TECHNICAL AND INSTRUMENTAL DEVELOPMENTS

The study of the development of the neutron technique and the research of new methods are the two permanent goals of the instrumentation activity of the LLB. Due to the wide field of condensed matter physics, 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 spectrometers (neutron flux, versatility). Optimised neutron focussing and better polarising systems are now installed on the spectrometers. It also includes the developments of the sample environment facilities (high magnetic fields (12T), cryostats (down to 80mK), furnaces (1800°C), pressure systems (up to 40 GPa)…). 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 in some cases with external laboratories.

In summer 1997, the Zircaloy housing core has been replaced during the annual shut-down of the Orphée reactor. Three months were necessary for this essential operation, scheduled a long time ago. It has been successfully achieved with assiduous care by the staff of the Orphée reactor.

During the period 97-98, among the technical activity of the LLB, we find a few important technical realisations. Two of them are new instruments: the reconstruction of the reflectometer PADA (named now PRISM) and the neutron resonance spin-echo spectrometer (NRSE). These spectrometers were designed and built in collaboration with respectively "le Service de Physique de l'Etat Condensé (CEA-SPEC)" and the Technical University of München.

The experimental installations of LLB are continuously modernised. We wish to mention the realisation by the electronics staff of LLB, of a more rapid and powerful data acquisition and control system. Several important progresses have also been obtained in the sample environment facilities, as high-pressure devices specifically adapted to inelastic, diffraction or small angle experiments. Very important and due to improvements of the neutron technique, is the original 3-axis measurements of the spin-wave spectrum on very small samples grown by molecular beam epitaxy.

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

DEVELOPMENT OF SPECTROMETERS.

A neutron resonance spin-echo spectrometer.

A ***NRSE spectrometer***, which in contrast to a standard spin-echo relies on RF-coils rather than on static magnetic fields, had been built by the Technical University of München (Germany) and installed at the guide G1-bis, a polarised supermirrors guide. Both techniques can perform high-resolution energy measurements (neV) of condensed matter. The active part of NRSE acts over several centimetres, on the opposite to the traditional NSE, where the Larmor precessions occur over typical distances of several meters. A direct consequence of this peculiar point is a great flexibility of the instrument, especially for the large scattering angles from 5° up to 100°. This spectrometer is then complementary of the NSE spectrometer MESS at LLB, which is adapted to studies of long characteristic time processes at low angles. It has been designed with a wavelength range of 3Å to 12Å and a beam size at the sample position of 4*4 cm^2. The distance between the RF-coils has been set to 1.8m providing a maximum effective integral path of 1600G.m. Such characteristics give an access to a wide range of time (5ps to 15ns) at q values larger than $0.05Å^{-1}$. In these q and time domains, the neutron flux on this instrument is comparable to that of IN11 at ILL. It is well suited for studying the complex dynamics of the liquid-glass transition. The NSRE spectrometer at LLB is, at the moment, the unique machine of this type operating in the world.

A Polarised Reflectometer for the Investigation of Surface Magnetism, PRISM.

The neutron reflectivity technique has emerged less than 15 years ago. It is devoted to the studies of solid and liquid surfaces and interfaces. At LLB, among its three reflectometers, one has been recently rebuilt as a new ***polarised reflectometer with polarisation analysis***, named PRISM. As the neutron flux is a key point of the reflectometry experiments, several solutions have been operated to increase the flux. The focussing of 100mm high beam of the neutron guide to around 15mm high at the sample position and the multi-layers monochromator have required a huge modification of the spectrometer (the previous spectrometer was using a graphite monochromator). The wavelength resolution $\delta\lambda/\lambda$ has changed from 0.6% to 4%. These two improvements lead to increase the flux by a factor 15. A near future development will be the installation of a position sensitive detector based on the “microstrip” technique (realised in the frame of the European programme ***XENNI***), which will allow off-specular studies on magnetic systems. The PRISM spectrometer is a good reflectometer, very well suited to magnetic studies; its new design renders it world leading.

TECHNICAL DEVELOPMENTS, SAMPLE ENVIRONMENT AND DATA TREATMENT.

