

Study of slow dynamic processes in magnetic systems by neutron spin-echo spectroscopy

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TOPICAL REVIEW

Study of slow dynamic processes in magnetic systems by neutron spin-echo spectroscopy

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Online at stacks.iop.org/JPhysCM/18/R231**Abstract**

The use of the neutron spin echo (NSE) technique in the study of slow dynamic processes in magnetic systems is reviewed. NSE provides the highest energy resolution of all inelastic neutron scattering techniques, hence it is used mainly in studying systems that show very slow dynamics, which cannot be resolved otherwise with neutrons. For spin glasses, which show a dramatic slowing down of the spin dynamics in the vicinity of T_g , research using NSE has made significant contributions to our understanding of the nature of the phase transition and the line shape of the spin relaxation function. In the study of the critical dynamics in magnets, NSE has proven predictions of dynamic scaling theory to be correct. Geometrically frustrated magnets and their unusual properties at low temperature have seen an upsurge of interest in recent years, and NSE has been able to make some key contributions to this field. The case of $\text{Gd}_2\text{Ti}_2\text{O}_7$ is presented in some detail, a system in which ordered spins and paramagnetic spins coexist.

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

Neutron spin echo (NSE) is a particular neutron scattering technique that uses the spin precession of polarized neutrons in a magnetic field to achieve very high resolution for measuring the neutron energy transfer in the scattering process. The method was invented by Mezei in 1972 [1]. The main applications of the technique are found in quasielastic scattering experiments, to study relaxation processes which are centred around zero energy transfer. Contrary to other inelastic neutron scattering techniques, such as triple axis spectroscopy (TAS) or time of flight spectroscopy (TOF), NSE measures the intermediate scattering function $S(Q, t)$ in reciprocal space and time. The accessible time window spans roughly from 10^{-12} to 10^{-7} s, covering the range from the microscopic timescale of atomic collisions and spin exchange to macroscopic times of slow relaxation processes. Compared to other experimental techniques that provide dynamic information, such as μ SR, NMR, ESR, Mößbauer spectroscopy, or ac susceptibility, it is the simultaneous access to spatial information (via Q) that sets inelastic neutron scattering and NSE apart. The mentioned microscopic techniques probe local behaviour at $R = 0$ (that is, one averages over all Q 's), whereas bulk techniques measure averages over macroscopic sample volumes, corresponding to $Q = 0$.

The first spin echo spectrometer was the IN11 instrument at the Institute Laue–Langevin (ILL), which became operational around 1978. Since that time, a constant stream of publications in the scientific literature has shown the strength and popularity of the technique.

The most comprehensive references are the neutron spin echo books which encompass all technical aspects and a wide range of applications in different scientific areas [2, 3]. More specialized review articles on the spin echo method are also available, with emphasis on applications in soft [4, 5] and hard [6, 7] condensed matter physics.

2. Experimental details

A technical description of a spin echo spectrometer will not be given here, but can be found elsewhere [2, 3]. In this section only a few specific aspects will be mentioned that become important at one point or the other.

NSE is rather unconventional for a neutron technique, in the sense that Q - and energy-resolutions are completely decoupled and one does not trade beam intensity for energy resolution. This is a consequence of how the information on the energy transfer of the scattering process is obtained. The energies of the ingoing and scattered neutrons are not separately determined, but the energy transfer is measured in one step via a neutron spin precession angle. The obtained resolution depends on the strength of the magnetic precession fields, and here the limits are simply given by what fields can be handled technically. Performing a spin echo in the earth's magnetic field will roughly provide cold triple-axis type energy resolution, and with a field of a few tenths of a tesla at the other extreme the resolution can be 10^4 – 10^5 times better.

In NSE one is not too concerned with energy transfer, because one directly measures the intermediate scattering function $S(Q, t)$, which is the time Fourier transform of the usual scattering function $S(Q, \omega)$. Measuring a Fourier transform has the important consequence that the correction for instrumental resolution is (mathematically) very simple. This is done by dividing the data of a scan by a reference scan measured with an elastic sample. Therefore, NSE has a crucial advantage over other neutron techniques when one needs to determine the line shape of a relaxation function in the time domain. With magnetic systems one is often (not always) in the fortunate situation that one can use the sample itself as elastic reference, provided it can be cooled to a temperature low enough to freeze out all dynamic processes in the measurement window.

