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NEUTRON SPIN ECHO SPECTROSCOPY

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1. INTRODUCTION

Quasi-elastic neutron scattering has long been an invaluable tool for studying the complexities of very slow dynamical processes associated with stochastic phenomena as diverse as molecular rotation and tunnelling, diffusion, polymer reptation, magnetic relaxation and glassy dynamics. The characteristic time constants of such processes are typically in the range $10^{-12}\,\mathrm{s} < \tau < 10^{-6}\,\mathrm{s}$, and hence quasi-elastic neutron scattering studies must always be made at very small energy transfers with a correspondingly high energy resolution. However, whilst time-of-flight and triple-axis-based backscattering spectrometers provide the highest resolution currently available with conventional neutron instrumentation, this resolution, at best $\Delta E \sim 0.3\,\mu\mathrm{eV}$ with an incident neutron energy of ~ 2 meV, is unfortunately inadequate for many studies of dynamical processes for which characteristic time scales are longer than $\sim 10^{-10}$ s. This significant constraint on the dynamic range of quasi-elastic neutron scattering, and the associated implications for the investigation of slow dynamics in matter, is simply a consequence of the so-called Liouville theorem which, through conservation of the phase space density of neutron trajectories, strongly couples the effective neutron count rate of a conventional neutron spectrometer directly to its resolution. For an ideally optimized generic neutron spectrometer, the Liouville theorem shows that the count rate is at very best proportional to the square of the instrumental resolution. Therefore to extend the time window of a conventional backscattering spectrometer by the necessary three orders of magnitude would correspondingly reduce the effective count rate by six orders of magnitude. This is clearly an unacceptable sacrifice in what is an already severely intensity-limited experimental technique.

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Fortunately, in 1972, Mezei successfully demonstrated that it was possible to circumvent the restrictions imposed by the Liouville theorem by using an extremely ingenious and elegant technique known as neutron spin echo (NSE) [1]. By effectively decoupling instrumental energy resolution from incident neutron monochromatization, and hence from beam intensity, neutron spin echo offers gains of at least two orders of magnitude in resolution over conventional quasi-elastic scattering spectrometers. This gain is achieved not simply by producing a neutron beam with better defined incident energy and by analyzing more accurately the energy of the scattered neutrons, but instead by determining the relative number of Larmor precessions executed by spin polarized neutrons travelling through a well defined magnetic field before and after the scattering sample. By utilizing Larmor precession in this way each neutron effectively carries its own microscopic stop watch through which its change of velocity, and hence its change of energy, in the scattering process is uniquely and accurately defined.

In this chapter we shall consider the basic principles of the neutron spin echo technique, its practical realisation, and some of its recent applications in the study of slow dynamics and relaxation phenomena in physics and chemistry.

2. POLARIZED NEUTRON BEAMS, LARMOR PRECESSION AND SPIN FLIPPERS

As the Larmor precession of polarized neutrons lies at the very heart of the spin echo technique, it is perhaps worth reviewing some of the more fundamental properties of polarized neutron beams and the techniques employed in their manipulation.

The neutron is a fermion. It carries a spin 1/2 and an associated angular momentum of $\pm 1/2\hbar$ The spin angular momentum component operators of the neutron expressed in matrix form are:

$$s_x = \frac{\hbar}{2}\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \quad s_y = \frac{\hbar}{2}\begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} \quad s_z = \frac{\hbar}{2}\begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \qquad \{1\}$$

whilst the operators

$$\boldsymbol{\sigma} \equiv \frac{2}{\hbar}\mathbf{s} \qquad \{2\}$$

are known as the Pauli spin operators.

By virtue of its spin the neutron also has a magnetic moment, $\boldsymbol{\mu}_n$ with

$$\boldsymbol{\mu}_n = \gamma_n \mu_N \boldsymbol{\sigma} = 2\gamma_n \mu_N \mathbf{s}/\hbar = -9.66\times 10^{-27}\,\mathrm{J.T^{-1}} \qquad \{3\}$$