A recurrent demand in neutron spectroscopy concerns **the increase of the neutron flux.** This goal can be now achieved due to important improvements either in multilayers guides or in the single crystal manufacture. 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 the natural nickel). These improvements eventually combined with focussing methods are more and more applied to the instrumentation of LLB. The intensity gains thus obtained ranged in between 2 and 4-5 (even 15 mentioned above for PRISM, where the graphite monochromator was replaced by a multilayer monochromator).
Such upgrades can be “heavy” works, as the 2T operation, decided two years ago, to provide the best ***3-axis spectrometer with polarised thermal neutrons*** in the world. Several solutions have been adopted to increase the flux. Good single crystals of Heusler alloy ($AlCu_2Mn$) have been realized in collaboration with ILL. The size of both the monochromator and the analyser are increased; they are bent vertically as well as horizontally. In order to benefit from these technical improvements, an increase of the beam size of the 2T chanel was required and thus a major modification of the 2T output and of its dense concrete shielding. The installation of the whole system is scheduled during the reactor shut-down, in April 99.
A new possibility is now proposed for inelastic neutron scattering measurements. It allows users to compare data obtained in the different neutron laboratories in the world, but also with other techniques, such as NMR. This method (recently settled at LLB) gives ***absolute measurements of inelastic scattering on 3-axis spectrometer***. It will be useful in the understanding of the origin of the measured intensity. As example, it could be a step to relate the magnetic fluctuations observed in YBaCuO to the superconductivity mechanisms.
Optimising the neutron flux on the spectrometers may also offer new opportunities. As a matter of fact, recently, ***3-axis inelastic scattering experiments on molecular beam epitaxy grown samples*** have been tempted. 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 is well below that usually needed in neutron studies (typically a few cm^3). Thus, such experiments on small samples are very encouraging and promise a new way for the development of the neutron scattering spectrometry.
Besides, since November’97, the powder diffractometer 3T2 is equipped with a new ***focussing Ge monochromator***. The cut-off angle is still $2\theta_M \approx 90°$, the wavelength 1.225Å, but due to the properties of the crystals (size and mosaic), the flux at the sample is four times higher than before. However, here, as the incident beam of 3T2 doesn’t illuminate the whole monochromator, the focussing feature is then not fully used. An increase of the height of this beam, similarly to the 2T operation, is under consideration.
A focussing system has also been installed on the spin-echo spectrometer MESS. In order to balance the lack of intensity due to the high-resolution energy of this spectrometer, several ***focussing guide elements coated with a non magnetic Cu^{65} isotope*** have been mounted in the first precession arm of the spectrometer. At the wavelengths commonly used on the spin-echo MESS, 6 and 8Å, the gain of intensity is in between 3 and 5. A specific ***pneumatic system of positioning of the different guide elements*** leads to flexibility in the choice of the energy resolution and the flux.

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. Depending on the

study involved, several pressure cells have been specifically designed. One made with an invisible alloy ($Ti_{0.34}Zr_{0.66}$) (coherent scattering length close to zero) allows to reach temperatures up to 800K under 1kbar; it has been used for studies of molecular liquids. Besides, a ***high pressure cell for small angle neutron scattering*** was realised to study supercritical fluids. It has sapphire windows and a great care of the temperature regulation allows a stability of ±30mK at 400°C.
On the other hand, in solids, much higher pressures (> 10GPa) are generally required to induce a phase transition. Such an increase of pressure is generally obtained by an important decrease of the sample volume. During the last years, a Paris-Edinburgh high-pressure cell has been adapted to a 3-axis inelastic neutron scattering spectrometer, where focussing monochromator and analyser crystals had been previously settled. ***Inelastic scattering experiments up to 12GPa*** are now possible on samples of effective volume of $10mm^3$. This high-pressure equipment is now at the disposal of any user and several experiments have been carried out successfully for studying the phonon dispersion of different compounds.
In the domain of neutron diffraction, the magnetic studies under very high pressure require an important technical development both of the diffraction spectrometer (for the flux) and of the pressure cell. As a matter of fact, pressures higher than 10GPa are necessary for magnetic structure studies on powders or single crystals. During the last three years, several focussing systems (in the horizontal and vertical planes) made with Ni-Ti supermirrors ($3\theta_c$) have been developed on the powder diffractometer G6-1. The angle of focussing is variable in order to choose the optimal ratio of the intensity and resolution. The maximum gain of intensity achieved is about 7. Two pressure cells, with sapphire (P < 10GPa) and diamond (P > 10GPa) anvils, can rotate freely inside the cryostat, allowing to study textures or magnetic domains distributions. Recently, measurements on a GdAs sample of ~1 mg weight have been done at a pressure of 43GPa! The G6-1 ***diffractometer for high-pressure studies, MICRO***, using special focussing devices to increase the neutron flux, is now operating half the year. This spectrometer is devoted to magnetic studies in the 50GPa pressure range. A new multidetector covering a high solid angle range is under construction at the EMBL (Grenoble); with the expected gain of counting rate, measurements at higher pressures could be tempted in the future.

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 specific 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, a very peculiar cone-plate cell has been realised: it allows the measurements of the scattering intensities in the three directions of the velocity with respect to the scattering vector q. The difficult observation of scattering in the plane (velocity, velocity gradient) could only be made owing to the huge penetration depth of neutrons in the materials. Another ***cone-plate shear cell***, with quartz or sapphire windows, was designed with a velocity gradient range 10^{-3} to 200 s^{-1}, specially adapted to the study of viscous systems such as polymers.

The neutron scattering techniques are also very interesting to study materials. At LLB two diffractometers are specially devoted to **applied research in materials science and technology**, for **texture and strain-stress measurements**. Among the recent improvements in this field, the ***DIANE diffractometer*** (G5-2) has been recently equipped with a **mechanical test machine** for strain measurements under applied stress or during fatigue tests. With its detector installed two years ago, this spectrometer is very useful to industry, for the determination of residual stresses in materials. Its gives to the LLB, the opportunity to participate to the European Brite-Euram program, ***TRAINSS*** (TRAining Industry in Neutron Stress Scanning) and in the international ***VAMAS*** project, which aims at the definition of a standard method for the measurements of residual stress by neutron diffraction.