Another strength of the method lies in the large dynamic range. The magnetic precession field can typically be changed by three to four orders of magnitude in a single scan. Such a large range has proven essential in fitting the correct model to many experimental data sets.

In NSE one is able to use a beam with coarse $\Delta\lambda/\lambda \sim 15\%$ monochromatization, because the echo principle holds for all wavelengths simultaneously. This means not only a coarse resolution in Q , but also implies a $\sim 45\%$ spread of $\Delta t/t$ in a measurement, because $t \propto \lambda^3$ [2, 3]. This is not a real problem as long as the wavelength dependences of Q and t in $S(Q, t)$ partly cancel each other (for a modified diffusion process with a stretched exponential relaxation $S(Q, t) \sim \exp(-DQ^2t^{2/3})$ the cancellation is exact). However, sharp features in the dependence of correlation times in the sample with Q are likely to create problems in the data analysis. The relative spread $\Delta t/t$ in a measurement is constant, and therefore it does not set a limit to the highest attainable energy resolution (equivalent to the longest meaningful t).

In NSE experiments with magnetic samples one almost always employs three-directional neutron polarization analysis (often referred to as xyz polarization analysis) to measure the magnetic part of the static structure factor $S(Q)$ [8, 9]. To do this the instrument is run in polarized diffraction mode without echo but with a π flipper. One measures six cross sections with the magnetic field along three axes x , y , and z ('up' and 'down' for each). Certain linear combinations of these cross sections cancel the nuclear coherent and incoherent scattering contributions, thus allowing the separation of the purely magnetic scattering. The echo amplitude, which itself is also purely due to magnetic scattering, is then normalized to the magnetic $S(Q)$. Since one uses polarized neutrons, one always identifies the magnetic scattering unambiguously. For example, in the studies of the critical dynamics in ferromagnets (see section 4) this was very important because of intense nuclear background scattering at small angles.

3. Spin glasses

A spin glass is the magnetic state of a system in which competing nearest neighbour interactions created by disorder lead to freezing of the magnetic moments in random directions at low temperature. The spin glass state is furthermore characterized by strong history dependence and slow recovery of equilibrium after external perturbation. One of the main reasons why spin glasses have attracted interest for so many years is that their properties are seen as fairly universal and applicable concepts are relevant for other scientific areas (biology and evolution, for example). Experimentally, spin glasses may be identified by the following signatures. In ac susceptibility a peak or 'cusp' is observed in low magnetic fields at a certain temperature T_g which depends on the frequency. The magnetization below T_g shows a strong history dependence and differences between field-cooled and zero-field-cooled measurements. A slow relaxation exists in the remanent magnetization. Neutron diffraction and specific heat reveal the absence of long range magnetic order, but a broad feature in specific heat may exist above T_g , and neutron diffraction generally shows short range order. In the archetypical spin glasses such as CuMn, AuFe, or $\text{Eu}_x\text{Sr}_{1-x}\text{S}$, the magnetic species resides in random sites in the host lattice, and the particular freezing phenomena are believed to result from the simultaneous presence of ferro- and antiferromagnetic couplings between the spins. Extensive reviews on spin glasses can be found in the literature [10–14].

Since the spin dynamics is one of the keys to understanding spin glasses, it is not surprising that NSE has been used in numerous experimental studies since it was invented [15–27]. The early experiments have had a strong influence on the development of the whole field.

It should be noted that purely antiferromagnetic interactions in structurally well ordered systems may also lead to spin glass behaviour, when these interactions are geometrically frustrated. Examples for this behaviour are found in the systems $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$, $\text{Y}_2\text{Mo}_2\text{O}_7$, or CsNiCrF_6 , which will be discussed in section 5.

3.1. The dynamics above the freezing temperature T_g

In the study of the high temperature dynamics in spin glasses, neutron scattering in general and NSE in particular have revealed that (i) the paramagnetic phase is already anomalous well above the freezing temperature T_g , showing non-exponential relaxation, and (ii) there is a dramatic slowing down of the dynamics when temperature is decreased below $\sim 1.5 T_g$. For the following discussion it is convenient to write the intermediate scattering function as the product

$$S(Q, t) = S(Q) \cdot s(Q, t) \quad (1)$$

where $S(Q)$ is the static structure factor and $s(Q, t = 0) = 1$. Notice that both $S(Q)$ and $s(Q, t)$ are simultaneously measured in an NSE experiment. NSE shows that in the temperature range as high as $3\text{--}10 T_g$ the spin relaxation $s(Q, t) \equiv s(t)$ is generally wavenumber independent but strongly non-exponential in time. The conclusion first reached from this observation was that a broad distribution of individual (exponential) relaxation processes is present in the system above T_g :

