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μSR investigation of spin dynamics in the spin-ice material $Dy_2Ti_2O_7$

J Lago[1], **S J Blundell**[2] **and C Baines**[3]

[1] Dipartimento di Fisica *A Volta*, Universitá di Pavia, I27100 Pavia, Italy
[2] Clarendon Laboratory, Department of Physics, University of Oxford, Oxford OX1 3PU, UK
[3] Paul Scherrer Institute, Laboratory for Muon Spin Spectroscopy, Villigen, CH-5232, Switzerland

E-mail: lago@fisicavolta.unipv.it

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Abstract
We present a detailed muon spin relaxation (μ^+SR) study of the spin-ice material $Dy_2Ti_2O_7$. Polycrystalline samples of this material have been studied in the temperature range $0.02\ \mathrm{K} < T < 300\ \mathrm{K}$ in applied longitudinal magnetic fields of up to 2.7 T. At high temperature our study confirms previous bulk results showing that dynamics are dominated by spin-phonon-mediated relaxation of the electronic spin system. Below about 60 K the depolarization of the muon ensemble becomes too fast and the signal is almost completely lost. However, the recovery at lower temperature of the resolved asymmetry to roughly 1/3 of the total value allows us to confirm that it is the development of strong quasi-static local fields and not only dynamical effects that causes this depolarization. Finally, we find evidence of remanent spin fluctuations at temperatures as low as 0.02 K, well inside the quenched spin-ice state. We tentatively ascribe these to the hyperfine interaction with the ^{161}Dy and ^{163}Dy nuclear spins.

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

1. Introduction

The discovery in the late 1990s of the so-called spin-ice state in the low-temperature regime of the pyrochlore $Ho_2Ti_2O_7$ [1] has led to a period of intense research into the physical properties of this novel, and somewhat unexpected, magnetic state. The pyrochlore lattice, a three-dimensional (3D) arrangement of corner-sharing tetrahedra, is known to display the highest degree of geometrical frustration: a system of antiferromagnetically coupled isotropic spins sitting on the vertex of the tetrahedra shows a macroscopically degenerate manifold of ground states and thus does not order as $T \rightarrow 0$ but remains in what Villain called the 'cooperative paramagnetic' state [2]. For Ising spins, instead, it is the ferromagnetic system that is frustrated [3]. The pyrochlore oxides $Ho_2Ti_2O_7$ (HTO) and $Dy_2Ti_2O_7$ (DTO) [4] are experimental realizations of this latter case [1, 5]. The strong Ising anisotropy arises in these compounds from the local crystal field at the rare-earth positions and constrains the Ho^{3+} and Dy^{3+} ions ($J = 8$ and 15/2, respectively) to lie along the local [111] axes. The effective ferromagnetic coupling, on the other hand, results from the combination of dominant long-range dipolar interactions ($D_{nn} = 2.35$ K) with antiferromagnetic nearest-neighbour exchange ($J = -0.52$ and -1.24 K for HTO [6] and DTO [7], respectively). As a result, below ~1 K the system sets into the disordered spin-ice state characterized by the '2-in 2-out' spin configuration analogous to proton displacement vectors in Pauling's model of hydrogen disorder in water ice (for a review, see [8]). In fact, the residual configurational entropy measured for these materials is close to Pauling's predicted value for ice [8, 9].

The static magnetic properties of spin-ice materials $Ho_2Ti_2O_7$ and $Dy_2Ti_2O_7$ as well as their Sn analogues $R_2Sn_2O_7$ (R = Ho, Dy) are well accounted for by the dipolar spin-ice model [7, 10], which successfully describes the static spin–spin correlations, measured by neutron scattering down to 50 mK [6, 11–13], as well as the field dependence of the magnetization along the main cubic symmetry directions [14–17]. The dynamical properties of the spin-ice materials are also well established for $T > 1$ K. The main thermal spin relaxation process quenches out at $T \sim 16$ K [18, 19], below which a regime of quantum tunnelling relaxation sets in that persists down to ~1 K [20, 23], where HTO and DTO enter the spin-ice state. In this regime, direct measurements of the frequency- and time-dependent spin–spin correlation functions using neutron scattering techniques largely rule out any spin dynamics on the experimental timescale down to 50 mK [20] and [21][4]. Bulk measurements are also consistent with the freezing of spin relaxation mechanisms below ~0.7 K [19, 22, 23]. However, a loss of about 9% polarization in the $t = 0$ limit in neutron spin echo (NSE) measurements on HTO suggests that a small percentage of the spin system remains dynamic in this regime with a characteristic relaxation time of $<10^{-12}$ s [20].