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. In this sense, ***FullProf***, a program for the determination of complex crystalline structures from powder and single-crystal diffraction patterns, is extended to data treatments of X-rays (including from synchrotron sources). ***WinPlotR*** is a new tool working on Windows (95, 98, NT) systems, for data plot and fit of powder diffraction patterns. These programs, including a lot of examples and help, are at the disposal of the scientific community on Internet. In the same mind, the treatment program of SANS data, PASIDUR, has been adapted to the data file format of ILL. Furthermore, this program, used on the PACE spectrometer of LLB, allows fully automatic changes of the configuration (collimation, detector distance, wavelength, tuning, sample position, temperature…) and the data acquisition.

All the spectrometers at LLB and several spectrometers in other neutron laboratories in the world are equipped with the intelligent ***control system for data acquisition***, named “LLB DAFFODIL System”. Its philosophy relies upon independent execution of three main functions (positioning, counting and position sensitive detector acquisition) together with versatility and simplicity of use for the physicists. In the past two years, a major upgrade of this control system has been studied by the electronic group of LLB. With the new counting module, a fully automatic acquisition mode for neutron polarisation and analysis is possible. Besides, the huge memory of the acquisition module allows all kinds of association of the multidetector cells, and the performance of the Time of Flight mode have been extended. A further expansion to kinetics experiments on multidetectors can now be considered.
The possibilities of the “DAFFODIL” system have been extended by the ***electronic service*** of LLB to manage different kinds of analogic signals (ADC, DAC, input-output voltage), to program logarithmic time scales for fast acquisition. All these facilities are used in several home made instruments: the rheometers, for the viscosimeters and the light scattering spectrometers. On its side, the ***computing group*** goes on the development of data acquisition programs; up to now about ten spectrometers of LLB are running with their recent programs. Besides, as only 5 persons are working in this group, they cannot solve all the problems encountered by the large number of PC’users. They took the decision to spread a PC’s administration system, Windows NT, to survey most of the PC machines.

The LLB has participated during the period 1996-1999, in two areas of the XENNI program (the 10-Member European Network for Neutron Instrumentation) : that of polarising multilayers and that of multidetectors. Several large surface etched transmission polarisers have been realised and tested on the reflectometers at LLB. It is now possible to produce polarisers with less than 10% of absorption (instead of 30% for the conventional ones).
The technology of optical gratings is used to build a new type of position sensitive detector with a high spatial resolution, the microstrip detector. With UV lithography makes it possible to achieve large surface arrays with periods down to 200 nm. This type of detector has very large counting rates and very low noise.

PERSPECTIVES.

Among the improvements planned for the forthcoming years, one can mention:

- a new multidetector covering a high solid angle range, under construction at the EMBL (Grenoble). It will be installed in 1999 on the powder diffractometer for high-pressure, MICRO ;
- another multidetector for the new PRISM reflectometer, based on the microstrips technique, in the frame of the European program XENNI ;
- in autumn 1999, the final installation of the polarised neutron option on the thermal 3 axis spectrometer 2T ;
- a double chopper for the EROS reflectometer, which will give variable Δq values (between 1% and 4%), is under tests at LLB.

Furthermore, we are considering the replacements of the mechanical selectors of the SANS spectrometers. As a matter of fact, since they are partially coated with supermirrors, the guides where the SANS instruments are installed have a maximum flux of the wavelength distribution around 2 to 3Å, which cannot be used with the present selectors. In order to meet a demand of users, i.e. to increase the available q range, a Dornier selector was ordered at the end of last year. It is planned to be installed on PACE. An intensity gain of about 20% (due to the transmission) and a possible choice of smaller wavelengths (due to the maximum velocity) are expected.
Finally, to end this summary of the technical developments at LLB, we would like to stress two projects of new instruments. A second **time of flight spectrometer,** with high flux**,** would allow to satisfy the important number of proposals for experiments, notably in biology. The time focussing technique studied allows an increase in flux of approximately one order of magnitude compared to choppers-designed spectrometers, as Mibémol, with a comparable energy resolution (40 to 200 µeV). This increase is mainly due to a monochromatisation of the incident beam using crystals operating in Bragg geometry. We will use vertically and horizontally focussing monochromators.
A **Small Angle Neutron spectrometer (TPA), at very low q,** is under consideration in order to extend the existing possibilities in SANS. The scattering vector range aimed 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, biophysic gels… As the manufacture of large position sensitive detectors for neutrons with a good resolution (~1mm) is a major problem, we plan to use an image plate. At the beginning, a pin hole collimation will be used; further possibilities in this direction will be studied later on.
As a conclusion, all these developments are very encouraging and 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.