$$s(t) = \int f(\tau) \exp(-t/\tau) d\tau, \quad (2)$$

where $f(\tau)$ describes the distribution [28]. For example, such a distribution could be due to spatial inhomogeneities, that is, different activation energies and relaxation times for different volume elements of the sample. After all, spin glasses are structurally not ordered and the individual magnetic ions reside in different environments. For example, Mezei discussed a distribution of relaxation times τ as a result of the presence of thermally active Arrhenius processes with different activation energies [18]. A constant distribution of energies between zero and E_{\max} leads to

$$s(t) = \frac{1}{E_{\max}} \cdot \int_0^{E_{\max}} \exp\left[\frac{-t}{\tau_0 \exp(E/k_B T)}\right] dE, \quad (3)$$

which has been used to fit the data in the upper panel of figure 1 with $E = 80$ K and $\tau_0 \sim 4.5 \times 10^{-12}$ s [18]. However, in such a situation it is not intuitive to understand the length scale (wavenumber) independence of the relaxation dynamics, because such inhomogeneities would likely introduce some correlation between size and relaxation time. Furthermore, comparisons of NSE and μSR experiments by Uemura *et al* have provided evidence in favour of spatially homogeneous dynamics [29]. Therefore a more likely explanation is that each spin has a spectrum of relaxation channels, and that the dynamics is intrinsically non-exponential.

To determine the exact functional form of the relaxation function, a model of hierarchically constrained dynamics may be applied to justify the stretched exponential ('Kohlrausch') form

$$s(t) = \exp\left[-(t/\tau)^\beta\right], \quad (4)$$

where $0 < \beta \leq 1$ [30]. The basic assumption in this model is that the magnetic moments interact and cannot relax independently; more specifically, their degrees of freedom are locked and a given group must adopt a certain configuration before a subset can relax. Another model was proposed by Weron [31], which assumes more generally that individual moments and their environment do not remain independent during relaxation. Then the time necessary for a single

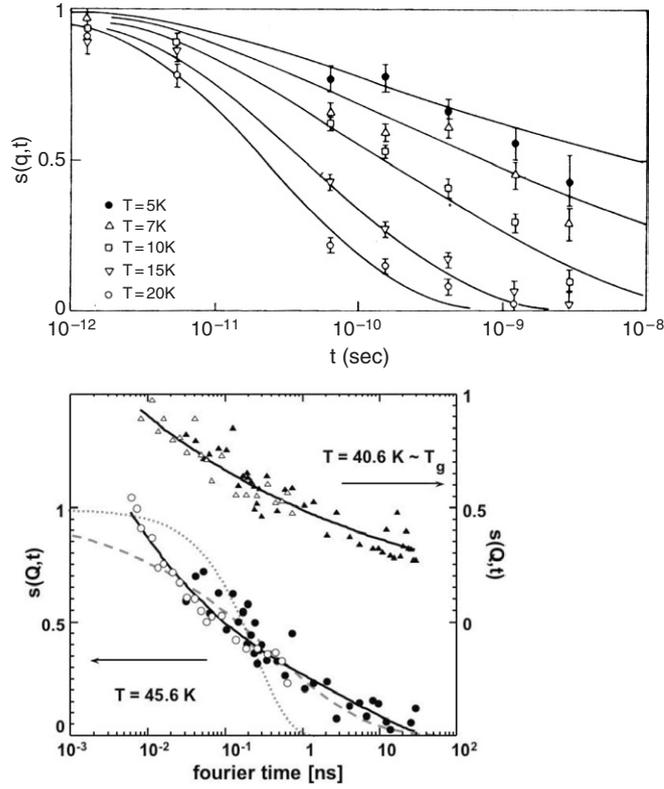


Figure 1. An early and a more recent example for line shape studies above T_g . Top: spin relaxation in $\text{La}_{0.7}\text{Er}_{0.3}\text{Al}_2$ ($T_g \sim 3.5\text{ K}$) at various temperatures, measured at IN11 (ILL). The lines are fits to a model discussed in the text (figure from [20]). Bottom: NSE spectra of $\text{Au}_{0.86}\text{Fe}_{0.14}$ spin glass ($T_g \sim 40.6\text{ K}$) at $Q = 0.08\text{ \AA}^{-1}$ measured at IN15 (ILL, full symbols) and at SPAN (HMI, Berlin, open symbols). The continuous line through the 45.6 K data is a fit to an Ogielski function (see text). The dashed and dotted curves correspond to a stretched exponential and a simple exponential decay, respectively. The data at 40.6 K are fitted to a simple power law (figure from [27]).

