

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 $0.9 \cdot 10^7 \text{ n.cm}^{-2}\text{s}^{-1}$ at the sample position ($\lambda=4.8\text{Å}$, $\Delta\lambda/\lambda=0.15$).^{*}

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.

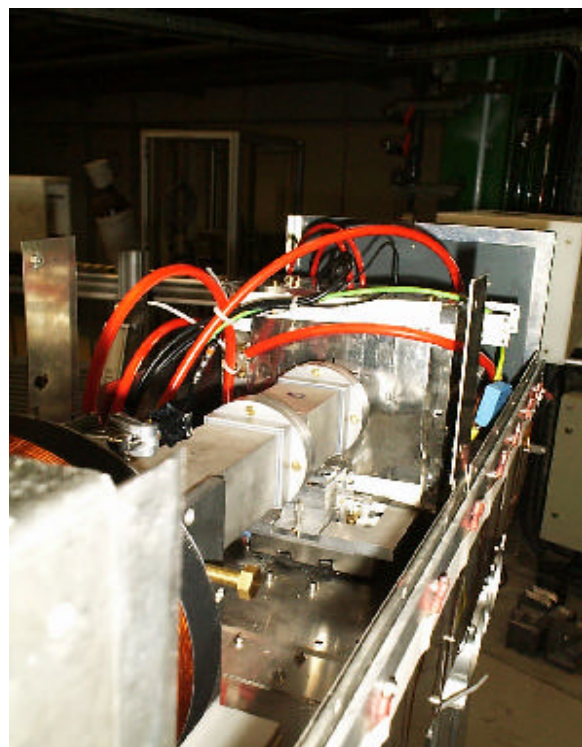


Figure 1. The NRSE instrument G1bis at Saclay

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 $\text{Ca}[(\text{NO}_3)_2]_{0.4}[\text{RbNO}_3]_{0.6}$. 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 $\text{Ca}[(\text{NO}_3)_2]_{0.4}[\text{KNO}_3]_{0.6}$, 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 $\text{Ca}[(\text{NO}_3)_2]_{0.4}[\text{RbNO}_3]_{0.6}$, 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 $Q=1.7\text{\AA}^{-1}$ are shown in figure 2.

The count rate was nearly $60\text{ neutrons.s}^{-1}$, 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.

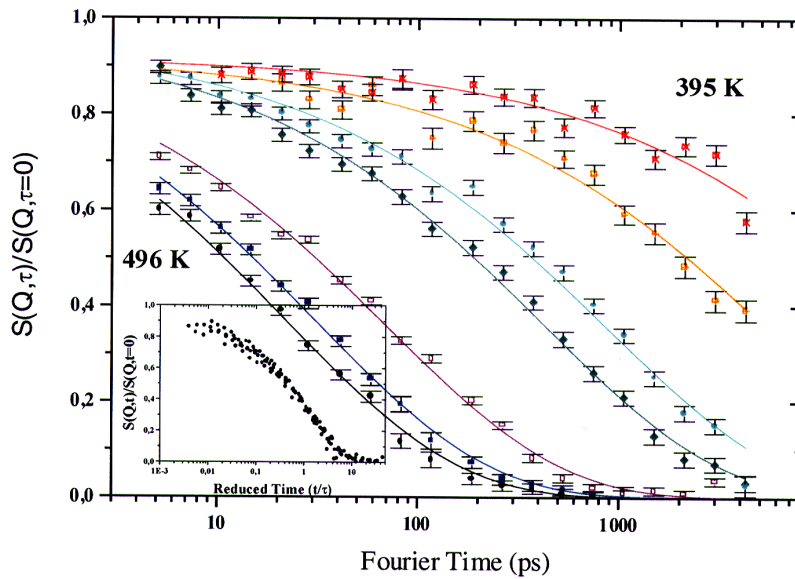


Figure 2 : Intermediate scattering function $S(Q,t)$ for $\text{Ca}[(\text{NO}_3)_2]_{0.4}[\text{KNO}_3]_{0.6}$ at $Q=1.7\text{\AA}^{-1}$ on the NRSE spectrometer G1bis.

References

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