Experimental report

12/09/2022

Proposal: **4-04-507** **Council:** 10/2020

Title: Hyperfine coupling as a probe of the unconventional nature of the Nd2Zr2O7 pyrochlore magnet

Research area: Physics

This proposal is a continuation of 4-05-778

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Samples: Nd2Zr2O7

Instrument	Requested days	Allocated days	From	To
IN16B	4	4	21/06/2021	25/06/2021

Abstract:

We wish to continue our investigations on the hyperfine spectrum in the frustrated pyrochlore magnet Nd2ZrO27. Our first observations are quite far from other results on Nd based compounds and also far from current theoretical descriptions, hence suggesting that we are at a turning point in the understanding of the physics of these systems. In this context, the objective of this continuation is first to ensure the accuracy of our present data at low temperature and second to continue our temperature dependence investigations on IN16B.

Experimental report on 4-04-507: Hyperfine coupling as a probe of the unconventional nature of the $Nd_2Zr_2O_7$ pyrochlore magnet

Context : Recently, the $Nd_2Zr_2O_7$ pyrochlore material has attracted a lot of attention among the family of highly frustrated magnets [1-3]. Current understanding of this material assumes the existence of a high temperature paramagnetic phase, which transforms upon cooling in a spin ice phase, and eventually in an all in - all out antiferromagnetic (AIAO) magnetic ordering below T_N=300 mK. The previse value of the ordered moment apparently depends on the single crystal / powder nature of the sample. An ordered moment of m=0.8 μ_B is found in crystals, whereas a value closer to 1.1 μ_B is found in powders. This phase also supports peculiar spin excitations which encompass a flat mode at low energy (E_0=70 µeV) with a pinch point pattern on top of dispersive excitations [4-6].

In this context, the study of the hyperfine coupling between the Nd electronic and nuclear spins is of specific interest. In particular, the spin ice and AIAO spin configurations play the role of magnetic fields expected to lift the degeneracy of the nuclear spin multiplet, giving rise to peaks at E = ±A m, where "m" is the *local* Nd electronic magnetic moment and A ~ 1.33" µeV/ μ_B (see figure 1) [9]. According to theory, one thus expects E = 2.9 µeV in the spin ice phase [10], and E= 1.1 µeV in the AIAO phase.

Previous results: During the September 2020 cycle, we had the opportunity to carry out the experiment at IN16B on $Nd_2Zr_2O_7$ using a dilution refrigerator to probe these hyperfine excitations. Surprisingly, we could identify not only 1 peak in each regime, but several peaks (on the positive energy side) at about 2.4, 3.2 µeV and a third peak around 1 µeV. Those peaks start to form below about 750 mK, but no significant evolution could be observed on entering the AIAO phase down to the base temperature of 80 mK (see figure 2).

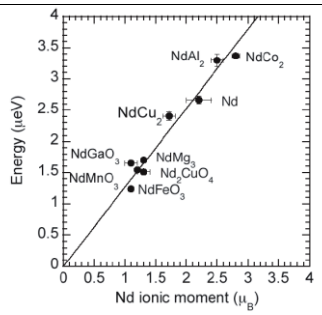

Figure 1: Plot of the energy of the inelastic signal in several Nd-based compounds (from [9]). This leads to A ~1.33 µeV/ μ_B.

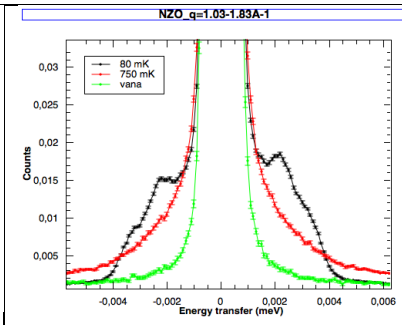

Figure 2: IN16B measurements on $Nd_2Zr_2O_7$ at 80 mK (black) and 750 mK (read). Vanadium is shown for comparison. Several peaks are observed at 80 mK, while a strong quasielastic contribution is present at 750 mK.

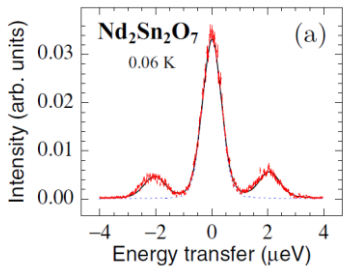

Figure 3: Inelastic neutron scattering measured on IN16B in $Nd_2Sn_2O_7$ showing the hyperfine field splitting induced by the Nd ordering (from Ref. [11])

This first experiment leads to strange results. However, the measurement conditions were not optimal for several reasons:

I. Thermalization in $Nd_2Zr_2O_7$ has always been an issue. The fit of our data is consistent with this base temperature, yet the SANE team realized that the sample stick was not perfectly screwed, casting some doubts on the actual temperature. Long time was spent at low temperature to wait for thermalization (adding Helium in the cell during the experiment), yet the temperature dependence could not be measured precisely.

II. In addition, a large quantity of powder sample was used (about 10 g), to fill a 15 mm cell. This may be at the origin of a temperature gradient in the cell but also of multiple scattering.