relaxation process depends stochastically on two other random variables: the dissipation rate and a waiting time. This model leads to a functional form of the relaxation

$$s(t) = [1 + k(t/\tau)^\beta]^{-1/k}, \quad (5)$$

where k is a measure of the interaction strength. The stretched exponential form is recovered in the weak interaction limit ($k \rightarrow 0$). In a different approach, Ogielski has studied the dynamics of spin glasses in Monte Carlo simulations [32]. Above T_g his results were best described by an empirical form

$$s(t) = t^{-x} \cdot \exp[-(t/\tau)^\beta], \quad (6)$$

where $\beta = 1/3$ at T_g , and increasing as T increases. At T_g and below he found a pure power law, which is also consistent with dynamic scaling theories which predict a pure power law form of $s(t)$ at T_g [27].

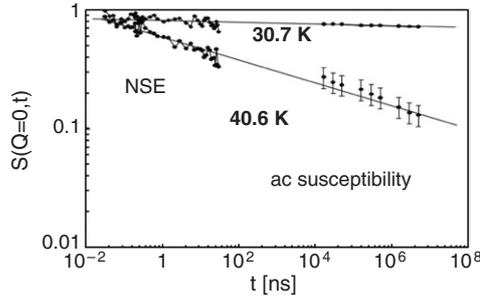


Figure 2. NSE spectra combined with $s(Q = 0, t)$ values deduced from macroscopic ac susceptibility measurements below and around T_g . The sample was a $\text{Au}_{0.86}\text{Fe}_{0.14}$ spin glass ($T_g \sim 40.6$ K). In the close vicinity of T_g the pure power law decay of $s(Q, t)$ holds over an impressively large dynamic range of more than nine orders of magnitude in time. Figure from [27].

3.2. The dynamics below the freezing temperature T_g

In the study of the low temperature spin glass phase, it has been experimentally verified that at T_g the line shape of the relaxation function changes and becomes a power law, thus confirming Ogielski's simulations and the scaling argument. Using the notation from equation (1) above one can combine NSE data with high frequency susceptibility by [17, 19]:

$$\chi(\nu) \propto \lim_{Q \rightarrow 0} \frac{S(Q)}{kT} [1 - s(Q, t)], \quad (7)$$

where $t = 0.7/2\pi\nu$, and thus increase the dynamic range of NSE spectra considerably. Equation (7) expresses that, at any given frequency, the observed susceptibility response corresponds to the fraction of the time dependent correlations which can relax within the corresponding time interval. This relation allows one to nicely bridge neutron scattering and ac susceptibility results. An example of such an analysis was recently given by Pappas *et al* (see figure 2). The combination of NSE and high frequency susceptibility data provides a very large dynamic range (nine orders of magnitude in this case), over which the power law decay of $s(Q, t)$ is observed at T_g and below. The power law decay was also found by re-analysing older $\text{CuMn}(5\%)$ data, which were combined with macroscopic dynamic (ac) susceptibility measurements and covered nine orders of magnitude in time [19]. These results constitute the most direct evidence for a phase transition in spin glasses at T_g , which in turn implies that the non-exponential temporal relaxation observed at T_g is an intrinsic, homogeneous feature of the phase transition. This strengthens the argument for the homogeneity of the non-exponential relaxation in spin glasses suggested by Uemura *et al* [29].

Equation (7) can also be used to demonstrate that the cusp in ac susceptibility is dynamic in origin. As neutron scattering shows, in CuMn the structure factor $S(Q)$ has no anomaly when the sample is cooled through T_g ; hence, all changes in $\chi(\nu)$ are due to changes in $s(Q, t)$. On the other hand, in the spin glass $\text{La}_{0.7}\text{Er}_{0.3}\text{Al}_2$ the cusp is a result of a competition of both terms [19]. In this compound $S(Q)$ changes very rapidly below $T = 10$ K due to the build-up of ferromagnetic short ranged correlations.