In fact, the possibility of persistent dynamics in the low-temperature frozen spin-ice regime had already been hinted, albeit on a different timescale, by early μSR work by Harris *et al* on a single crystal of HTO, which, at 40 mK, showed a slow relaxation of the resolved asymmetry (1/3 of the full asymmetry being already lost at this temperature in the instrumental dead-time of the ISIS pulsed facility, this being attributed by the authors to the size of the largely static local field from the Ho^{3+}) [24]. In contrast, later work by Dunsiger and co-workers, also on single-crystal samples of HTO, showed a complete depolarization of the beam in $t \ll 1$ μs in zero field at low T, which the authors attributed to rapid Ho^{3+} fluctuations rather than inhomogeneous static local fields [25]. This sharp decay of the beam polarization has, since then, been observed in preliminary measurements on polycrystalline samples of DTO for $T < 50$ K [26].

Here we present μSR results collected on a polycrystalline sample of DTO in the temperature range $0.02 \text{ K} < T < 300$ K in longitudinal fields (LF) up to 2.7 T. In agreement with previous work, the relaxation becomes faster on cooling and the beam is almost completely depolarized in $t < 1$ μs for $T < 60$ K. However, below 40 K there is a recovery of the resolved asymmetry up to 1/3 of the full value. We argue that it is a combination of diminishing electronic fluctuations and the development of inhomogeneous internal fields that explain the μSR response in the 'hot-paramagnetic' regime of spin ice. At 20 mK, the repolarization of the beam by the application of a longitudinal field is consistent with a disordered magnetic state in which the Dy^{3+} moments are essentially static on the experimental time window. On the other

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[4] Quasi-elastic neutron scattering results on HTO collected with the IRIS spectrometer (ISIS, Rutherford Appleton Laboratory, UK) show that the signal is resolution-limited for T well above the quenching into the frozen spin-ice state, thus signalling an upper limit of $\sim 10^{-9}$ s for the characteristic time of the dynamics.

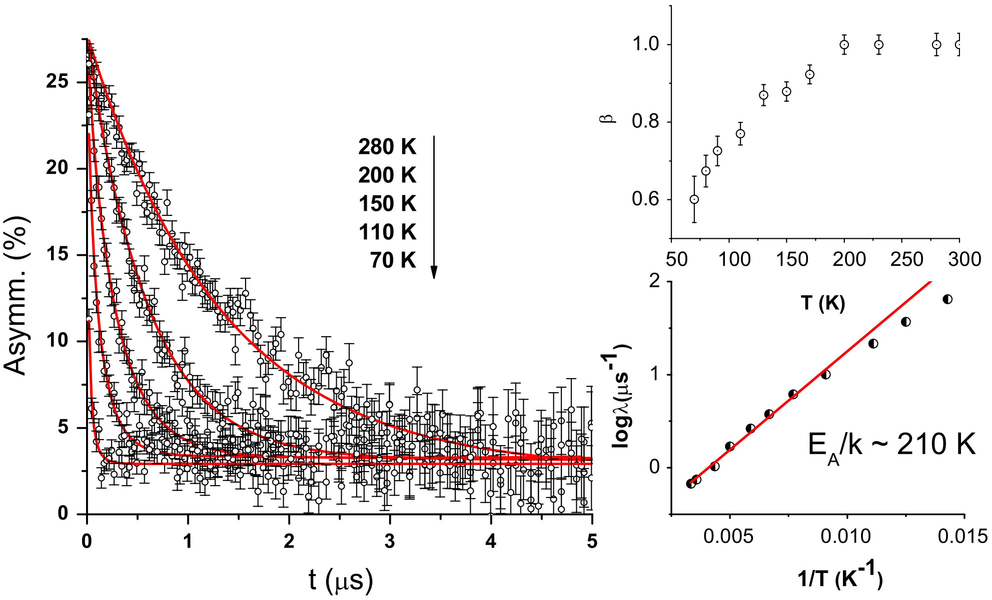

Figure 1. High-temperature muon depolarization spectra in a 50 G longitudinal field. Solid lines are the fit to an stretched exponential function $P_z(t) = a_0 \exp(-\lambda t)^\beta + b_k$. Top inset: thermal evolution of the β exponent. Bottom inset: temperature dependence of the derived muon relaxation rate λ. The solid line demonstrates the thermally activated nature of the spin relaxation at high T, with activation energy $E_a/k_B \sim 210$ K.

hand, however, the presence of a small slowly relaxing component indicates the persistence of dynamic fluctuations in the millikelvin regime, which we tentatively ascribe to hyperfine coupling with the Dy nuclear moments.

