Experimental report

25/05/2022

Proposal: 5-32-930 **Council:** 4/2021

Title: Correlations in the spin-liquid ground state of the Ising triangular antiferromagnet NdTa7O19

Research area: Physics

This proposal is a new proposal

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Samples: NdTa7O19

Instrument	Requested days	Allocated days	From	To
D7	7	7	24/06/2021	01/07/2021

Abstract:

A classical disordered, macroscopically degenerate ground state was predicted for the Ising triangular antiferromagnet already in 1950, while additional non-commuting exchange terms were later suggested to possibly lead to its quantum counterpart, a quantum spin liquid. Several complementary experiments that we have undertaken on a novel triangular antiferromagnet NdTa7O19 suggest that it might be the first to realize such a state. Our previous INS study has revealed that its crystal-electric-field ground-state Kramers¿ doublet is characterized by highly anisotropic Ising exchange interactions between effective spin-1/2 degrees of freedom, while our muSR study has suggested a quantum-disordered ground state with low-frequency excitations that are found to persist down to at least 66 mK. Here we propose to investigate spin correlations as a unique fingerprint of the spin-liquid ground state via magnetic diffuse scattering on the D7 spectrometer, which would allow us to further characterize and better understand this enigmatic ground state.

The measurements were performed with polarized neutrons on a 5.4-g sample at base temperature of 50 mK and at 5 K. The incident energy of the neutrons was 3.55 meV and their wavelength was 4.8707 Å. This allowed measurements in the wave-vector range Q = 0.3 − 2.5 Å^{-1}. We note that this range allows for the detection of nearest-neighbour antiferromagnetic spin correlations, when present, despite the relatively large nearest-neighbour distance of 6.22 Å. For standard calibration of detectors, measurements on vanadium were first performed and quartz polarization corrections were made. The diffraction mode of the instrument was used, where the scattering intensity was effectively integrated up to the neutron incident energy. The three-directional *XYZ* polarization analysis was utilized to separate the magnetic contribution to the neutron scattering cross section from the incoherent and nuclear coherent contributions (Fig. 1).

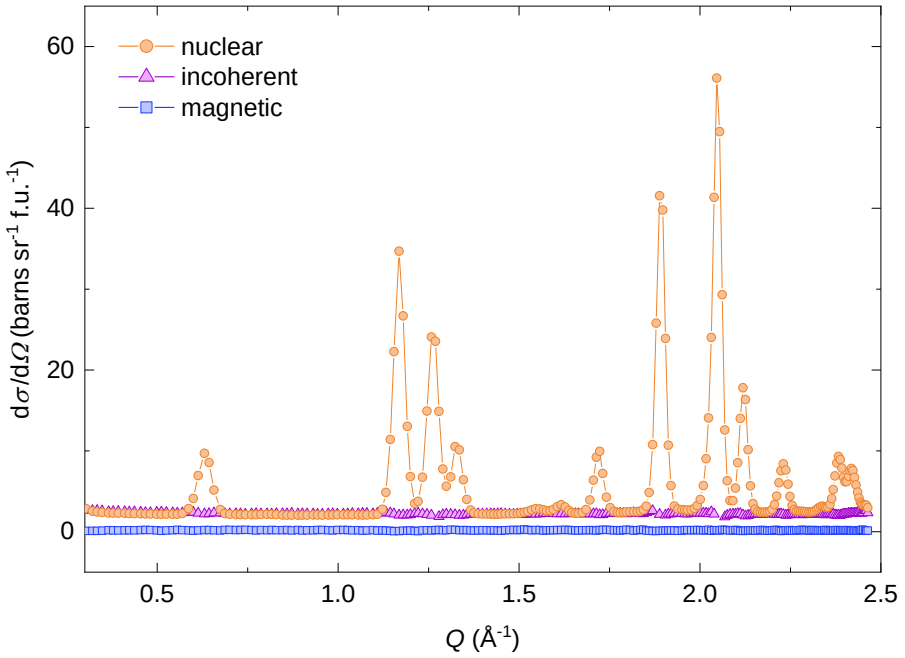

Fig. 1: Magnetic, incoherent, and nuclear coherent neutron scattering cross sections in $NdTa_7O_{19}$ at 50 mK obtained from the *XYZ* polarization analysis.

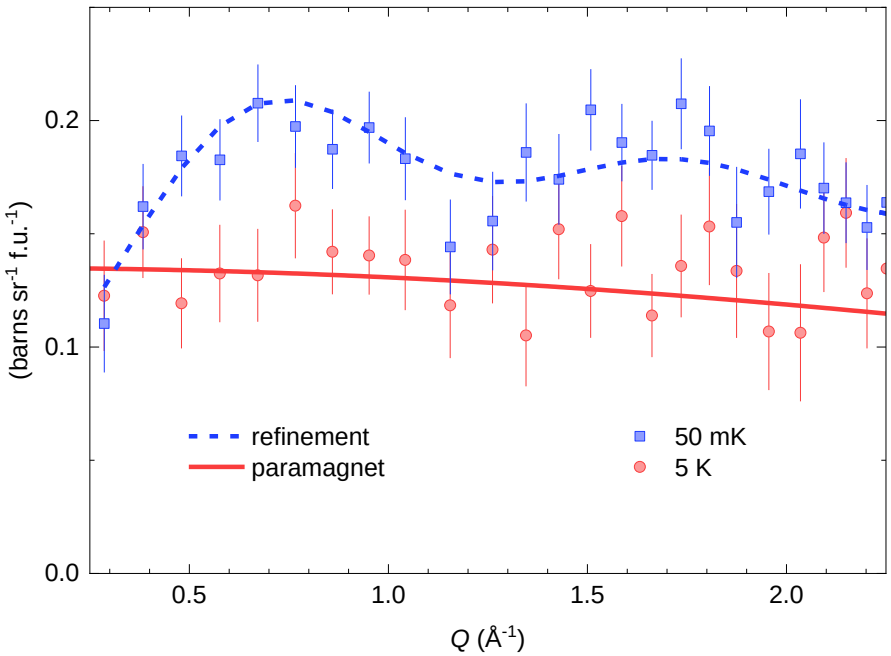

Fig. 2: The magnetic neutron scattering cross section in $NdTa_7O_{19}$ at 50 mK and 5K. The former reveals diffuse scattering due to nearest-neighbour antiferromagnetic spin correlations predominantly of the Ising type (refinement), while the latter is simply paramagnetic.

The magnetic part of the scattering cross section shows no sharp peaks even at the base temperature (Fig. 1), thus demonstrating absence of long-range magnetic ordering. On the contrary, broad diffuse magnetic scattering is clearly observed (Fig. 2). At 5K, the diffuse scattering is due to paramagnetism. It shown monotonic decrease with increasing Q due to the Nd^{3+} form factor and is nicely modelled with no ajustable parameters if the average g factor (taken from other measurements) is considered (Fig. 2). The diffuse scattering is clearly enhanced at 50 mK when compared to 5K. Moreover, at the base temperature it exhibits oscillations in Q, which nicely correspond to antiferromagnetic correlations between nearest neighbours within triangular planes.

We have modelled the diffuse scattering at 50 mK with reverse Monte Carlo refinements (Fig. 2) using Spinvert Suite. For these refinements we used a spin supercell of 3×3 unit cells, which was fitted to the experimental data, and performed 300 proposed refinements per spin for 216 independent spin configurations that were calculated in order to obtain good statistics. We find excellent agreement with the experiment for nearest-neighbour spin correlations perpendicular to the triangular planes dominating over the in-plane spin correlations, thus yielding an Ising-like nature of the investigated triangular antiferromagnet.