An alternative conclusion was reached by Heffner *et al* [33], who compared NSE and μSR data and concluded that the observed relaxation function in CuMn above and below T_g can be fitted to a modified power law $s(t) \sim t^{-\nu} + \text{const}$, with a non-zero constant only below T_g which would imply static ordering. Clearly, the NSE data available at the time were not good

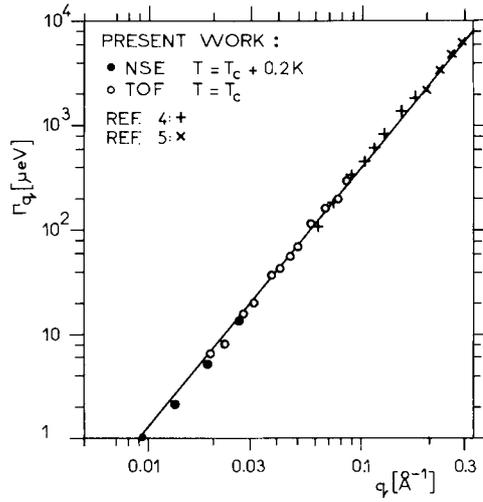


Figure 3. Critical dynamics in Fe. Results from different experiments of the wavenumber dependence of the spin relaxation rate in Fe at T_C . Figure from [39].

enough to identify the line shape of $S(Q, t)$ unambiguously. The problem of the ‘true’ line shape is still under debate today, with more and better data becoming available.

4. Critical dynamics in isotropic ferromagnets

The critical dynamics in magnets has been studied experimentally and theoretically, with activity peaking in the 1970s and 1980s. Neutron scattering (NSE in particular) is well adapted to study the critical fluctuations in the vicinity of T_C because one gets access to space and time information simultaneously. As T_C is approached, the fluctuations become increasingly slower and larger in scale. Therefore one needs cold neutrons to access low Q and the best possible energy resolution to get as close to T_C as possible. Dynamic scaling theory predicts that the critical scattering yields the same line shape at all Q , and that at $T = T_C$ the energy line width Γ , which is inversely proportional to the lifetime τ of the fluctuations ($\Gamma = \hbar/\tau$), scales as

$$\Gamma = A \cdot Q^z, \quad z = (5 - \eta)/2, \quad (8)$$

where A is a constant and the Fisher exponent, η , is a small positive number, $\eta < 0.1$ [34]. As one approaches T_C , one expects a cross-over from exchange interaction to dipolar interaction between magnetic moments as the leading energy term. Such a cross-over was expected by theory to correspond to a change of the critical exponent to $z = 2$, which would occur at a certain critical wavenumber ($Q_d = 0.045 \text{ \AA}^{-1}$ for Fe, $Q_d = 0.147 \text{ \AA}^{-1}$ for EuO). In addition, there was experimental evidence for such a cross-over [35].

Making use of the previously unavailable energy resolution of NSE, Mezei was able to extend other neutron scattering studies [36–38] of the critical fluctuations in Fe and EuO significantly towards lower Q and longer fluctuation times [39–41]. He showed that the expected cross-over was not observable with neutrons in either Fe or EuO (see figure 3). In conclusion, it became apparent that isotropic ferromagnets have a common dynamic scaling function, but that in a certain (Q, T) range the scaling is masked by the dipolar interaction, that is, by the self-interaction of the fluctuations [7].

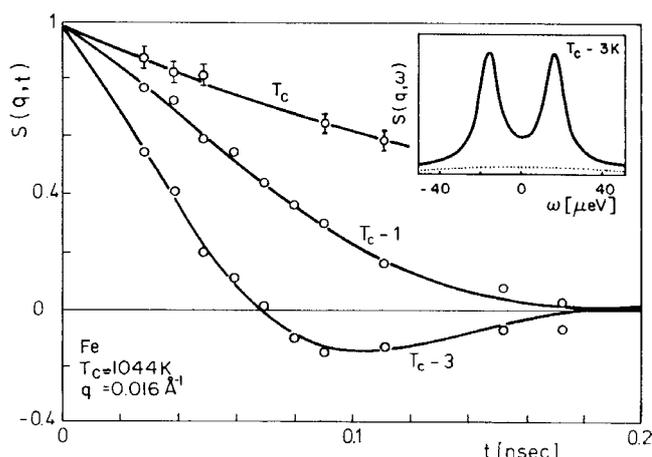


Figure 4. Critical scattering in Fe measured by IMNSE. At $T < T_C$ the lines correspond to fits to standard magnon theory including dipolar interaction. At T_C the fit is to a diffuse Lorentzian peak. The inset shows $S(Q, \omega)$ corresponding to the $T_C - 3$ K curve. Figure from [44].

