Nuclear Forward Scattering Study of Relaxation in the Geometrically Frustrated Ferromagnet $Dy_2Ti_2O_7$

$Dy_2Ti_2O_7$ is a pyrochlore in which the Dy^{3+} ions lie on the vertices of a network of corner-sharing tetrahedra. Spins arranged in such a configuration cannot minimize all of their exchange interactions simultaneously if these are antiferromagnetic. This phenomenon, called "geometrical frustration," results in a highly degenerate ground state and exceptionally complex dynamics. $Dy_2Ti_2O_7$ [1], along with similar pyrochlores $Ho_2Ti_2O_7$ [2] and $Ho_2Sn_2O_7$ [3], are ferromagnetic. However, in these materials, strong crystal fields constrain the rare-earth ion spins to lie along the (1 1 1) directions that connect the corners of each tetrahedron to its center, and so they too demonstrate geometrical frustration.

Nuclear forward scattering (NFS) probes the hyperfine fields created at each resonating nucleus by the electrons surrounding it. Thermal fluctuations of the electron spins affect the hyperfine fields and thus the observed NFS spectra. We used NFS at the undulator beamline **BL35XU** to measure the Dy ion spin-flip rate in a 98-μm-thick $Dy_2Ti_2O_7$ crystal foil [4]. We collected NFS time spectra showing the intensity of the resonantly emitted photons as a function of time after the arrival of the initial synchrotron radiation pulse. The nuclear resonance used here is that of ^{161}Dy (natural abundance 19%) with energy 25.65 keV and natural lifetime 42 ns.

The foil was inserted in a cryostat, and its NFS time spectra were collected over a temperature range from 8 to 287.3 K by an array of 16 thin avalanche photodiodes with time resolution below 200 ps and efficiency ~17%. Previous experiments done without such a fast detector (Ref. 15 of [4]) could not capture the fast NFS oscillations caused by the large hyperfine splittings of the ^{161}Dy nucleus. The bandwidth of the beam incident on the foil was limited to about 0.5 meV by a high-resolution monochromator, which passed a flux of about 2×10^8 photons/s. The beam ran parallel to the foil's (1 1 1) direction and was polarized within 3° of its (1 −2 1) axis. NFS data were typically available 4 ns after the exciting pulse, but a correction to the detector efficiency (~20%) was needed at times before 8 ns after the excitation. This has been applied.

Figure 1 shows selected NFS time spectra. At temperatures below about 45 K, the beats due to the magnetic splitting remain clear. The spin flips broaden the resonance linewidths, resulting in the observed faster decay. At intermediate temperatures, the spin-flip rate is high enough to collapse the

resonance spectrum into a single broad line, as reflected in the rapid decay observed in our sample at 55 K. Finally, at high temperatures, the average time between spin flips is much less than the nuclear resonance lifetime. Thus the nuclei see only an average hyperfine magnetic field of zero, and only the electric field gradient affects the decay.

The NFS time spectra from 8 to 90 K and at 287.3 K permitted definite determination of the hyperfine fields and spin-flip relaxation times. The hyperfine fields were assumed to consist of a magnetic field **H** and a parallel electric field gradient with a single component, V_{zz}. The relaxation was included by using the model of Blume and Tjon [5]. Theoretical fits to selected spectra are shown in Fig. 2.

The relaxation times τ from 30 to 90 K could be well fit to an Arrhenius equation, $\tau = \tau_0 \exp(E_a/k_B T)$, where T is the absolute temperature in deg K. Figure 3 shows the Arrhenius plot. The high-temperature limit τ_0 of the relaxation time is $15.76^{+3.03}_{-2.53}$ ps, and the activation energy is 271.9±8.1 K. While the activation
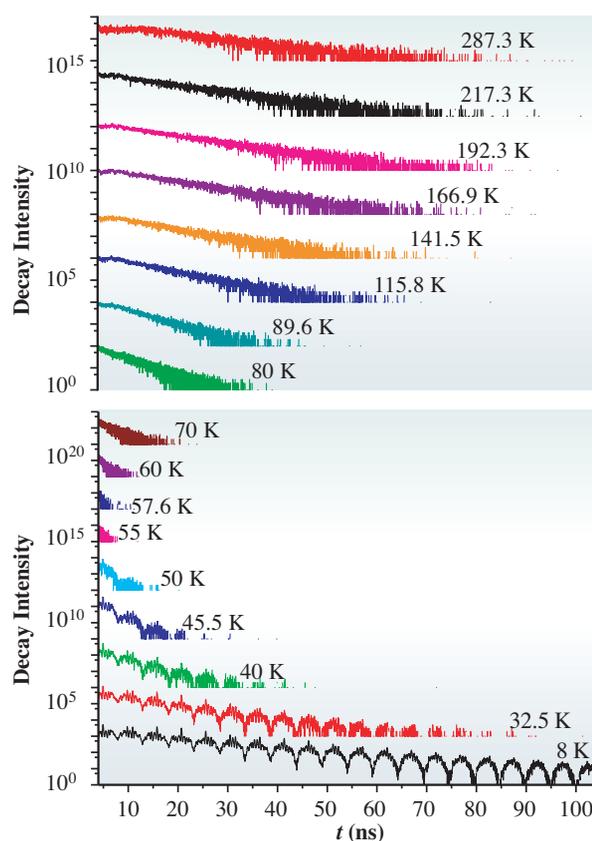