2. Experimental details

Muon spin relaxation measurements were performed on polycrystalline samples of $Dy_2Ti_2O_7$ (synthesized from a stoichiometric mixture of Dy_2O_3 and TiO_2 at 1300 °C) using the GPS and LTF spectrometers at the LMU facility at the Paul Scherrer Institut (Switzerland). For a review of the μ^+SR technique see, for example, [27, 28]. Essentially, information about the dynamics of the local fields in the sample is obtained by monitoring the positrons emitted by the muons in their decay in detectors placed in the forward (N_F) and backward (N_B) positions relative to the polarized muon beam. The muon-spin relaxation function (or asymmetry) is given by

$$G(t) = \frac{N_B(t) - \alpha N_F(t)}{N_B(t) + \alpha N_F(t)} \propto P_z(t) \qquad (1)$$

where α is an instrumental parameter that accounts for the F/B detector efficiency and $P_z(t)$ is the average muon-spin polarization function. $P_z(t)$ describes the muon depolarization inside the specimen and thus provides information on the time evolution of its internal fields.

3. Results and discussion

3.1. The paramagnetic and 'hot' spin-ice states, $T > 1$ K

Figure 1 shows the high-temperature time evolution spectra of DTO in a small LF of 50 G (used to decouple the nuclear spin system without affecting the electronic one). As expected, the depolarization of the muon beam becomes faster with decreasing temperature and, below approximately 60 K, the asymmetry is almost entirely lost in the electronic dead time. Data above this temperature were fitted to a power exponential function

$$P_z(t) = a_0 \exp(-\lambda t)^{\beta} + b_k \tag{2}$$

where a_0 is the total relaxing asymmetry and b_k is a background term arising from muons stopping in the sample surroundings. The exponent β evolves from a value of 1 at high T to $\sim$0.60 at 70 K (top inset of figure 1). The bottom inset in figure 1 shows the fit of the derived muon depolarization rate λ to an exponential $\lambda \propto \mathrm{e}^{(E_\mathrm{a}/k_\mathrm{B}T)}$, yielding an activation energy $E_\mathrm{a}/k_\mathrm{B} \simeq 200$ K. This value is in good agreement with that reported earlier for both DTO [19, 18] and HTO [20], which corresponds to the theoretically calculated gap between the ground state and first excited doublets [29, 30]. The $\propto \exp(E_\mathrm{a}/k_\mathrm{B}T)$ dependence and the energy scale of E_a thus indicate that an Orbach process [31, 32] involving transitions to/from the first excited doublet are the main contribution to spin dynamics in this temperature range.

At around 60 K the muon ensemble is completely depolarized in $t \ll 1$ μs. Further cooling the sample below 40 K shows, however, a recovery of the resolved $t = 0$ asymmetry up to roughly a third of a_0 that then decays with a rate that decreases with T and levels off below about 12 K (see figure 2(a)). Such a behaviour cannot be explained as simply resulting from fluctuating spins but, as shown below, is to be understood as a combination of the slowing down of the electronic fluctuations and the development of an internal dipolar field that appears static in the experimental time window. It is the change on cooling of the ratio between the relaxation rate $1/T_1$ of the electronic fluctuations and the width of the local field distribution that yields the observed thermal evolution of the asymmetry.

In a disordered magnetic system such as spin ice, the μSR response in the regime of coexisting electronic fluctuations and strong spin correlations that give rise to a distribution of internal fields with finite *rms* value B_i is usually well described by a dynamical Kubo–Toyabe function [33, 34]. Its actual shape depends on the relative values of ν, the characteristic frequency of the spin fluctuations, and $\Delta = \gamma_\mu B_i$ (where $\gamma_\mu = 135.5$ MHz T^{-1} is the muon gyromagnetic ratio): for rapid fluctuations, $\nu/\Delta \geqslant 5$, the relaxation simplifies to a simple (or power) exponential decay $P_z(t) = \exp(-\lambda t)$. In the slow fluctuation limit ($\Delta/\nu \leqslant 1$), on the other hand, the depolarization curves are characterized by a rapid initial decay of the asymmetry due to the transverse components of the static field distribution and the effect of spin dynamics is only sensed as a slow relaxation of the characteristic '1/3-tail' of the static Kubo–Toyabe function.

