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 0.05$Å^{-1}$. 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.

NEUTRON RESONANCE SPIN-ECHO SPECTROSCOPY AT LLB

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Neutron spin echo spectroscopy (NSE) has been discovered by F. Mezei in 1972[1]. The basic principle of the technique is to use the neutron spin to monitor the neutron velocity changes induced by scattering processes. The measured signal $P(\mathbf{Q},\tau)$, i.e. the polarisation of the scattered beam at the echo point, is proportional to the cosine transform of the scattering function $S(\mathbf{Q},\omega)$. NSE provides the highest energy resolution nowadays achievable in neutron scattering and is consequently a very useful tool for the measurements of slow relaxation processes occurring in many fields of condensed matter physics.

In 1987, a new scheme for NSE spectroscopy was proposed by R. Gähler and R. Golub[2]. In this technique, called Neutron Resonance Spin-Echo (NRSE), the two high magnetic field coils are replaced by four radiofrequency coils; it is directly inspired by the principle of Nuclear Magnetic Resonance. On the contrary to the traditional NSE, where the Larmor precession occurs over typical distances of several meters, the active part of NRSE acts over distances of several centimetres ; the rest of the neutron path has to be magnetically shielded. As a consequence of the localisation of the fields, this technique allows an increased flexibility within the same covered Fourier time domain.

A spin echo spectrometer based on this principle has been constructed in collaboration between the TU München and the LLB and was installed at the neutron guide G1bis. This spectrometer has been designed for measurements over scattering angles ranging from 5 to 100 degrees. Complementary to the conventional NSE spectrometer of the LLB, which is specially adapted for low angle studies, it bridges a gap in the NSE spectroscopy at the LLB. Several innovations have been realised, such as a new design for the radiofrequency coils in order to minimise the stray fields and the consecutive decreases of the polarisation (see figure 1). A polarising section ($FeCoTiN_x$ supermirrors) has been implemented into the curved guide[3] providing a useful neutron wavelength range between 3 and 12 Å ; the polarised neutron flux is of $0.9\ 10^7$ $n.cm^{-2}s^{-1}$ at the sample position (λ=4.8Å, $\Delta\lambda/\lambda$=0.15). *

The distance between the NRSE coils has been set to 1.8 m providing an effective field integral path of ~1600 G.m whereas the sample to detector distance has been kept shorter than 3 meters. This leads to intensity accessible Fourier time domain comparable to the classical NSE spectrometer IN11 of the High Flux Reactor of the ILL.

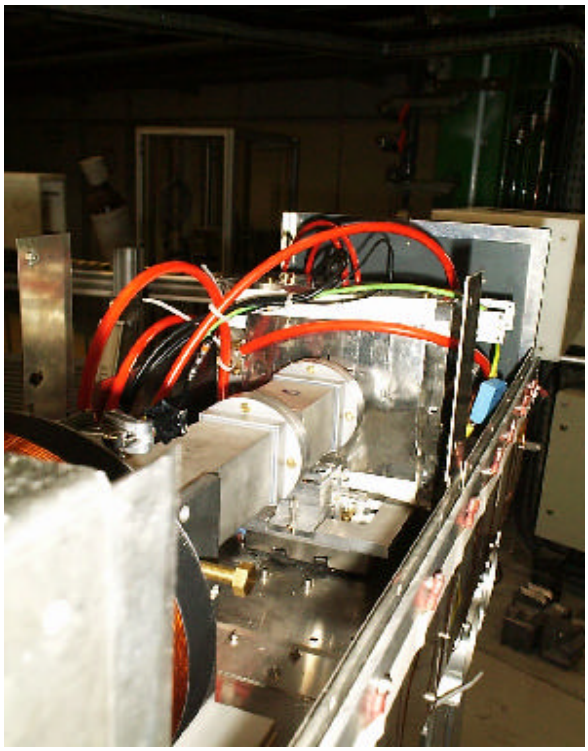

Figure 1. The NRSE intrument G1bis at Saclay

Tests measurements were performed late 1996 and during 1997, but real experiments only started after the commissioning of the new guide and were completed in 1998.

For example, measurements were performed to study the temperature-wave-vector dependence of the slow relaxation processes of a ionic mixture $Ca[(NO_3)_2]_{0.4}[RbNO_3]_{0.6}$. In such systems in which a liquid to glass transition is observed, complex dynamics occur over large time scales. These dynamics cannot be understood by a simple cross-over from vibrational excitations to structural relaxation.

Experimental data show additional intensity, which can be ascribed as rattling of particles in transient cages formed by their neighbours, as it has been quantitatively modelled in mode coupling theory[4]. The predicted scaling laws have been verified in a couple of systems, the most prominent example being $Ca[(NO_3)_2]_{0.4}[KNO_3]_{0.6}$, which is believed to stand as a model for glass forming liquids[5]. This believe has

* From gold Activation measurement.

been challenged by recent dielectric measurements[6] on a chemically homologous system $Ca[(NO_3)_2]_{0.4}[RbNO_3]_{0.6}$, which have shown a quite different evolution of the dynamic susceptibility.

NSE spectrometry is specially suited for the study of the slowest part of the relaxation, called α-relaxation. This process enters the time range of the spectrometer, for temperatures significantly above the calorimetric glass transition. Typical spectra obtained at $Q=1.7Å^{-1}$ are shown in figure 2. The count rate was nearly 60 neutrons.s^{-1}, and the polarisation at $\tau=0$ was around 0.6. Measurements were performed at temperatures ranging between 395 and 496K. The continuous lines represent fits with the Kohlrausch-Williams-Watts (KWW) function. The inset shows the same spectra, but now shifted in time according to the time-temperature shift principle. The relaxation times extracted from the individual KWW fits were taken as scaling times. Apparently the data obey the time-temperature shift principle.

In summary, the NRSE spectrometer is well suited for high-resolution studies involving slow dynamics within microscopic and mesoscopic length scales. These cover wide domains such as physics of glasses and liquids, biology, polymer science, critical phenomena…. Technical developments under progress should allow to perform studies on crystals within the very next future.