NSE also revealed that, at T_C , the line shape of the fluctuations in time is a simple single exponential [41]. This result was contrary to theoretical calculations which predicted a different line shape [42, 43]. In these theoretical studies the spin correlation function was calculated using a simplified version of mode-coupling theory.

Ferromagnets present a challenge in that the neutron depolarization usually occurring in the ferromagnetic phase destroys the echo signal. Therefore, Mezei's original paper reported the measurement in Fe at $T_C + 0.2$ K [39]. A potential way out of this problem was presented in a study by Farago *et al* of magnon dynamics in Fe near T_C [44] (see figure 4). In their experimental set-up, called *intensity modulated* neutron spin echo (IMNSE), the authors used separate polarizer/analyser pairs in both spectrometer arms and two $\pi/2$ flippers on each side. Thus the number of precessions can still be compared for the two sides, and the echo principle remains the same. At the sample, however, the number of precessions in the first spectrometer arm is coded into intensity (by virtue of the spin analyser in front of the sample) and not into precession angle. Thus one is able to make the measurement even if the beam polarization is destroyed in the sample, but one pays a severe intensity penalty. This is probably the reason why IMNSE has not been used more often.

A more recent study of the spin dynamics near T_C in materials showing colossal magnetoresistance was reported by Heffner *et al* [45]. In the ferromagnetic manganite $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$, $x \simeq 0.3$, the authors found at least two types of spin relaxation processes suggesting spatial and temporal inhomogeneities in the Mn spin dynamics.

5. Geometrically frustrated magnets

Unlike common spin glasses like CuMn or AuFe, which are disordered, and where the distribution of distances between magnetic ions leads to competing interactions, geometrically frustrated magnets are structurally well ordered.

Still, a situation may arise in which the regular spatial arrangement of the spins in a lattice is incompatible with their couplings, that is, a situation in which the spins cannot simultaneously satisfy all their pair-wise interactions. The 'typical ingredients' for this are magnetic moments with antiferromagnetic nearest neighbour interactions, residing on a lattice

of triangles, such as the edge-sharing triangular or *Kagomé* lattices [46, 47], or on lattices involving tetrahedra such as the face-centred cubic and the pyrochlore lattices [48]. However, antiferromagnetism is not a strictly necessary condition as the discovery of ‘spin ice’ has shown [49]. Frustrated magnetism is a popular field of current condensed matter research, and reviews on the topic can be found in the literature [50–54].

Typically, when a geometrically frustrated magnet is cooled to very low temperature, the spins find it ‘difficult’ to form an ordered ground state. It is often observed that, if the system settles into an ordered state, the phase transition occurs only at a temperature much lower than the Curie–Weiss temperature (which gives the scale of the magnetic interaction energies). In fact, the ratio of the phase transition temperature and the Curie–Weiss temperature has been established as an indicator for the degree of frustration in a system [52]. However, one may also observe that such a system enters either a spin-glass-like state (see below), or remains dynamic even at the lowest attainable temperature such as $\text{Tb}_2\text{Ti}_2\text{O}_7$ [55, 56]. One often finds a fragile balance between the different magnetic terms in the Hamiltonian (exchange, dipolar and anisotropy energies), and the resulting ground state at low temperature can have a large degeneracy. Another general signature of frustration is a shift towards low energy of the spectral weight of excitations in the system. These slow low-energy excitations are often in the NSE measurement window [57].

As an example, the case of $\text{Gd}_2\text{Ti}_2\text{O}_7$ is presented here in some detail. It shows a very unusual magnetic state in which ordered and paramagnetic moments of the same magnetic species (Gd in this case) coexist that reside on equivalent positions in the lattice. Thus one can assign different ‘degrees of frustration’ to different sublattices. Other examples of such magnets are Mn_2P [58], UNi_4B [59], CePdAl [60], and TbNiAl [61].

