TECHNICAL AND INSTRUMENTAL DEVELOPMENTS

The development of the neutron technique and the research of new methods are two permanent goals of the LLB activity. Due to the wide field of condensed matter research in which the neutron spectrometry is involved and often even unique, this domain of activity is very large. It ranges from the designs of new spectrometers, but also implies improvements of the existing instruments (neutron flux, versatility). Optimised neutron focussing and better polarising systems are now installed on the spectrometers. Technical developments also concern the sample environment facilities: high magnetic fields (12T), cryostats (down to 50mK), furnaces (1800°C), pressure systems (40GPa)… The wide use of these devices also requires the development of raw data acquisition and data treatment systems, as simple as possible to use. All this specific neutron instrumentation generally results of collaborations between the researchers and the technical staffs (designs and drawings, "small" mechanics, electronics, computing) of the LLB, and more recently European networks between centers.

During the period 1999-2000, among the activities of the LLB, we find some important technical realisations. The major one is the rebuild of the 2T channel with a larger geometry of the output neutron beam. This important change close to the reactor core was accompanied with a new more brilliant thermal triple axis spectrometer with the polarised neutron option, owing to the higher flux of 2T. The experimental installations of LLB are continuously modernised or implemented with new options, such as the grazing angle diffractometer. Several specific neutron instrumentation projects are also developed at LLB, generally for peculiar needs of the laboratory. As example, microstrip detectors and polarising multilayers have been and will be produced and installed on several spectrometers of the laboratory. We wish to mention the powerful data acquisition and control system realised by the electronics staff of LLB, which is spread not only in neutron spectrometry, but also for X-rays or light diffraction in other laboratories.

Besides these important developments, numerous other realisations in various domains have been carried out (new sample environments, new acquisition or data treatment programs…), which render the domain of application of neutron scattering always larger.

DEVELOPMENT OF SPECTROMETERS.

A recurrent demand in neutron spectroscopy concerns the **increase of the neutron flux**. Huge progresses have been obtained in this way, due to important improvements either in multilayers guides or in the single crystal manufacture. For instance, with supermirrors guides (even polarizing), the maximum critical angle for neutron reflection (keeping a good reflectivity) is now around $3\theta_c$ (θ_c is the critical angle of naturel Nickel). These improvements, eventually combined with focussing methods, are more and more applied to the instrumentation of LLB.

Such upgrades can be heavy works, as the 2T operation almost finished this year. The latter consisted in an increase of the size of the neutron beam cross-section, 50*120mm^2 instead of 40*75mm^2. The aim of this operation was of course to get higher flux at the sample position, but also owing to the higher flux to provide a world leading ***thermal triple axis spectrometer with polarized neutrons***. Several solutions have been adopted to increase the flux. Better single crystals of Heusler alloy ($AlCu_2Mn$) have been realized in collaboration with ILL. The size of both the monochromator and the analyzer were increased; composed respectively of around 40 and 27 elements, they are bent vertically as well as horizontally. In order to fully benefit from these improvements of the spectrometer, an increase of the beam size of the 2T output was required and thus a ***major modification of the thermal beam tube 2T*** and of its dense concrete shielding. The dismounting of the old 2T neutron beam output and the installation of the new one have been performed during the reactor shut-down, in April 1999. The operation was a full success, realized in due time by the team of the Orphée reactor.

Now the thermal flux on the 2T spectrometer is much higher: the counting rate has been increased by a factor 3. Optimising this spectrometer thus offers new opportunities. Recently, non-polarised experiments on molecular beam epitaxy grown samples have been performed. They successfully led to the determination of the spin-wave spectrum of MnTe samples, of thickness of about 4-6μm and even of 1μm. Such a sample volume (~0.25mm^3) is well below that usually needed in neutron studies (typically a few cm^3). These experiments on small samples are very encouraging and promise a new way for the development of the neutron scattering spectrometry. The polarized option is being installed in 2001.