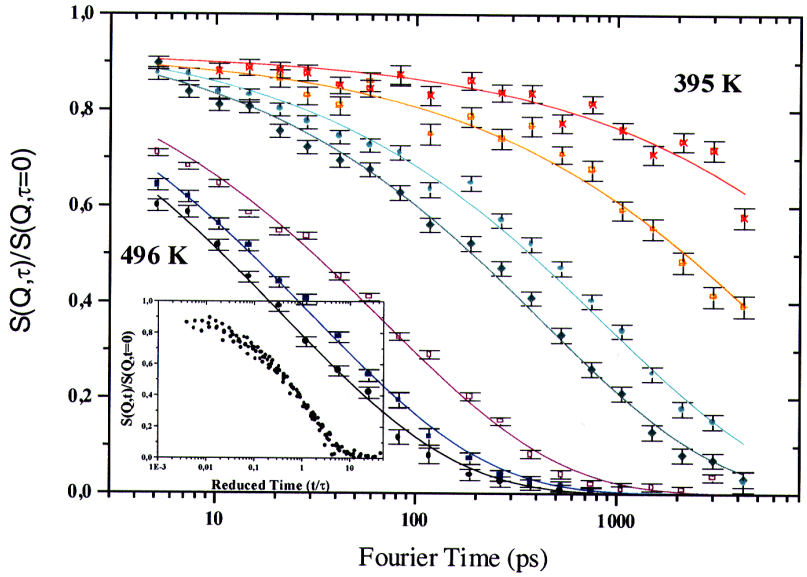

Figure 2 : Intermediate scattering function S(Q,t) for $Ca[(NO_3)_2]_{0.4}[KNO_3]_{0.6}$ at $Q=1.7Å^{-1}$ on the NRSE spectrometer G1bis.

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NEW MONOCHROMATOR AND FOCUSING GUIDE ON THE POLARISED NEUTRON REFLECTOMETER PRISM : A POLARISED REFLECTOMETER FOR THE INVESTIGATION OF SURFACE MAGNETISM.

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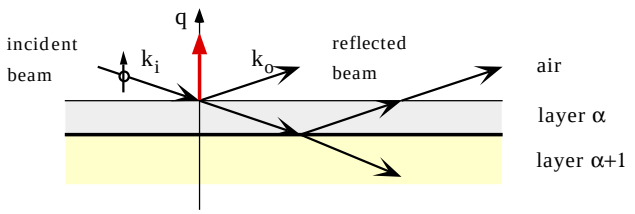
The neutron reflectivity technique has emerged less than 15 years ago. It appeared as a key technique in the study of polymers and magnetic thin films. Topics such as polymer interdiffusion, di-block copolymers ordering have been addressed. Besides, especially after the discovery of the giant magnetoresistance (GMR) effect, neutron reflectivity has been successfully applied to the study of magnetic multilayers and ultrathin films. Problems such as the magnetic ordering in rare-earth multilayers, surface magnetism and anisotropy in ultrathin magnetic layers have been solved. However, in order to extend the possibilities of the neutron reflectivity, an increase of the flux was necessary. In that aim, we have recently rebuilt a polarised reflectometer with polarisation analysis, dedicated to the study of magnetic thin films.

Neutron reflectivity principle

The neutron reflectivity technique consists in measuring the reflection of a neutron beam on a thin film (liquid, polymer or solid) at grazing incidence. The measured reflectivity as a function of the scattering wave vector **q** gives information on the chemical composition, the thickness of the layers and the roughness of a multilayer system. Neutron reflectometry is especially interesting to study polymers because of the large scattering length of hydrogen, and of the possible labelling of polymer chains by selective deuteration.

Moreover, the neutron is a ½ spin particle and the magnetic interaction with unpaired electrons is very large (as large as the nuclear scattering). By using polarised neutron reflectivity with polarisation analysis, it is possible to obtain information on magnetic systems: the type of magnetic ordering in multilayers systems (ferro, anti-ferro or helicoidal) or more generally the magnetisation profiles throughout the depth of magnetic thin films (along the scattering vector **q**).



Reflectivity geometry on a multilayer system.

To increase the available neutron flux, three different solutions are or will be put in place :

- increase of the wavelength spectrum width dl/l by using a multilayer monochromator,
- focussing of a 100 mm high neutron beam onto a 15 mm high region at the sample position,
- use of a position sensitive detector.

The previous PADA reflectometer was a two axis spectrometer using a graphite monochromator which had an excellent wavelength resolution $\delta\lambda/\lambda$ (~0.6%). The new PRISM spectrometer is still located on the guide G2 of the reactor Orphée. Here is the description of this instrument, schematically represented in figure 2.

The incident beam is deviated and monochromatised by a 3 m guide (M) made of nickel-titanium multilayers. The direction of the monochromatised beam makes an angle of 2.4° with the direction of the main guide. This part of the guide has been built and mounted by the CILAS company. The angular deviation of 2.4° is however not enough to move away the output beam from the main guide at the sample position. Thus, we have mounted two 1.80 m long $2\theta_c$-supermirror neutron guides (B) after the monochromator. The total beam deviation, from the main guide at the sample position is of 900 mm. This is enough for the shielding around the main guide (50mm lead, 250mm concrete) and the sample environment (cryostat, magnetic field). Since most of the studies on magnetic samples are realised on samples less than 20mm wide, we have focussed the beam vertically on the sample position. This vertical focussing is realised by a 8 m conical neutron guide (C) made of $2\theta_c$-supermirrors, which is interrupted twice. These two interruptions allow the introduction of the polarising (P) and flipping systems. The transmission polariser is made of Fe/Si multilayers deposited on 50 mm high silicon substrates (these mirrors were provided by Th. Christ from the HMI in Berlin). The incidence angle on the polarisers is small, 0.3°, in order to reject long wavelength neutrons generated by the monochromator system. The polarisation efficiency is 0.97 and the transmission of this polariser is 70%. At the sample position, a 50 mT field allows to maintain a good polarisation efficiency. The analysis system (A) is similar to the polariser except that the height of the device is 80 mm. It enables a full analysis of the

reflected beam, which is highly divergent in the vertical direction.

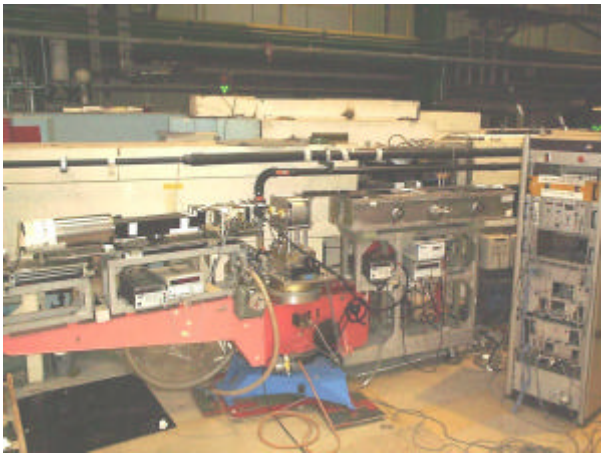


Figure 1. The Polarized reflectometer PRISM

With these two improvements (new monochromator and focussed beam), the available flux has been multiplied by a factor 15 and is presently of 5×10^5 neutrons/cm².s after analysis on the detector with a resolution $\delta\theta = 0.03°$.