III. Finally, our observation are far from theoretical expectations, as well as from other observations on Nd based compounds. As an example, IN16B measurements on a neighbor compound $Nd_2Sn_2O_7$ are displayed in Figure 3. In this case, the obtained local ordered moment was consistent with the ordered moment deduced from diffraction (1.8 µB).

A second experiment was thus scheduled on IN16B to solve these issues. We used the low flux mode (at variance with the first try), wavelength 6.27 A, and standard Si111 yielding an energy resolution with a FWHM ~ 0.75 µeV. To avoid the uncertainty on the sample temperature and its homogeneity, we use a copper annular cell (internal diameter 14 mm, external 16 mm, height 4.5 cm). It provides two copper surfaces for the thermalization and a smaller thickness of the sample. The cell was filled with 1 bar of Helium at low temperature to ensure a better thermalization (the background did not evolve in the previous experiment when adding more Helium in the cell).

We measured at several temperatures from 50 mK up to 10 K. Part of the data are shown below in two different forms, as an energy-temperature map (figure 4) and classical spectrum (figure 5) at low temperatures. In this second experiment, a single broad peak forms below the Néel temperature at an energy close to 1.5 µeV.

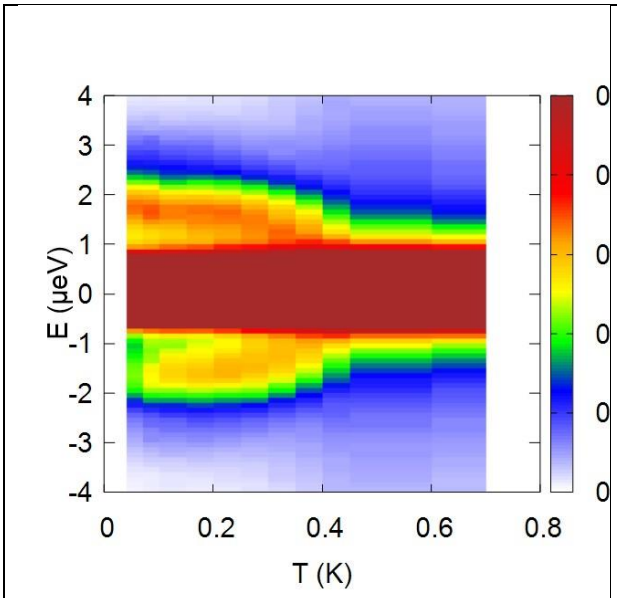

Figure 4 : raw data taken at IN16B. The data have been gathered to plot a map as function of energy transfer and temperature.

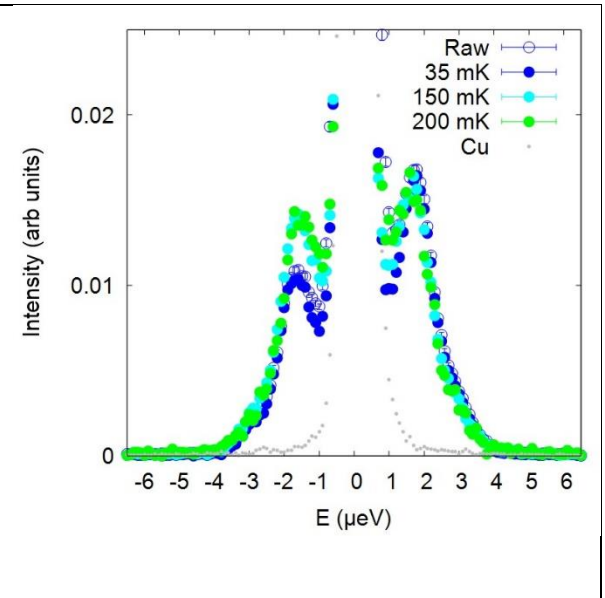

Figure 5 : raw data taken at various temperatures. The empty can ("Cu") is also shown for the sake of comparison.

Figure 6 gives the averaged energy <E> of these spectra (red points, left y-axis), along with the corresponding magnetic moment <m> determined based upon the relation E = A m (blue point, right y-axis). Above T_N, the value of <E> should be understood as arising from a quasi-elastic scattering (not shown here). Two conclusions can be drawn based on this second experiment:

I. Below T_N, the local magnetic moment, if the above relation is applicable, is large, and even larger than the moment determined from diffraction: close to 1.8 µB instead of 1.1 expected from diffraction.
II. Moreover, no static field exists, at the nuclear time scale, in the spin ice state just above T_N.

Further analysis is ongoing to figure out this intriguing result.

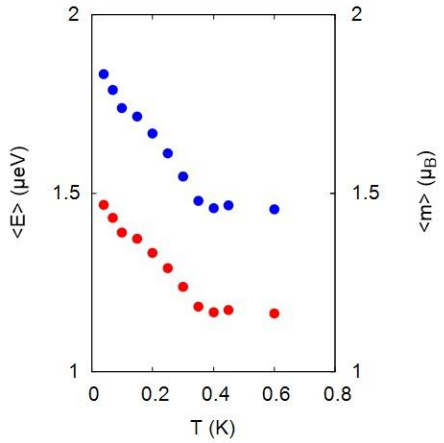

Figure 6 : The left y-axis gives the average energy of the raw data as a function of temperature. The right y-axis is the ordered (electronic) magnetic moment as deduced from the E=Am relation (see [9]).

References

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