A new surface diffraction spectrometer has been mounted on the reflectometer EROS at LLB. Its originality lies in its working mode, a Laue type configuration. It is diffraction measurements at grazing incidence by a white neutron beam. In the grazing incidence geometry, a part of the beam is reflected while another part is transmitted. The latter, the evanescent wave, may be diffracted by Bragg planes perpendicular to the surface. It is then possible to get information concerning the in-plane ordering of a sample from the diffraction signal recorded by a position sensitive detector. Preliminary experiments have already been performed on non-magnetic crystals and on a magnetic Co epitaxial thin film. With the latter, four evanescent Bragg peaks have been measured, corresponding to the four possible spin combinations (++,--,+-,-+). After these promising first measurements, the ***grazing incidence diffraction*** set-up will be installed as a new option of the reflectometer EROS.

The renewal of the detection system of the spectrometer 7C2, the spectrometer devoted to studies of liquids and disordered systems, has started this last year. The wavelength used on this spectrometer is 0.07nm. The old detector was a "banana" (640 cells covering 128°) filled with BF_3 gas. It had a bad detection efficiency (17%), especially for this low wavelength. On the opposite, the microstrip detector technique developed at LLB (see the description in the next paragraph) is filled with 15 bars of ^{3}He gas, which gives a detection efficiency of 80-85%! Thanks to the latter improvement of the detection, the replacement of the banana detector was decided. The spatial resolution of the first microstrip detector of the future spectrometer, has been especially adapted for 7C2. The resolution is 2mm in the horizontal direction and 5mm in the vertical direction. Thirteen detectors of this type will be placed closer to the sample (radius of 1m instead of 1.5m), in quincunx (see Fig. 1) in order to avoid blind detection angles. The total angular detection range will be 130°, with a better resolution in the horizontal plane (0.12° instead of 0.2° previously).

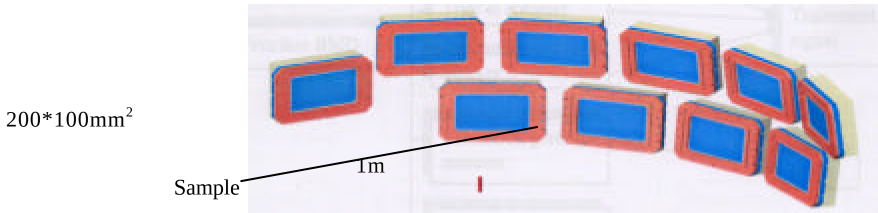

Figure 1. Layout of the future detection system of the spectrometer 7C2: 13 microstrip ^{3}He detectors (200*100mm^2) are placed in quincunx around the sample. One central detector will cover the small q domain.

Finally, we would like to stress the progress in the development of a new spectrometer, the project named **TPA** (Très Petits Angles), for small angle neutron scattering at very low q. The scattering vector range aimed with this new instrument is 10^{-4} - 10^{-2}Å^{-1}. It would allow the studies of large scale objects (1000Å) such as giant micelles, cell membranes, cavities, precipitation in alloys, biophysical gels… The principles are those of classical small angle spectrometers, pin hole collimation and 2D detection, but with a good resolution. For the collimation, methods to increase the neutron flux, such as focusing mirrors or neutron lens, will be studied later on. Concerning the detection, as the manufacture of big detectors for neutrons with a resolution of about 1mm is a present problem, the choice of the image plate technique has been made. An image plate for neutrons designed by MAR Research is now optimised on the guide G5ter. However, this kind of detector being very sensitive to γ radiation, studies of collimator and detector shielding are presently performed. The replacement of the mechanical selector by a supermirror monochromator is envisaged to get rid of the γ coming from the selector.

TECHNICAL DEVELOPMENTS, SAMPLE ENVIRONMENT AND DATA TREATMENT.