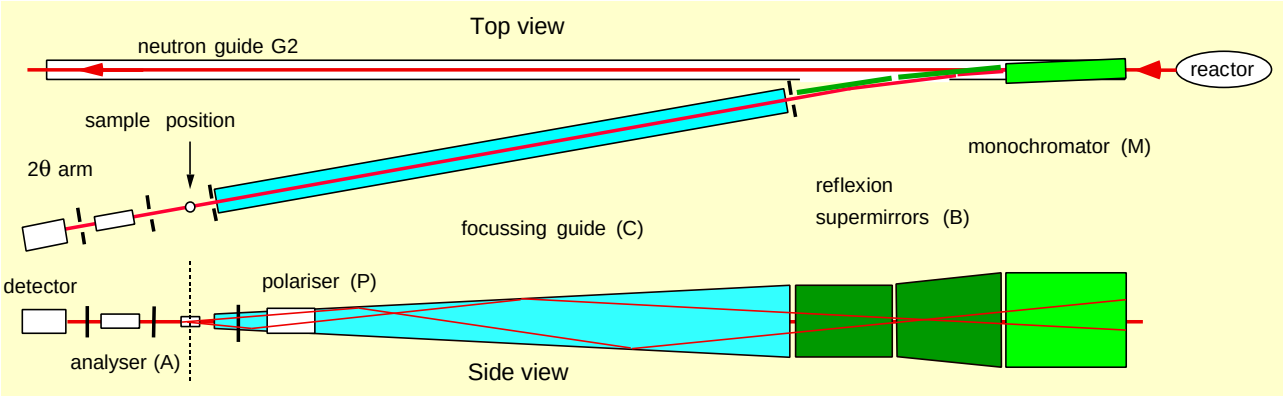


Figure 2. Scheme of the PRISM spectrometer and of its focussing guide, top view and side view

This high neutron flux will allow two main progresses in the reflectivity experiments. It will drastically reduce the measuring times of the reflectivity curves, and will permit measurements in various conditions of magnetic field and temperature. The second gain is the scattering vector range, twice as large as before. ($1.10^{-2} < q(Å^{-1}) < 0.25$). The wavelength is now 4 Å with a resolution $\delta\lambda/\lambda$ of 4%. The angular resolution $\delta\theta$ can be varied from 0.01 to 0.06°. The intensity dynamic range of measurement is now typically between 10^5 to 10^6 neutrons/s on a 1 cm² sample for an 8 hours full analysis scan and a resolution $\delta\lambda/\lambda = 4\%$ and $\delta\theta = 0.05°$ (see figure 3).
During 1999, a position sensitive detector, based on the micro strip technology (currently developed in the frame of the European XENNI program) will be installed. It will allow off-specular studies on magnetic systems.

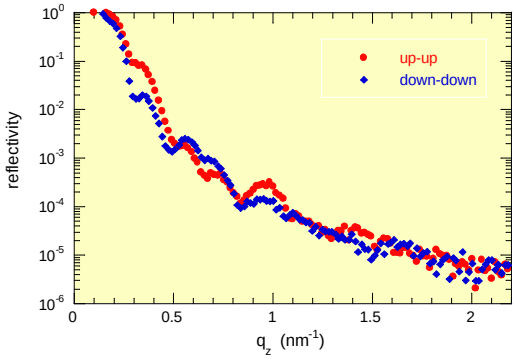


Figure 3. Reflectivity of a spin-valve system (Mn/Fe/Co/Ru /Co/NiFe) under a 0.3T field. The measurement time was 3 minutes per point on a 1 cm² sample

3-AXIS INELASTIC NEUTRON SCATTERING ON MOLECULAR BEAM EPITAXY GROWN SAMPLES

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Optimizing the use of neutron beams on triple axis spectrometers implies focusing by **curved monochromators and analysers**, bent vertically as well as horizontally (Figure 1).

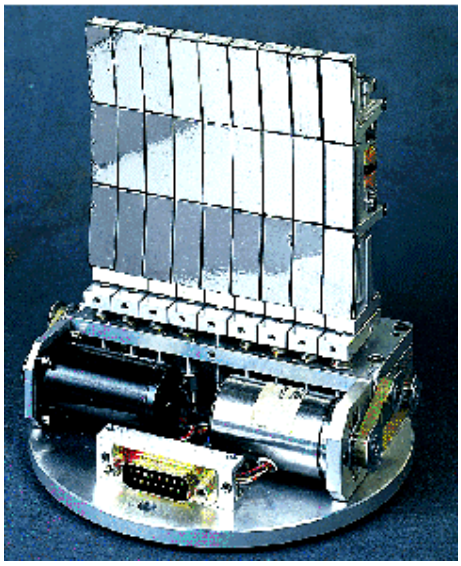

Figure 1. Focusing analyser of the 3-axis spectrometer 1T

The gain in intensity, keeping a low background level, offers new opportunities of **inelastic neutron scattering** measurements on small size samples (**a few mm^3**).
Thus, **MBE grown samples** have been used to measure **spin waves on MnTe**. Such measurements are illustrated on Figure 2. MnTe is often used to obtain by substitution of Mn by non magnetic ions (e.g. Zn or Cd) diluted magnetic semiconductors. These compounds have the Zinc blende (ZnS) cubic structure and the evolution of their magnetic properties depends on exchange integrals, responsible of the magnetic order of the pure compound. But pure MnTe has not the blende structure, but that of hexagonal NiAs! **Only MBE grown samples of MnTe have the blende structure**, because of uniaxial constraint due to epitaxy. Such samples offer therefore the possibility to **determine exchange integrals** by measuring the **spin wave spectrum of MnTe**, with inelastic neutron scattering. Raw data measured on MBE grown samples are illustrated on figure 2.
They are obtained on samples of **4 and 6 mm width** grown at the Institute of Physics of the Polish Academy of Sciences in Warsaw, on an AsGa substrate with a buffer of 2 or 4 µm of CdTe. With a surface of about 2 cm^2, **the sample volume was about 1 mm^3**. The only known information was the value of the anisotropy gap of the spin wave spectrum, that previous Raman scattering measurements had determined as ≈1.05 THz.

