The Neutron Spin-Echo (NSE) technique achieves high energy resolution by encoding the neutron energy into a neutron spin Larmor precession angle. Polarized neutrons are sent through two identical magnetic fields (large precession coils) before and after the sample (see figure 1). At the sample, the atomic spins cause a $\pi$ spin flip. If the scattering process is strictly elastic, the precession angles in the two fields are identical, irrespective of the initial neutron velocity distribution, and the beam polarization is preserved. Small energy transfers lead to a change in the precession angle of the outgoing beam and, hence, to a decrease in the measured polarization. Neutron Spin Echo measures the spin-autocorrelation function as the intermediate scattering function $S(q,t)$. This is directly determined by scanning the magnetic fields in the coils and measuring the polarization (see figure 2).

The magnetic properties of "spin ice" Ho$_2$Ti$_2$O$_7$ and Dy$_2$Ti$_2$O$_7$ have been intensively studied recently because:
1) They are topologically analogous to crystalline water ice and show the same residual entropy at low temperature.
2) They were the first examples for geometrically frustrated ferromagnets.

Using a combination of neutron spin echo and ac-susceptibility two different types of dynamical processes have been revealed. Spin dynamics at high temperatures ($T > 15$ K) is due to a thermally activated single-ion process that is distinct from the process that dominates at lower-temperatures ($1 < T < 15$ K). The low-temperature process involves quantum spin tunneling, as quasi-classical channels of relaxation are exhausted in this temperature range.

We have also studied the extent to which magnetic voids assist in the spin relaxation, by partially substituting Ho with non-magnetic Y. At higher temperatures the thermally activated dynamics is unchanged and persists to low Ho concentration. At low temperature, $T < 50$ K, a new relaxation process appears which is more than 10 times faster than the main relaxation process, but has only a small spectral weight and does affect the quantum relaxation.

Figure 1: Schematic of the N65-NSE Spectrometer in magnetic setup with the Q-coils.

Figure 2: Raw data from Y-doped H$_2$Ti$_2$O$_7$, of first magnetic Echo measured at the NCNR.

Figure 3: The normalized $S(q,t)$ from several spin ice compounds. The pure compound (bottom) reveals a $q$-independent single relaxation process which does not completely freeze by 0.3 K. Introducing Y causes the system to freeze at elevated temperatures, reveals a classical to quantum phase transition and introduces a second relaxation process (left).

Figure 4: Measurements of a.c. susceptibility of Ho$_2$Ti$_2$O$_7$ in an applied magnetic d.c. field. Such measurements show two peaks in the imaginary part of the susceptibility. A peak at around 15 K has been mysterious for some time and was understood only after spin echo measurements [5]. Another peak at around 1 K marks the macroscopic spin ice freezing. The right panel in the figure shows the position of the 15 K peak.