlap regime. As the overlap regime only comprises some data points, it is at present impossible to resolve this contradiction.

3.d. a better spatial resolution of the detector

The Q-resolution of the instrument is defined by the aperture sizes, the beam divergence (collimation distance), the wavelength spread $\Delta\lambda/\lambda$ of the velocity selector and the spatial resolution of the detector, i.e. the "cell" size. Recent improvements of the collimation system have, amongst others, introduced diaphragm changers with the possibility of inserting small apertures and the possibility of rotating the selector, leading to a decrease of the bandwidth $\Delta\lambda/\lambda$.

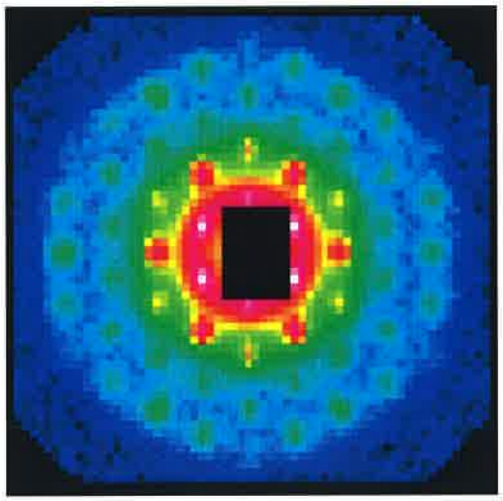

Fig.4:

Rheo-SANS of micellar block copolymer solutions
(Förster et al., D11 experiment 9-10-762, Aug. 2005)

A sudden ordering of the shear thinning micellar phase occurs above a shear rate of 200 s^{-1}, which is not yet understood The resulting scattering pattern of the fcc-structure shows about 50 peaks.

The peaks are not sharp and the blurring of the image illustrates the present limits of instrumental resolution.

Therefore, the detector "cell" size of 10x10mm limits at present the Q-resolution of the instrument. A striking example, which shows the unsatisfying resolution, is the very recent rheo-SANS experiment on micellar block copolymer solutions (see Figure 4). The micellar phase is used for the preparation of mesoporous oxides, which play a role for catalyst support or photonic crystals. The micellar solution is shear thinning and forms a shear-oriented fcc-structure with a strong ordering above a threshold value for the shear gradient of 200 s^{-1}. Although the resulting D11 scattering pattern indicates about 50 different peaks, the image resolution is very poor as some peaks are blurred because they comprise just 2 – 3 detector "cells". A finer spatial resolution would be most helpful for a obtaining a precise interpretation of the data.

3.e. maintaining a high detector efficiency

With addition to the remarks in section 4.b. it must be stressed that the instrument D11 is dedicated for low-Q studies. The majority of experiments is performed under conditions (neutron wavelength, detector distance, collimation distance) where the neutron flux at the sample is not excessive and the efficiency of the detector is an important parameter for obtaining data with good statistics in reasonable measuring times.

4. Conclusion

In the previous sections we have described the scientific requirements and the present limitations of D11 in terms of dynamic range, maximum count rate, accessible range of momentum transfer, detector resolution and efficiency. The project is aimed at an innovative evolution of the instrument with a "best possible" specification, in order to maintain ILL's leading position in small angle scattering.

As a "best possible" and cost-effective improvement it is proposed to replace the present ^{3}He detector by the ^{3}He - MWPC 128x128-7.5 (the former CERCA ^{3}He detector of D22). This detector has a considerably larger counting area thus a much better dynamic range of 15, a better efficiency and a better spatial resolution. After some modifications of the detector electronics, the maximum possible countrate (with 10% dead time loss) could be increased to about 300 kHz, i.e. a factor of 6 higher than the present D11 detector. Installing this detector at D11 would require a larger tank with an increased diameter. The consequence of a tank replacement with a diameter of $\approx$ 2m would require, however, a re-siting of the instrument IN12, an option which is at present under discussion at the ILL in the context of a general re-organisation on the siting of instruments.

It is also of interest to use the existing space in the building ILL7 and to increase the detector tank length to about 42m. In conjunction with the enhanced counting area, the tank elongation would considerably increase the totally available Q-range of the instrument and allow for measurements at significantly lower Q-values than possible at present. Table 1 summarizes the present status and the future project characteristics.

	presence	future	gain
Shape & Size (active area)	Circular, ≈ 3900 cm^2	Rectangular, ≈10000 cm^2	260 %
Spatial resolution	1 cm^2	0.5 cm^2	100 %
Max. countrate (deadtime)	60 – 70 kHz (0.85µs)	≥ 300 kHz (neglible)	x 6
Efficiency	60% (6Å), 70% (10Å) 65% (16.5Å)	≥ 80 % (6Å), ≥ 75% (10Å) ≥ 70% (16.5Å)	
Background	1 Hz integral	1 Hz integral	
Dynamic range Q_{max}/Q_{min}	7	15	100 %
Tank length and diameter	38m 1.5m	43m ≈ 2m	
Minimum Q	5 10^{-4} $Å^{-1}$	3 10^{-4} $Å^{-1}$	70 % (1/Q, size in real space)
Maximum Q	0.44 $Å^{-1}$	0.82 $Å^{-1}$	50 % (1/Q, size in real space)

Table 1: D11 project specifications: present status and future requirements