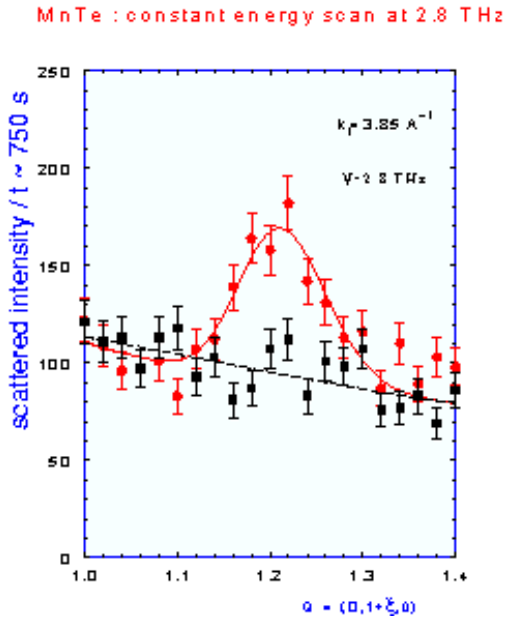

Figure 2: Raw data obtained at T=13 K. · and 75 K ■ Spin-waves only exist at low temperature.

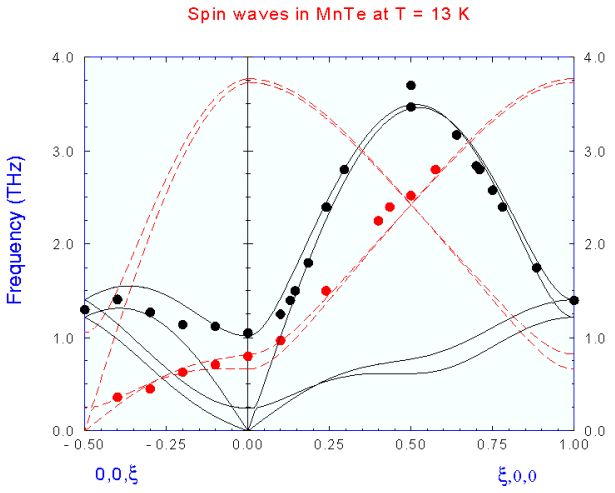

Figure 3. Black lines and symbols are respectively the measured and calculated spin-wave spectra at T=13 K for one magnetic domain in the sample.
Red dashed lines and red symbols correspond to modes in the second magnetic domain of the MBE sample.

Study of the sample by elastic neutron scattering [1] confirmed its structural and magnetic characteristics. The magnetic transition takes place at $T_N \approx 65$ K and is associated to a structural transition (tetrahedral distortion). The low temperature magnetic structure is **antiferromagnetic of type III**. If [1,0,0] is the crystallographic axis perpendicular to the layers, the doubling of the magnetic cell is along [0,1,0] or [0,0,1], due to the nature of the buffer. Therefore, in the MBE sample, 2 magnetic domains coexist and we have to account for the superimposition of 2 reciprocal lattices. This lowers the scattered intensity and hampers the analysis of the results. Anyhow, a pertinent set of data has been obtained with a counting time of about 15 minutes per step. On Figure 3 are reported the experimental results and the theoretical dispersion curves, calculated with a Heisenberg hamiltonian

A test measurement has been done on a **MnTe/ZnTe multilayer**, made of alternating 20 MnTe and 3 ZnTe atomic layers. The sample width was 1 μm, but it was a single domain. The long range magnetic order is preserved and spin gap at q=0 as well as the zone boundary mode have been observed.
Even though such experiments benefit of the high value of the Mn moment (spin 5/2) [2], they are very encouraging as a first step towards the study of dynamical properties of magnetic multilayers.

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[2] The inelastic results are not yet published.

NEUTRON DIFFRACTION UNDER VERY HIGH PRESSURES: EXPERIMENTS IN THE 40 GPa PRESSURE RANGE.

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The role of high pressures in solid state physics is essentially twofold: 1) pressure varies interatomic distances in a controlled way and therefore could be used to verify theoretical models, 2) it induces new phases, which may exhibit unusual characteristics. In practice, the possibility of using pressure strongly depends on the range of pressure needed and on the technical tools required to reach it. In soft matter, even pressures of less than 1 GPa may strongly change the interatomic distances and the physical properties. In contrast, in most solids, much higher pressures (10 GPa or more) are needed to produce significant change of the sample volume ($\Delta V/V$ >10%). In the recent years, crystal structures studies in the 100 GPa pressure range have strongly benefited from the development of third-generation synchrotron X-ray sources, whereas magnetic studies in the same pressure range are still much less developed. Anomalous magnetic X-ray scattering cannot be used because of the huge absorption of the pressure device, and Mössbauer studies do not provide any information about the long range magnetic order. Neutron diffraction is usually limited to much lower pressures (1–3 GPa). This is because pressures above 10 GPa could be reached only by a drastic decrease of the available sample volume (down to 10^{-4} to 10^{-6} mm^3 in the 100 GPa pressure range). Such a sample volume is well below that usually needed in neutron studies (typically a few mm^3 to a few cm^3).

At the beginning of the 1990's, a project was initiated at the LLB to develop magnetic studies under very high pressure using high intensity neutron diffraction [1]. The steady state reactor with a cold source provides the most convenient q-range for such studies.

The project encompasses both single crystal and powder diffraction under pressure. Single-crystal diffraction yields very detailed information about spin arrangements. Numerous studies using single crystal diffraction under pressures were performed at the LLB during the last years [1,2], most of them on the lifting detector spectrometer 6T2. Very recently a new superconducting magnet and a dilution refrigerator, compatible with the high pressure setup, have become available. Implementing high magnetic fields (up to 8 T) and very low temperatures (down to 30 mK) together with pressures up to 10 GPa opens new possibilities to single crystal studies.

On the other hand, powder diffraction is the most straightforward way to extend neutron studies well above 10 GPa. In this pressure range, single crystals could be easily destroyed by pressure inhomogeneities or by a first order structural transition.

The development of a specialized powder diffractometer, allowing neutron studies to be made at pressures up to 50 GPa, i. e. at higher pressures than in any other neutron facility, was the most ambitious part of the project. During the last three years the G6.1 diffractometer (equipped with a 400-cell multidetector) was transformed in a specialized spectrometer, optimized for the magnetic diffraction of small samples (less than 1 mm^3) under very high pressures. In this high pressure version, G6.1 is equipped with a double–stage focusing system inserted between the monochromator and the sample position (fig 1).