For DTO the exponential decay shown in figure 1(a) indicates that at high temperature the depolarization of the μ^+ ensemble is dominated by spin fluctuations involving the lowest-lying excited doublet. These fluctuations slow down as the temperature decreases and the spin system settles into the ground-state doublet. Ac-susceptibility measurements have shown that the high-temperature relaxation channel freezes at around 15 K [18, 20], which means that at low T (figure 2(a)) the system finds itself in the slow-fluctuation regime. However, the *rms* internal field that develops on cooling is so large already at temperatures as high as 20–30 K that the fast initial decay is lost in the electronic dead time and only the 1/3-tail is resolved. According to this interpretation, we have fitted the depolarization of the beam in this regime, which is due to the presence of residual dynamics, with the phenomenological function[5]

$$P_z(t) = \frac{a_0}{3}[(1 - f)\exp(-\lambda_{\mathrm{dyn}} t) + f] + b_k \tag{3}$$

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[5] The observed $\frac{1}{3}$ component could, in principle, be ascribed to an ordered static state with a large internal field so that only the longitudinal component is resolved. This, however, is ruled out by bulk magnetic measurements that fail to detect LRO in spin ice at any finite T.

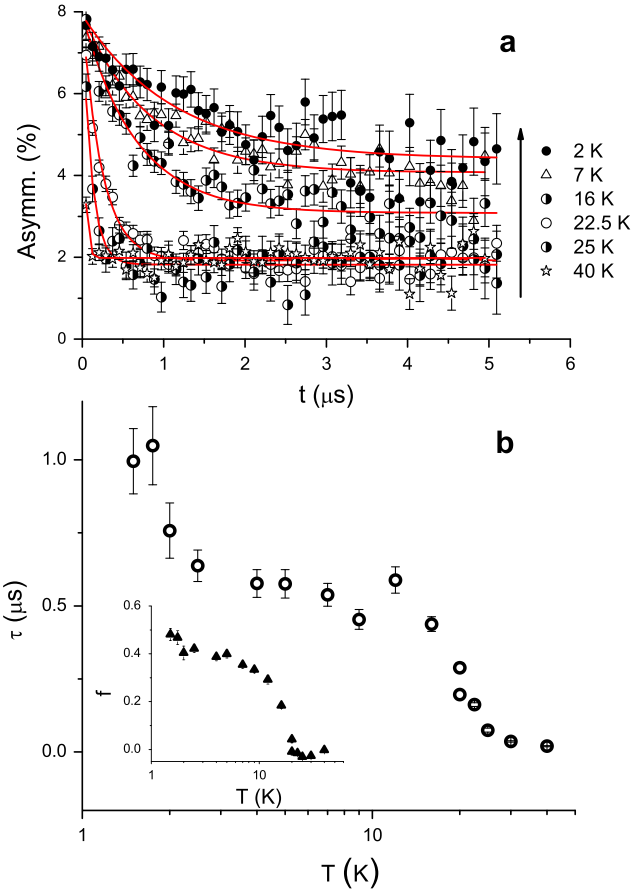

Figure 2. (a) Muon spin depolarization spectra for DTO below 40 K in an applied field LF = 50 G. The solid lines represent the fit to the function $P_z(t) = \frac{a_0}{3}[(1-f)\exp(-\frac{2}{3}\nu t)+f]+b_k$. (b) Thermal evolution of the derived characteristic relaxation time $\tau = 1/\nu$. Inset: thermal evolution of the non-relaxing component f.

where f accounts for the non-relaxing fraction that develops with decreasing T and $\lambda_{\text{dyn}} = \frac{2}{3}\nu$ is the dynamical muon spin depolarization rate responsible for the damping of the 1/3-tail. Note that the above expression implies a single correlation time $\tau = 1/\nu$, in agreement with results by Snyder and co-workers that showed the existence of a single or a very narrow distribution of τ at all temperatures [18, 35]. In fact, the derived characteristic time, shown in figure 2(b), reproduces the trend obtained by these authors from susceptibility measurements [35]: the flattening that occurs below $T \sim 15$ K marks the crossover from thermally activated dynamics to a quantum mechanical regime where relaxation occurs via tunnelling between the two degenerate states in the ground-state doublet [20, 35], whereas the upturn observable below ~2 K, at the onset of the frozen spin-ice regime, is interpreted as a re-entrance of a thermally activated regime probably involving coherent motions of small group of spins [35].