A major element in neutron spectrometry concerns the neutron detection. Unfortunately, the manufacture of large multidetectors is now a big problem for all neutron laboratories. The LLB has participated during the period 1996-1999 in the area multidetector of the XENNI program (the 10-Member European Network for

Neutron Instrumentation). In this frame, the LLB has developed ***microstrip gas detectors***. The latter are filled with high pressure He^3 gas to capture the neutrons. The conversion into electric signal is performed through a glass plate, on which a network of anodes and cathodes has been etched. Such device allows a two dimensional detection with a very good resolution (1.5 - 2mm). Several prototypes of such microstrip detectors have been tested on PRISM and EROS (the two reflectometers of LLB). They display a large dynamics and a low noise. At this moment, a large size detector $200*100mm^2$ is installed for a test period on the spectrometer 7C2, the spectrometer devoted to studies of liquids and disordered systems. It is a prototype for the renewal of the whole detection system of this instrument (described in the previous paragraph).

Besides, optical gratings used for the manufacture of microstrip detectors could also be used in the field of neutron optics. As a matter of fact, UV lithography makes possible to achieve large surface arrays (several cm^2) with periods down to 200nm. The neutron beam can be diffracted by such gratings. We envisage to use these gratings for the ***energy analysis of the white neutron beam***. As a matter of fact, the diffraction direction is a function of the wavelength. Thus, after diffraction by the grating, one can measure on a multidetector the whole reflectivity curve of a sample, without any 2θ rotation or use of a chopper.

Another improvement of the spectrometers at LLB concerns the replacement of the **mechanical selectors** of the SANS spectrometers. As a matter of fact, due to the supermirror guides installed in the past years, neutrons of short wavelength are now available at the end of the guides where the SANS machines are located. The wavelength corresponding to the maximum flux of the wavelength distribution, is now around 2 and 3Å. In order to meet the repeated demand of users to increase the q range, the PACE spectrometer is now equipped with a Dornier selector. An intensity gain of about 20% (due to a better transmission than the previous Hungarian selector) and a possible choice of small wavelengths (down to 2Å at 24000 rpm when tilting the selector) are expected. On this spectrometer, it is now also possible to remove automatically this mechanical selector and replace it by a guide element in order to perform with an additional chopper, **SANS measurements by time of flight** (TOF) on the white neutron beam. This technique has been used few times in the past at LLB. It allows us to perform SANS measurements in the whole q range in the same time with a good wavelength resolution (1-2%). We thus plan to get larger experience in the TOF procedure, in view of the future European Spallation Source (ESS). We also wish to develop TOF on the XY-multidetectors (PAXE, PAXY). As a matter of fact, thanks to the recent EuroPSD modules developed by the electronic group of LLB, such technical developments could be achieved rapidly and more easily now than previously at LLB.

Developments of the sample environment facilities are also under progress. In particular, neutron scattering experiments under **pressure** are carried out at LLB since several years. On the one hand, in soft matter, even low pressure (<1GPa) may strongly change the inter-atomic distances and the physical properties. In solids, much higher pressures are required to induce a phase transition as example. In the domain of neutron diffraction, the single-crystal diffractometer 6T2 can be used in a so-called “lifting detector” mode, which allows a variety of complex sample environments to be implemented. The equipment suite has been recently extended to encompass a powerful combination of high field (***6 Tesla*** with a ***cryomagnet*** from Oxford Instruments), very low temperature (***50 mK*** with the ***^{3}He-^{4}He dilution insert*** from Air Liquide) and high pressure (***7 GPa*** with the Kurchatov-LLB ***sapphire-anvil pressure cell***) devices (see Fig. 2). The potential of this experiment was demonstrated by recent studies of field-temperature-pressure phase diagrams in TmSe and TmTe compounds.

Figure 2. 6T2 spectrometer equipped with the cryomagnet (6Tesla), ^{3}He-^{4}He dilution insert (50mK). The high pressure cell (mounted inside the cryomagnet) allows to perform experiments up to 7GPa.