Fig. 1. Selected nuclear forward scattering time spectra collected from $Dy_2Ti_2O_7$ sample foil at various temperatures.

energy of $Dy_2Ti_2O_7$ is similar to those of $Ho_2Ti_2O_7$ and $Ho_2Ru_2O_7$, the τ_0 value for $Dy_2Ti_2O_7$ is about three times its value for the Ho pyrochlores [4].   One possible reason for this difference is that the effective exchange interaction is only 1.1 K in $Dy_2Ti_2O_7$, but is 1.8 K for $Ho_2Ti_2O_7$.

We find that |$\mathbf{H}$| and V_{zz} are flat throughout the low-temperature region, with their uncertainties increasing with temperature. |$\mathbf{H}$| lies between 559 and 583 T (765-798 MHz) up to 90 K, with uncertainties not exceeding 2.4 T or 3.3 MHz. eQV_{zz}, where Q is the electric quadrupole moment of the ^{161}Dy nucleus, lies between 1160 and 1294 MHz up to 60 K, with uncertainties not exceeding 100 MHz. Our values at 8 K are |$\mathbf{H}$| = 561.74±0.01 T and eQV_{zz} = 1293.63±0.09 MHz, in good agreement with previous measurements as discussed in [4]. The spectrum at 287.3 K yields a value of 493 T (675 MHz) for |$\mathbf{H}$|, with a large uncertainty of 114 T (156 MHz) consistent with a high spin-flip rate. The value for eQV_{zz} at this temperature, 364.8±3.5 MHz, differs sharply from that found at low temperatures, perhaps because the Dy ions here enter a crystal field excited state in which a different electric field gradient is exerted on the nuclei.

The NFS spectra taken from 100 to 220 K did not yield unique values for the hyperfine fields and relaxation times, though we could obtain reasonable fits by extrapolating these parameters from the low-temperature data.   These spectra could be approximated by simple exponential decays with decay lifetimes increasing from 9.6 to 12.6 ns from 115.8 to 192.3 K, falling slightly again to 12.0 ns at 217.3 K.

Fig. 3. Arrhenius plot of τ *versus* T in $Dy_2Ti_2O_7$. For the low-temperature data, the error bars lie within the squares. The solid line is the best-fit Arrhenius function (see text).   The dotted line is the extrapolation of this best fit to room temperature.

In sum, we have studied spin relaxation in $Dy_2Ti_2O_7$ using nuclear forward scattering of synchrotron radiation by ^{161}Dy. A new fast and efficient detector showed that the NFS technique can be applied to this isotope in a practical way. The activation energy of $Dy_2Ti_2O_7$ appears characteristic of titanate pyrochlores, but the high-temperature limit of the relaxation time is about three times that of Ho pyrochlores. The hyperfine fields agree well with previous measurements at low temperatures and are essentially independent of temperature up to 90 K.

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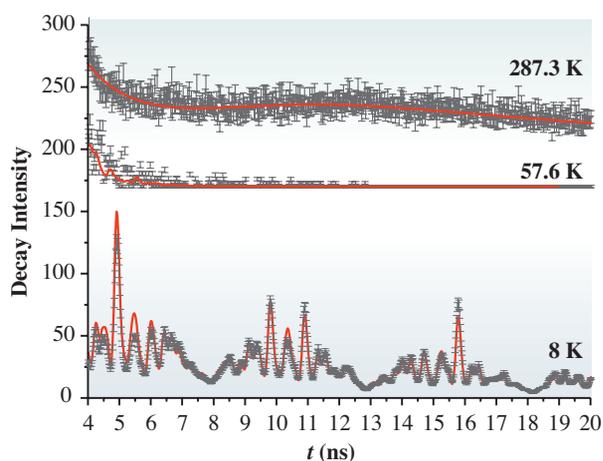*E-mail: John.Sutter@diamond.ac.uk

Fig. 2. Best fits of selected spectra from 4 to 20 ns. The data are given by the individual points with error bars. The red curves are the best-fit spectra. For clarity, the 8 K spectrum is scaled down by a factor of 50 and the one for 57.6 K is expanded by a factor of 3.
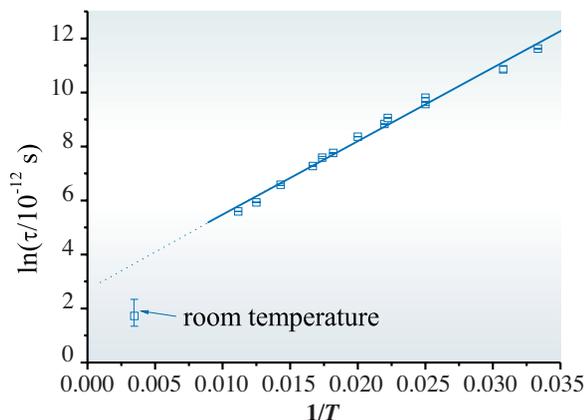
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