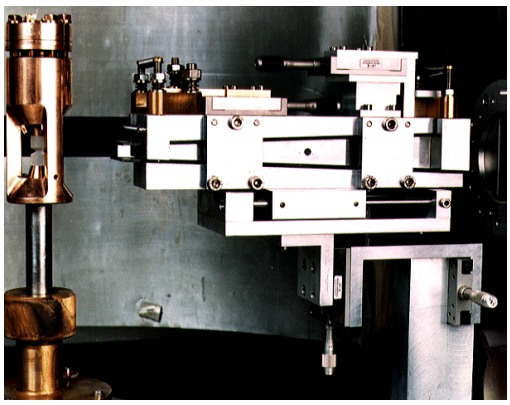

Figure 1. The pressure cell and the focusing device used in the high pressure version of the G61 spectrometer.

The focussing system, consisting of Ni-Ti supermirrors "compresses" the neutron beam both in horizontal and in vertical planes [3]. Using a double-stage system allows us to simulate a curved profile of the mirrors' surface. This is the optimal profile to get both the highest gain in intensity for a given length of the system (limited by the free space between the monochromator and the sample) and a smooth angular distribution of neutrons in the focused beam. The variable angle of focussing makes it possible to

choose the optimal trade-off between intensity and resolution for each experiment. The maximal gain in intensity obtained by focusing is 7.

Together with a high intensity of the incident beam, a very low background is crucial to measure small samples. A sophisticated protection and an especially designed cryostat allow us to reduce the background to a very small value – only a few counts/per cell/per hour.

We use pressure cells with sapphire (P < 10 GPa) and diamond anvils (P > 10 GPa). The implementation of the sapphire anvil cells for neutron diffraction is based on over 10 years of experience gained for such experiments in Kurchatov Institute (Moscow). A number of such cells, adapted to low temperature measurements, were developed and implemented in the neutron experiments (single crystal or powder) at the LLB during the last years. One of the most compact cells with sapphire anvils is shown in fig. 2. This cell can be rotated freely inside the cryostat, allowing to study textures or magnetic domain distributions. The cell is compatible with the dilution refrigerator and the superconductive magnet, making possible to use high pressures, magnetic fields and mK-temperatures in the same experiment.

Figure 2. This small pressure cell can be rotated inside a cryostat

The power of the above technique is illustrated here by the study of magnetic interactions in two closely related systems, the europium monochalcogenides and the gadolinium monopnictides. Both families, having the same crystal structure and the same electronic structure ($4f^7$) for the cations (Eu^{2+} and Gd^{3+}) are usually considered as "model systems" to study magnetic interactions in semiconductors and semi–metals. The neutron diffraction patterns of EuTe and GdAs under pressure are shown in figs. 3 and 4.

The pressure strongly modifies the magnetic properties of EuTe, resulting in a sequence of magnetic transitions at pressures around 10 GPa [4]. As the lattice constant decreases, the ferromagnetic exchange interaction increases very rapidly, leading to a transformation of the initial antiferromagnetic order to a ferromagnetic one, followed by an exponential increase of the Curie point [5].

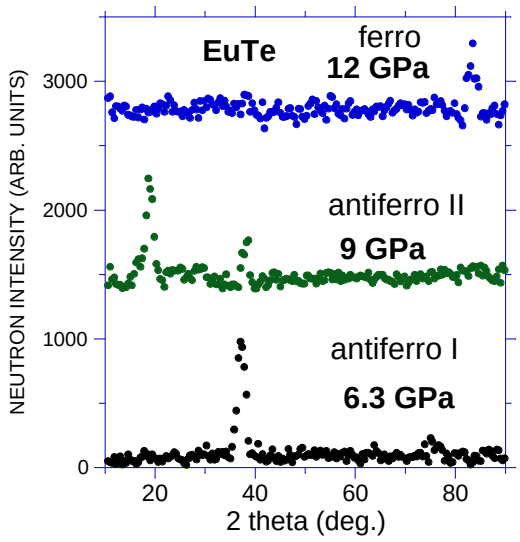

Figure 3. Magnetic neutron diffraction spectra under high pressure in EuTe.

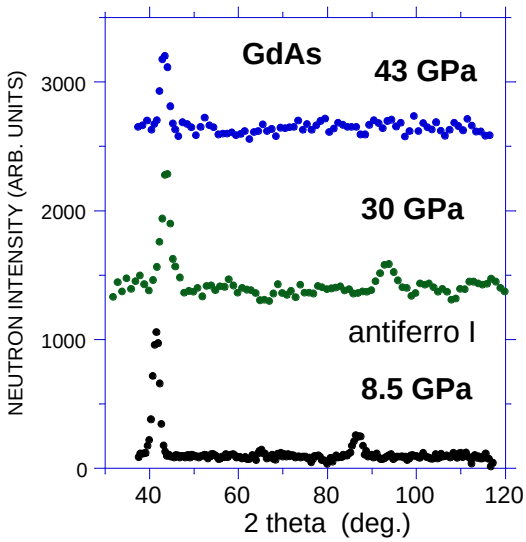

Figure 4. Magnetic neutron diffraction spectra under high pressure in GdAs. The GdAs sample was prepared by A.Ochiai (Univ. of Niigata, Japan).

The GdAs sample, having the same magnetic structure at ambient pressure, shows a very different behavior. Pressure only increases the Néel point. Even at 43 GPa, when the volume decrease reaches about 40%, the compound remains antiferromagnetic. In this measurement, the sample was as small as about 1 mg of weight (0.002 mm^3). This is likely the smallest sample under the highest pressure ever measured with neutrons.

Comparing the two systems (GdAs and EuTe) allows us to see the role of the band structure in the formation of the antiferro- and ferromagnetic exchange between the cations.

In the year 1999, we expect the next upgrade of the high pressure version of the G6.1 spectrometer ("MICRO" diffractometer). A new multidetector, optimized for small samples studies and covering a 7 times larger solid angle, is currently under construction in the European Molecular Biology Laboratory (Grenoble), in collaboration with A. Gabriel. The upgrade of the spectrometer will allow us to increase the available pressure range above 50 GPa and will also provide better possibilities to meet the needs of a growing number of experimental teams, suggesting various studies under pressure in many different fields of physics. Besides the traditional field of magnetism under pressure, new subjects appear, for instance the studies of mesoscopic structures such as the recently discovered carbon nanotubes.