However, despite their common thermal dependence, a puzzling three-orders-of-magnitude difference exists between the two sets of results: our data are sensitive to dynamics in a microsecond timescale, whereas Snyder's susceptibility data probe fluctuations on the millisecond timescale. Given the different experimental time windows of two techniques, the difference in τ could, in principle, simply indicate that ac-χ and μSR measurements are sensing different relaxation processes, albeit with the same temperature dependence and involving the same energy levels (since the derived activation energy is essentially the same for both techniques). This is, however, highly unlikely, as both susceptibility [18] and NSE [20] measurements of spin ice clearly indicate that the dynamics in this temperature regime is characterized by a single relaxation channel (in the case of the current μSR data, $\beta < 1$ in equation (2) probably results from the presence of two or more muon stopping sites that become magnetically inequivalent on cooling). Alternatively, the difference could arise from the different Q-space probed by the two techniques: whereas a bulk technique such as ac-χ probes excitations in $Q = 0$, in μSR (a local technique) one averages over all Qs. According to this interpretation, the observed difference in τ would imply the dispersion of the excitation, i.e. imply a certain degree of coupling among the rare-earth ions. However, the three-orders-of-magnitude disparity seems rather large to be the result of a Q-dependence and, more importantly, this apparent dispersion contradicts the single-ion character of the relaxation process as determined by NSE measurements [20, 36]. The different timescale of these two relaxation channels thus remains an outstanding problem that needs to be re-addressed in the future.

3.2. The frozen spin-ice state, $T < 0.5$ K

In order to get further insight on the nature of the frozen spin-ice regime, we have measured the LF dependence of the spectra at a temperature well inside this state (20 mK). Given the loss of the early part of the signal and to ensure that the observed change in a_0 is a true effect of the LF on the system and not an instrumental artefact, data were normalized by the field response of a silver plate of the same geometry (which allowed us to determine the change with applied field of the α parameter in equation (1). The value of α is first determined for each field using the silver plate and then used in the fit of the DTO data at the same field). The normalized spectra for LF up to 2.5 T are presented in figure 3, where two features are immediately noticeable: first, in zero field (ZF) the resolved asymmetry, accounting for roughly a third of the total value, relaxes with a sizeable relaxation rate $\lambda \sim 0.20\ \mu\text{s}^{-1}$ (from the fit to equation (3), implying a characteristic time $\tau \propto 10^{-6}$ s), indicating the persistence of electronic fluctuations down to base temperature. Second, the application of the LF causes the repolarization of the μ^+ ensemble. We first discuss this latter effect before speculating about the possible origin of the spin dynamics.

Neutron scattering measurements have shown that in zero applied field the frozen spin-ice state is characterized by short correlation lengths that extend not much further than neighbouring tetrahedra [1, 12, 37]. The muon therefore experiences a broad and random distribution of largely static local fields, so one expects a Kubo–Toyabe-like relaxation of the asymmetry [34]. Indeed, as for $2\ \text{K} \leqslant T \leqslant 12$ K, the ZF data are consistent with the expected rapid depolarization of the $2/3$ transverse component caused by the spread of quasi-static internal fields—not resolved in our experimental setup due to the size of the large local fields created by the Dy^{3+} moments ($\mu = 10.6\ \mu_\text{B}$); see below—followed by a slow relaxation of the longitudinal component by residual dynamics in the rare-earth spin system.