Since several years, numerous physical-chemistry systems (lamellar phases, giant micelles, liquid-crystalline polymers) are studied under shear. The experiments consist in applying a shear deformation with a characteristic time $1/\dot{\gamma}$, where $\dot{\gamma}$ is the velocity gradient. When this time is about some characteristic relaxation times of the complex fluid, important structure changes can be observed. The Small Angle Neutron Scattering technique is especially well adapted to such studies. Several in-situ **shear devices** have been realised: Couette or cone-plate shear cells. Recently, one Couette cell, with quartz windows, was improved since its velocity gradient range extended from 10^{-2} to 800 s^{-1}: this shear cell is specially adapted to the study of soft systems such as colloids. Another Couette cell, allowing studies at higher and better controlled temperature (up to 140°C), has been specially designed for small sample volumes (<500mm^3) (see Fig. 3).

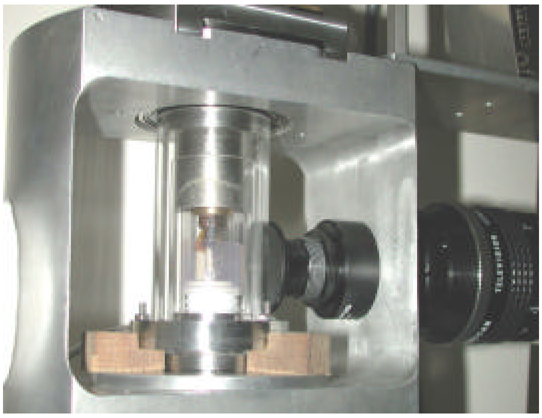

Figure 3. Couette cell, transparent for neutrons, designed to perform in situ small angle neutron scattering experiments under shear. The temperature is controlled up to 140°C. The gradient velocity range is 0.1 to 100 s^{-1}. A video camera records images of the sheared sample.

←→
20mm

The neutron scattering techniques are also very interesting to study materials or devices of technological interest. At LLB several spectrometers are specially devoted to applied research in materials science and technology, for texture and strain-stress measurements. The neutron radiography instrument, installed at the end of the guide G4 of the Orphée reactor, is widely used as a non-destructive testing technique. Such a technique is also interesting to measure some physical properties of materials. Recently, in collaboration with the University of Kiel, ***dynamic neutron imaging*** has been used for the first time to determine in situ ***viscosities and densities of silica melts***. The method consists in monitoring small neutron absorbing spheres falling in silicate melts. The furnace designed for these experiments allows measurements at temperatures up to 2000°C.

Other aspects of the neutron technique are data acquisition and control. All the spectrometers at LLB and several spectrometers in other laboratories in the world are equipped with the intelligent control systems for data acquisition developed by the ***electronic group*** of LLB. In the past, most contracts of this group were scientific collaboration projects, but ever since they integrated the Orphée Technologie's structure, their approach is more commercial and they are very reactive to the client's needs. In this way, the hardware designs have integrated downloading procedures so that the customer can upgrade the system as well as receive maintenance testing programs. Recently, they sold a data acquisition system for 2D detector to ILL, and have completed a contract with the Optic Institute of Orsay to renovate their X-ray spectrometer. In South Africa, they brought up-to-date the control and driving system of a 4-circles neutron diffractometer. A contract for the equipment of a light scattering spectrometer has been signed with the Tunisian Ministry of Education. Thus, the electronic group of the LLB has, since a long time, acquired enough experience to export and promote its know-how in spectrometer process control and data acquisition. Now, the trend is to equip, at LLB and abroad, not only neutrons but also X-rays and laser spectrometers.

On its side, the ***computing group*** of LLB has been working for the last years on a new instrument control software for data acquisition, named "***Programme Orienté Objet de Pilotage de Spectromètres***". The software architecture mimics the architecture of spectrometers, which are all doing different physical studies, but have all many common points (electronic counting devices, sample environments, motors, choppers). The instrument configuration (experiment and environment description) is defined in specific files. From these descriptions, the software adapts itself and constructs the corresponding Graphical User Interface and list of functions available on the spectrometer. It has been carefully defined to help its maintenance and its evolution. The software is now controlling 6 spectrometers of LLB.