$\text{Gd}_2\text{Ti}_2\text{O}_7$ belongs to the cubic pyrochlore family (lattice constant $a = 10.18(1)$ Å at 300 K). The Gd^{3+} spins ($S = 7/2$) reside on a pyrochlore lattice (see the inset in figure 5) of corner sharing tetrahedra and possess dominant near neighbour antiferromagnetic couplings. $\text{Gd}_2\text{Ti}_2\text{O}_7$ has two magnetic phase transitions at $T_N = 1.1$ K and $T_0 = 0.7$ K. Neutron diffraction revealed the unusual nature of the spin ordering [62, 63]. In the temperature range between T_0 and T_N three out of four spins order in a $4-k$ antiferromagnetic structure while the other spins remain completely paramagnetic. This is linked to the absence of two key magnetic Bragg peaks, the $(1/2, 1/2, 1/2)$ and the $(5/2, 3/2, 3/2)$. While cooling through T_0 , the previously ordered spins (ordered moment $7.0 \pm 0.1 \mu_B$) slightly tilt away from their easy directions, and the previously unordered spins acquire a small ordered moment ($1.9 \pm 0.1 \mu_B$ at $T = 250$ mK) but retain a large paramagnetic component, the dynamics of which persists down to the lowest attainable temperature. Reverse Monte Carlo simulations were used to show that the spatial arrangement of the paramagnetic moments does indeed yield a structure factor for the diffuse scattering that is consistent with the experiment. In a spin echo experiment performed at IN11 these results from neutron diffraction were directly confirmed (see figure 6). The diffuse scattering (see figure 5) was shown to be dynamic and the level of $S(Q, t) \sim 0.75$ at $T = 100$ mK compared well to the relative intensities of the Bragg scattering (static) and the diffuse scattering (dynamic) in this Q -range. The persistent spin dynamics was also confirmed in a μSR experiment [64].

The ‘spin ices’ $\text{Ho}_2\text{Ti}_2\text{O}_7$ and $\text{Dy}_2\text{Ti}_2\text{O}_7$ are two other well studied members of the rare earth titanate family $\text{R}_2\text{Ti}_2\text{O}_7$ [49, 65]. These systems have already been extensively reviewed [66, 67]. Here it may suffice to mention that by combining NSE with ac susceptibility results, it was possible to identify a cross-over temperature $T_0 \sim 16$ K, well above the freezing which occurs at $T_f \sim 1$ K, at which the nature of the spin dynamics changes [68]. Susceptibility alone could tell that there is an anomaly at T_0 , but its nature could only be revealed with the help of the spatial information from neutron scattering (in this case, through

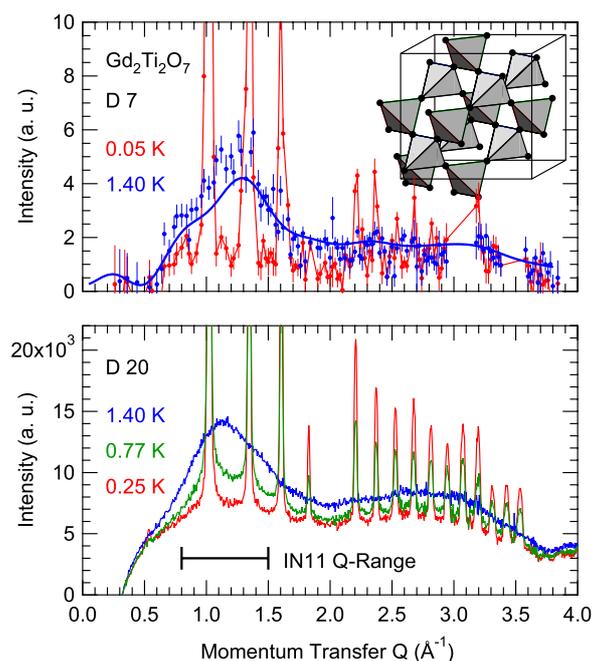


Figure 5. Magnetic powder diffraction data of $\text{Gd}_2\text{Ti}_2\text{O}_7$, measured using polarized neutrons (D7, top panel) and unpolarized neutrons (D20, bottom panel) at ILL. The top panel shows the magnetic part of the scattering, which is separated using polarization analysis, and also shows the fit resulting from reverse Monte Carlo simulation at 1.4 K (see text). In the bottom panel, a diffraction pattern measured at higher temperature is subtracted, hence all peaks shown are magnetic. Also shown is the Q -range of the spin echo experiment at IN11. The inset shows the pyrochlore structure.