In a Kubo–Toyabe relaxation, as the applied field is increased, the vector sum of the local and applied fields becomes increasingly aligned along the field direction z and the muon polarization is restored. Full recovery is achieved for $B_\text{appl} \geqslant 5B_i$. From the data plotted in figure 3, where full saturated asymmetry is obtained for $B_\text{appl} = 2.5$ T, one could thus estimate an average static component of the dipolar field at the muon site of ~0.5 T, a value that is only slightly lower than that reported by Dunsiger from HTO single-crystal data,

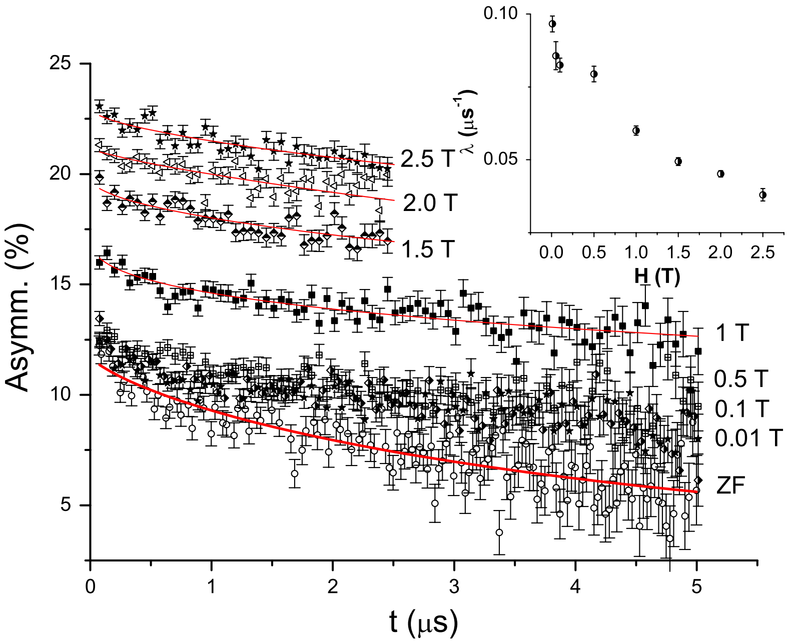

Figure 3. Longitudinal field dependence of the muon depolarization spectra for DTO at 0.020 K. The red solid lines represent the fit of the data to a simple exponential $P_z(t) = a_0 \exp(-\lambda t) + b_{\mathrm{tot}}$. Inset: estimated relaxation rate in the range 0.01 T $\leqslant H \leqslant$ 2.7 T. Note that $\lambda = 0.20\ \mu\mathrm{s}^{-1}$ in ZF.

$B_i \sim 1$ T [25], and similar to the value estimated for isostructural $\mathrm{Tb_2Mo_2O_7}$ ($B_i \sim 0.7$ T), with a comparable magnetic moment [38]. However, this estimate is based on the assumption that the Zeeman energy of the Dy^{3+} moments in the given LF does not exceed the exchange and dipolar interactions that stabilizes the spin-ice configuration, which is not correct for DTO: with $D_{\mathrm{nn}} = 2.35$ K and $J = -1.24$ K, an applied field of 0.3 T already exceeds them and is capable of altering the characteristic '2-in 2-out' spin structure. In fact, depending on the orientation along which the field is applied, the system adopts a number of new spin configurations at relatively low field intensity [1, 39, 40] and, for $\mu_0 H \simeq 3$ T applied along any of the main crystallographic orientations, the Dy^{3+} moments adopt a non-collinear ferromagnetic state [16, 39]. Thus, although the repolarization of the muon beam is generally a signature of static internal fields in a magnetic system, one could not, without additional neutron or susceptibility data, take it as a definitive proof in the case of spin ice. Strictly speaking, we can only say that our current data are *consistent* with a frozen disordered state, as shown by neutron and susceptibility data.

Now, the damping of the longitudinal component at ZF reveals the existence of residual spin fluctuations with a characteristic time $\tau \sim 10^{-6}$ s ($\lambda = 0.2\ \mu\mathrm{s}^{-1}$ in ZF) in a temperature regime in which dynamics are ruled out, at least in the ac-χ (10^0–10^{-4} s) and neutron scattering (10^{-9}–10^{-14} s) domains. In fact, kinetic arguments have been put forward to explain the absence of the predicted phase transition into an ordered state at 0.18 K [39] and the hysteretic behaviour of the magnetization below 0.5 K [41]: below this point the system gets locked into one of the '2-in 2-out' spin configurations, as the remaining thermal energy is incapable of flipping the Dy^{3+} moments above the barrier generated by the effective ferromagnetic coupling. Recently, a magnetocaloric study of DTO [42] has shown a crossover at ~0.3 K into a regime characterized by extremely slow relaxation due to the minimal spin flip probability.