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DATA ACQUISITION & CONTROL SYSTEM FOR NEUTRON SPECTROMETERS AT THE LLB

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Laboratoire Léon Brillouin (CEA-CNRS)

There are 24 neutron spectrometers installed around the ORPHEE research reactor, all equipped with an acquisition and control system known as « **DAFFODIL SYSTEM** ». This system has been developed by the electronic group of the laboratory. Its backbone is the IEEE 488 BUS. Up to 32 peripherals can communicate with the user host computer via this standard communication protocol. The DAFFODIL SYSTEM is composed of a few IEEE 488 devices, which are designed for independent execution of complete functions (see figure 1).

The past two years, three main functions **(positioning**, **counting** and **Position Sensitive Detector acquisition** including **Time Of Flight)** have been particularly developed at the LLB for a maximum integration and simplicity of use. They form the core of the data acquisition equipment for any kind of neutron spectrometer.

For each function, an intelligent IEEE 488 microprocessor board has been designed. These boards constitute the Euro series (**EuroMove**, **EuroScale**r and **EuroPsd**), which are at present installed on the experiments. This triple function subsystem comes in the form of a single (see figure 2) or double Europe crate. It contains the location for the Euro series modules, power supplies for the electronic system as well as for the motors and the peripherals boards for motor control, readout and display of encoded axis and diverse I/O.

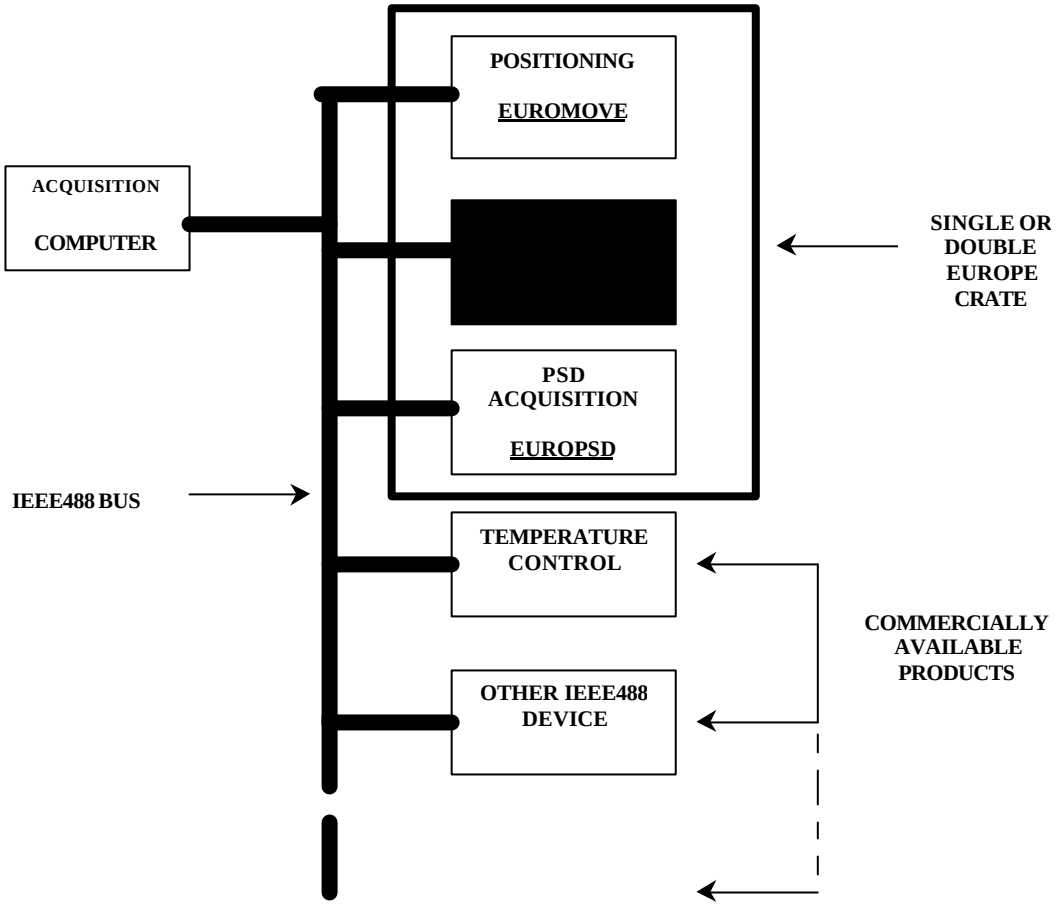

Figure 1. Outline description of the LLB Daffodil system.

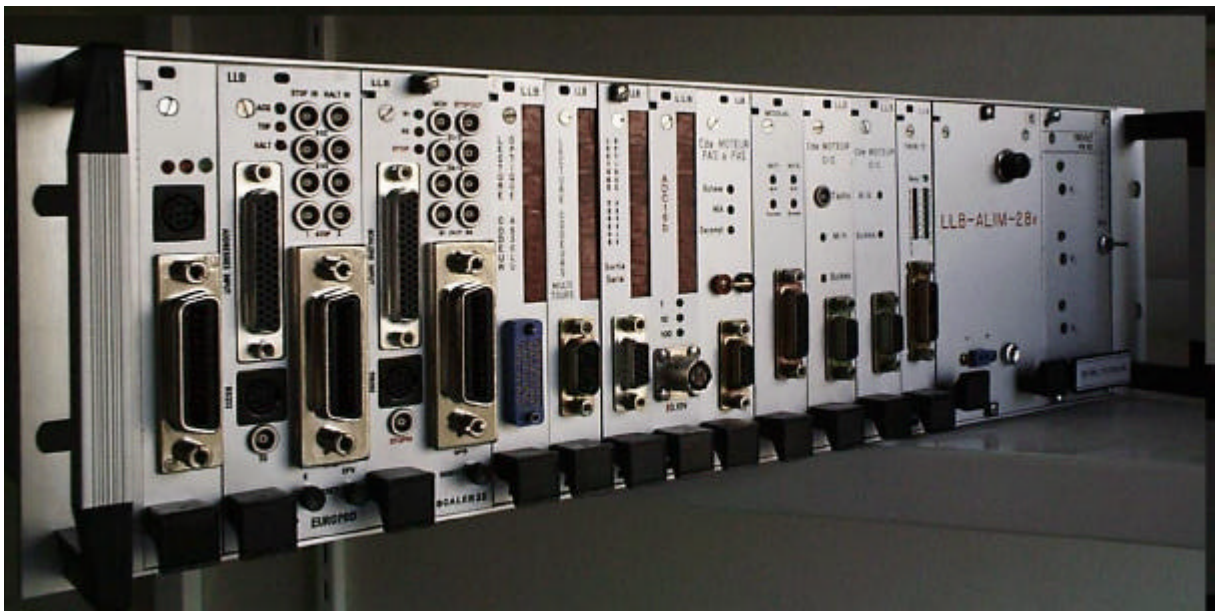

Figure 2. Single Europe crate for the Daffodil System of LLB

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THE EUROMOVE MODULE

The EuroMove module is a positioning controller. It is a single Europe board (100 x 220 mm^2), which can control **up to 10 axis simultaneously** through its own bus. This in-house design bus has been studied to allow on the one hand the mixing of power and logical signals and on the other, to provide a very simple and efficient interface to slave boards (encoder, motor, I/O boards etc.). Since the Europe crate contains the power supplies for the motors, **there is no need for another distinct power crate** nor additional connections.