To the development of instrumentation, are added those of **programs for data analysis** suited to each method. As example, in some treatment programs of diffraction measurements, new fits to various functions (the Rietveld analysis, pseudo-Voigt decomposition method…) are always implemented. ***FullProf***, a LLB-made program for the determination of complex crystalline structures from powder and single-crystal diffraction patterns, is widely used in physics, chemistry of condensed matter. It has recently been extended to data treatments of X-rays, including from synchrotron radiation sources. Now, crystal superstructures, like incommensurate structures, can be treated. A simulated annealing optimisation has also been included in the program to solve crystal and/or magnetic structures. Fullprof program, including a lot of examples and help, is at the disposal of the scientific community on Internet (http://www-llb.cea.fr/fullweb/powder.htm). Started in 1990, important developments of CRystallographic Imaging using Maximum Entropy (CRIME) have resulted in the maturation of two programs: GIFTED (Generalized Inverse Fourier Transforms for Electron Densities) aimed at x-ray / unpolarized-neutrons, powder / single-crystal, Patterson / Fourier densities and ACE (ACEntric) aimed at Polarized Neutron data and used to reconstruct 3D magnetization densities no matter how large the flipping ratios. Recent applications involve imaging disordered protons in molecular crystals (aspirin, $Ni(NH_3)_6Cl_2$), field-induced magnetic symmetry breaking in ($Nb_{3-x}S_4$) and detection of fractional oxygen in Hg-based high-Tc superconductors.

PERSPECTIVES.

Among the improvements planned for the forthcoming years, one can mention:
- the renewal of the detection system of 7C2 (described in a previous paragraph): thirteen microstrip detectors filled with ^{3}He gaz with an efficiency of 80-85% will increase the counting rate by a factor 4.
- the upgrade of MUSES, the Neutron Resonance Spin-Echo (NRSE) spectrometer, recently installed at G1bis. The NRSE spectrometry, an alternative version of high magnetic field Neutron Spin Echo, is especially well suited for large angle measurements. It allows to measure intermediate scattering functions, from 0.05 to 3 $Å^{-1}$. In order to improve the counting rate of the spectrometer, we plan to implement a multi-angle analysis device. This requires the construction of a second "precession" arm covering a scattering angle of 15°. It includes 25 ^{3}He detectors, the development of appropriate symmetry NSE coils for the measurements of the small time part of the spectrum and two curved or "banana like" resonance coils. The data acquisition unit developed by the electronic group of the LLB will be installed. A possible future development of TOF spin echo spectrometry could be made on this spectrometer. The development and construction of the proposed options will be performed in the frame of the collaboration with the Technische Universität München.

The LLB is also involved into European networks for the forthcoming years (2000-2003). One program concerns the neutron polarisation (in particular the supermirror polarisers and analysers), the European Neutron Polarisation Initiative (ENPI). Besides, the LLB is a member of the European contract, TECHNI (Technology for Neutron Instrumentation program), for the studies of microstrip detectors and gratings.

For the neutron physicist community, the European Spallation Source is a big challenge for the future. The LLB, with respect to its position and responsibility, should play an important role in this project. Namely, concerning the instrumentation, several techniques can be modified and/or developed in order to operate in TOF mode. We have already mentioned the possibility to perform SANS experiments by TOF on PACE (the spectrometer devoted to isotropic scattering). The SANS technique is, at the present, not considered to have a big advantage of the TOF mode. For this reason, a lot of experience could be gained on the SANS spectrometers at LLB, including measurements on multidetectors. Several other possibilities could be also settled, like in the powder diffraction spectrometry. Concerning the TOF, a lot of improvements, as focusing guides, could be made on MIBEMOL, the high resolution TOF machine of LLB, in order to increase the flux.

As a conclusion, all these developments are very encouraging as well as motivating. They render the neutron spectrometry more and more useful and determinant to any research at the microscopic level (structure and dynamics) in physics, chemistry, biology and materials science.