Thus, we suggest that a relaxation mechanism involving energy exchange with the ^{161,163}Dy ($I = \frac{5}{2}$) nuclear system might explain the relatively fast dynamics observed in our study in the frozen state at such low temperature[6]. Although a simple scenario involving phonon-induced electron spin flips which couple to the nuclear moments is ruled out by the absence of sufficient thermal energy, one can invoke quantum mixing of nuclear and electronic levels to allow low-temperature relaxation of the electron spins via thermal fluctuations in the nuclear spins (which occur down to lower temperatures). This mechanism has been used to explain the short T_1 observed in the millikelvin regime in rigid insulators doped with paramagnetic impurities [44, 45] and can be visualized as a small-amplitude 'wobbling' of the electronic spin about the $\langle 111 \rangle$ axis induced by nuclear spin flips. In fact, a related idea involving small 'incoherent oscillations' of the rare-earth moments about the $\langle 111 \rangle$ axes was proposed by Ehlers *et al* [20] in order to explain the loss of roughly 9% polarization in the $t = 0$ limit in NSE measurements of HTO. Their results, however, imply a characteristic time $\tau < 10^{-12}$ s, which is much shorter than that estimated in our current study. Given the speculative nature of our discussion, it is difficult to argue in favour or against either estimate. However, the loss of polarization in the NSE data can be observed in temperatures well into the paramagnetic regime (up to at least 200 K [20]), which is something that appears difficult to reconcile with the current interpretation of the high-temperature dynamics which the same authors proposed in terms of a single-ion relaxation process: the fact that equation (2) provides a good description of muon depolarization curves above 70 K (figure 1) with $a_0 \sim 0.27$ (close to the full asymmetry expected for the instrument) and a value of the non-relaxing background ($\sim$0.02), as expected from muons stopping in silver rules out the possibility that a significant portion of the spins in DTO might experience relaxation processes much faster than those experienced by the majority and detected in the experiment. That is, according to our μSR results, a putative second high-T relaxation channel must act on *all* spins and not only part of them. This said, it seems unlikely that the wobbling of the spins as described above can cause such an effect at high temperature once spin flips and transitions to/from excited states are restored, or at least not suffer a significant change in its magnitude on cooling as the system sets into the ground state doublet, i.e. in what could be classically described as locked along the $\langle 111 \rangle$ local axes. The missing NSE fraction, however, remains constant up to 200 K [20]. Note as well that, although NSE and μSR probe different Q ranges, the NSE signal of spin ice (there is *a priori* no reason why DTO should behave differently from HTO at high temperatures) has been found to be Q-independent in the range of the measurements ($0.1 \leqslant Q \leqslant 0.4$) [20], as expected for a single-ion process, which, again, rules out the possibility that a dispersive character could be the origin of the different energy scale of the relaxation channel as detected by the two techniques (if it is, in fact, the same process in the two windows). Finally, we want to point out that the loss of $t = 0$ amplitude in the NSE signal, ascribed to the presence of an unknown fast relaxation process, is not unique to spin ice but appears ubiquitous among geometrically frustrated magnets [46–50], where its origin, or indeed the disparity between the observed NSE and μSR results, has not yet been worked out.

4. Conclusions

There are two main conclusions that can be drawn from the current work. First, data in the paramagnetic and 'hot spin-ice' regimes (i.e. $T > 1$ K) demonstrate that the depolarization of the muon ensemble is essentially caused by the development on cooling of strong,

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[6] Preliminary results on HTO have shown a much faster spin relaxation in this compound than in DTO at 0.020 K [43], which can be explained by the difference in the hyperfine coupling in the two materials and thus provide further evidence for the nuclear-induced relaxation of the electronic system in this temperature regime.

inhomogeneous quasi-static internal fields and is only marginally due to dynamic fluctuations of the electronic spin system, in agreement with results from bulk techniques. Second, and more importantly, we find residual spin dynamics well inside the frozen spin-ice regime ($T \ll 1$ K, a temperature range where other techniques show the spin system to be completely frozen), which we propose results from the mixing of electronic and nuclear energy levels, which induces a small-amplitude 'wobbling' of the electronic spin about the quantization axis. It will be interesting to explore whether such an effect is common to other frustrated magnets in which spin dynamics can be detected in the millikelvin regime coexisting with a frozen spin state.

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