The main role of the EuroMove module is to drive the motors through the motor boards, while monitoring the actual position of the mechanics via the encoder boards, to a position required by the computer. This module also provides a **real time display of the position** of each of the movements on the front panel of the encoder boards. The calculations are performed on 16 000 000 points, the displays showing 6 digits.

The onboard software embedded in the EuroMove module operates with different methods of positioning for each movement depending on its own encoder, motor and mechanic characteristics. In particular, it provides a **complete functional control of the pneumatic system** required by the « Tanzboden modules ».

Two types of boards, for D.C. and stepping motors as well as encoder boards for optical, magnetic and resistive sensor technologies have been designed. Thus, the user can choose the motorization and position encoding equipment that best fits his particular environment.

THE EUROSCALER MODULE

The EuroScaler module is basically a counting module for discrete detectors and monitors. Designed as a single Europe board, it is stand-alone and needs only the +5V to operate.

The Euroscaler module has an internal timer, a monitor counter and 32 scalers; it therefore can manage **a complete monitor preset or time preset operation on 32 detectors**. Moreover, it provides a **fully automatic acquisition mode for polarized neutrons** including up to 4 phases for polarized neutron analysis. The module outputs logical signals for driving the flippers. The duration for each of the phases and the dead time for the flipping of the magnetic field are programmable.

Input and output triggers for external synchronized systems like a EuroPsd module or another Euroscaler module are also available. For example, if there are more than 32 detectors on the experiment, several Euroscaler modules can be used, one of them will play the role of master and the others the role of slaves via their input triggers.

THE EUROPSD MODULE

For **data acquisition with fast Position Sensitive Detectors** (in particular those based on delay lines or microstrips), and to provide **Time Of Flight operation**, we have designed the **EuroPsd module** (see figure 3), which is stand-alone like the EuroScaler module.

The main function of the EuroPsd module is **histogramming memory**. It accepts a 18 bits to

accommodate a 512x512 cells PSD. Cells of the detector are associated **with 32 bits data counters**. One of the features of this module is the programming, through user friendly commands, of the allocation of the counters via a **routing memory**. This feature has many applications, for example, **converting a rectangular detector into a virtual circular detector**. Non adjacent cells can also be grouped together to form a multiparts « macrocell ».
In the Time Of Flight mode, the delay, the number of time channels (up to 4096) and their **width (200 nsec up to 7 minutes** by steps of 100 nsec) are programmable. Long duration time channels can be used in conjunction with another capability of the EuroPsd module, which is to follow and to distinguish several « frames » in a **kinetic experiment**.
With this EuroPsd module, the **maximum counting rate achievable is 5 MHz** with a null dead time, i.e. an address takes less than 200 nsec to be processed (routing, histogramming and T.O.F).

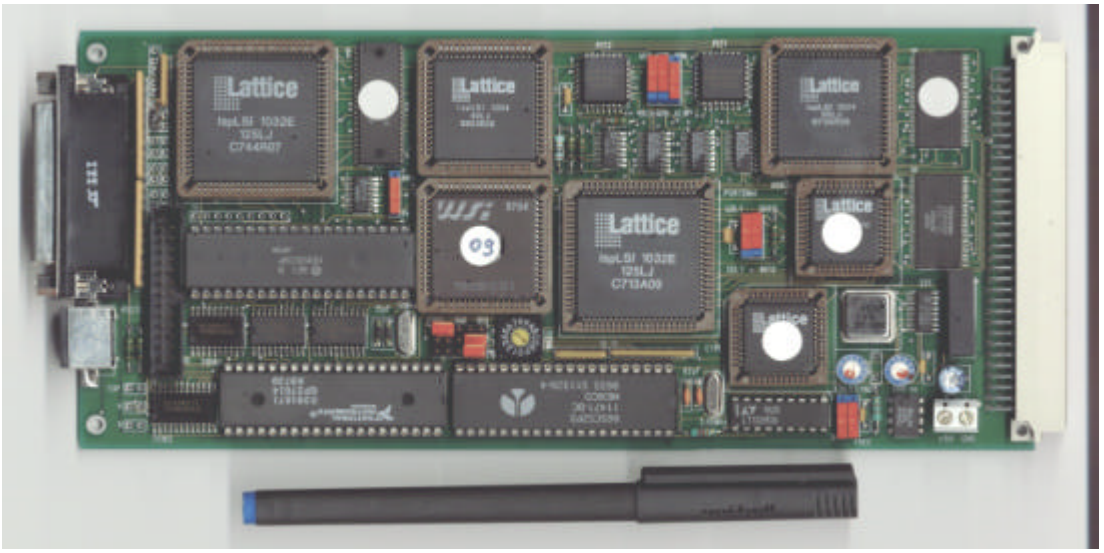

Figure 3. EuroPsd module for data acquisition and Time Of Flight

Several laboratories in the world are equipped with the LLB DAFFODIL system, among them K.F.K.I. (Hungary), I.T.N. (Portugal), Institute of Physics (China), Institute of Nuclear Technology (Greece), and more recently K.A.E.R.I. (Korea) in association with the CILAS company.

THE REPLACEMENT OF THE ORPHEE REACTOR'S HOUSING CORE

(SUMMER 1997)

The research reactor Orphée built by the CEA at Saclay in the late seventies went first critical in december 1980. Since then the reactor has provided the neutron beams used by physicists on the experimental spectrometers located around the reactor. During the summer 1997 the reactor was stopped for a major refurbishment : the remplacement of the housing core located in a heavy water tank.