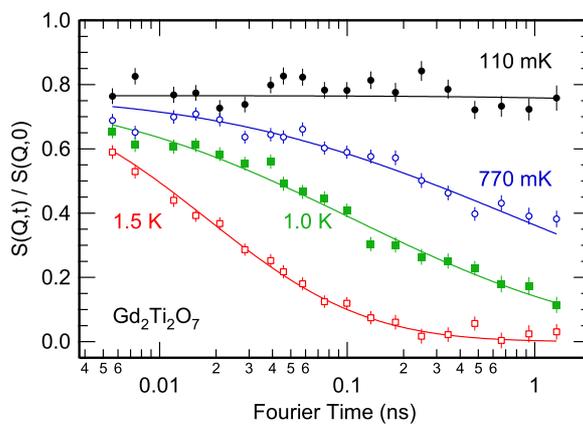


Figure 6. Neutron spin echo scans of $\text{Gd}_2\text{Ti}_2\text{O}_7$, averaged over the Q -range shown in figure 5. Lines are guides to the eye.

the absence of any Q -dependence of the scattering and the simple exponential line shape of $S(Q, t)$. At temperatures above T_0 , the dynamics consists of thermally activated single spin flips between the two states of the ground state doublet of the Ho^{3+} or Dy^{3+} ions. No indications for collective spin dynamics were found. On cooling below $T < T_0$ the dynamics

becomes independent of temperature, and it was suggested that in the lower temperature regime a quantum relaxation process is involved originating from the slowly fluctuating dipolar magnetic field.

As was mentioned earlier, geometrical frustration can lead to spin-glass-like freezing of the magnetic moments. Examples of such systems include $\text{SrCr}_{8-x}\text{Ga}_{4+x}\text{O}_{19}$ (SCGO) [69–72], $\text{Y}_2\text{Mo}_2\text{O}_7$ (YMoO) [73, 74] and CsNiCrF_6 [75]. Strictly speaking, they all possess some site disorder—in SCGO the Cr sublattice population is 89% at $x = 0$, and in YMoO a local distortion of the Mo tetrahedra was found on the 5% level using x-ray-absorption fine-structure measurements [76]. One may consider this site disorder as the nucleus of the particular spin-glass-like freezing in these materials, but nevertheless it is justified to view the frustration as arriving from the more or less regular arrangement of the magnetic moments on *Kagomé* (SCGO) or pyrochlore (YMoO) lattices. These systems differ from classical spin glasses in several of their properties, for example a quadratic T^2 dependence of the specific heat of SCGO below T_g , and a very rapidly vanishing spin relaxation in YMoO below T_g .

6. Outlook and summary

We have seen that NSE opens up very rich possibilities for research on the magnetic dynamics in a large variety of different systems. For the next decade or so it may be anticipated that the use of NSE will be much more extended to the study of new types of materials, such as magnetic nanoparticles [77–79], ferrofluids [80, 81], molecular magnets [82], random anisotropy magnets [83], or quantum magnets [84]. Nowadays molecular magnets such as ‘ Mn_{12} ’ cannot be studied with NSE because these materials are very dilute and the comparatively strong nuclear scattering by hydrogen or deuterium makes the signal to noise ratio very unfavourable. Progress in instrumentation [85, 86] and the advent of spallation neutron sources such as the SNS [87] will in the future make inelastic neutron experiments possible that previously failed due to limited intensity. There are certainly many promising systems available that warrant exciting experiments.

In the domain of frustrated magnets, a couple of new systems with very unconventional spin dynamics have been identified in recent years, for example $\text{Yb}_2\text{Ti}_2\text{O}_7$ [88, 89], or $\text{Gd}_2\text{Sn}_2\text{O}_7$ [90]. Studies will certainly also be extended to other families of compounds, such as the jarosites [91]. Introducing non-magnetic impurities often has important effects in geometrically frustrated magnets. In the context of pyrochlore physics, such a study using NSE has been published by Keren *et al* for the $\text{Tb}_{2-x}\text{Y}_x\text{Ti}_2\text{O}_7$ system [92].

It may also be expected that more integrated studies will be performed where NSE is combined with other methods. Experimental techniques providing microscopic dynamic information that come to mind are NMR, ESR, μSR and Mößbauer spectroscopy. These are local probes that have no access to Q information, but offer the possibility to study small and dilute samples because the intensity limitations are not nearly as severe as for neutrons. This is especially true for magnetic studies of systems in which Gd, Eu, Sm, or Dy occur, which all have naturally abundant isotopes with huge neutron absorption cross sections. In addition, by combining different techniques one gains access to a wider dynamic range in which spin relaxation processes can be observed.

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