This housing core, a square tube of 25 x 25 x 200 cm^3 is the backbone of the reactor as it contains the reactor core itself. It is the one barrier between the heavy water, from the tank, and the light water that cools the reactor core. The zircaloy 2 had been choosen for its characteristics both mechanical, close to those of the stainless steel, and physical (it does not interfere with the neutrons) to make the housing core and its 2 flanges. These two casted flanges are welded to both ends of the tube, and it is by them that the housing core is attached to the top and the bottom of the middle of the heavy water tank. The water tightness between the 2 types of water is done by two 0-ring metallic joints. The reactor core is supported by the grid core, located at the bottom of the housing core. The core is composed by eight fuel rods and one beryllium rod. So outside the tube there is the heavy water and inside it, there is the core itself cooldown by light water.

In reactor early life, year 1983, the metallic 0-ring upper joint, between the tank and the housing core, went to leak. So it has been replaced by a new one. This operation was done by removing the housing core from the heavy water tank and then the joint was changed by reputting it in its original place. The strain on the studs and nuts was increased to ensure the tighness of the system.

Meanwhile, it was obvious that the leak was due to the growth of the housing core that will, in the long run, lead to another leak. So it was decided to study the zircaloy growth by putting some samples inside the beryllium rod located in the middle of the reactor core itself. At the same time a new housing core was ordered.

The main difference between the two housing cores is that the new one is fitted at its top with a growth absorber made of a double stainless steel wall and flanges.

In the year 1995, it was foreseen, in the light of the study of the zircaloy, that the housing had to be changed by the turn of the century. It was then decided that it will be done in the summer 1997 to allow time to plan the operations. This planning was made by looking back in 1983 when the housing was first taken out to foresee the coming problems.

The main concerns were the four bottom studs and nuts as they had already given us troubles. It was decided to make some specific tools to destroy either the nut or, at worst possible case, the stud. The planning was done to start the refurbishment on the 15 july 1997.

The reactor was stopped on the 13, the core unloaded on the 14, and the heavy water replaced by light water on the 15. Before removing the housing core itself, several operations had to be done :

i) Remove the bars that link the control rods and their mechanism,
ii) Remove the rod trolley,
iii) Remove the loading-unloading anti-fault grid,
iv) Remove the core grid.

After that, it was possible to take out of the tank the housing core. The housing core was put in the canal to be disposed later. It was now possible to look inside the heavy water tank for an inspection done with a remote camera and also on the two 0-ring spans. It had shown that the two spans should be meticulously cleaned before we put back the new housing core.

Through out these tasks, the new housing core and all its parts were put together on a mock-up tank for a final check before it was put in its real location, the heavy water tank.

After all the checks, both on the span ring, on the tank and on the new housing with all its parts, it was then decided to put the new housing in place. When it was in place two tasks were done. First the three 0-ring joints were tested for water tightness. There is one more joint, the one between the zircaloy flange and the stainless steel of the growth absorber device. As the test gave us satisfaction, all the other parts were put back into place.

When all the operations have been completed a mock up core was loaded in the new housing core for hydraulic tests. When completed, we asked the French Safety Authorities for the green light to start the reactor. The reactor was then loaded with the fuel rods. We had the go ahead green light to start on the 27 october 1997.

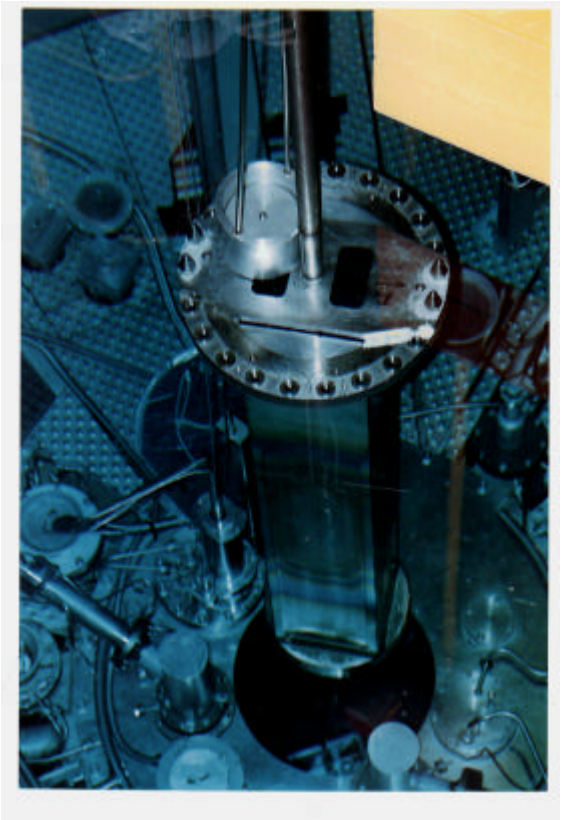

Figure 1. Removal under water of the old housing core

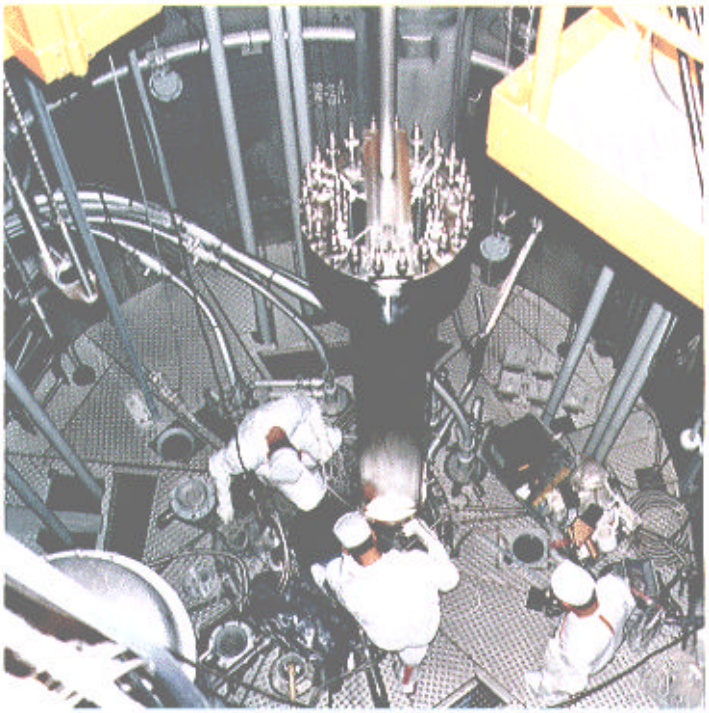

Figure 2. Installation of the new housing core