Experimental report

12/09/2025

Proposal:	**4-01-1857**	**Council:**	10/2024
Title:	Excitation Spectrum of the Triangular-lattice antiferromagnet (CD3ND3)2NaRuCl6		
Research area:	Physics		

This proposal is a new proposal

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Samples: (CD3ND3)2NaRuCl6

Instrument	Requested days	Allocated days	From	To
THALES	5	5	25/06/2025	30/06/2025

Abstract:

The prospect of merging the paradigms of geometric frustration on a triangular lattice and bond anisotropies in the strong spin-orbit coupling limit holds tremendous promise in the ongoing hunt for exotic quantum materials. The recently discovered organic quantum magnet (CD3ND3)2NaRuCl6 poses a rare candidate system to realize such physics, displaying a triangular arrangement of Ru3+ ions adopting the spin-orbital entangled j = ½ state. Thermodynamic and magnetometric data suggest a highly frustrated ground state with significant antiferromagnetic correlations but residual magnetic order below TN = 0.22 K. We hereby propose a single-crystal investigation on the magnetic excitation spectrum of (CD3ND3)2NaRuCl6 at zero field and in the fully polarized state, from which we will attempt to determine the effective spin Hamiltonian and pinpoint the relevance of potential bond anisotropies. With an expected bandwidth of ~ 0.5 meV and a suspected presence of continuum excitations, the cold triple-axis spectrometer THALES with the FLATCONE multi-analyzer setup will be ideally suited for this purpose.

Experimental Report:
Excitation Spectrum of the Triangular-lattice antiferromagnet $(CD_3ND_3)_2NaRuCl_6$

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The prospect of merging the paradigms of geometric frustration on a triangular lattice and bond anisotropies in the strong spin-orbit coupling limit holds tremendous promise in the hunt for exotic quantum materials. The recently discovered organic quantum magnet $(CD_3ND_3)_2NaRuCl_6$ poses a rare candidate system to realize such physics, displaying a triangular arrangement of Ru^{3+} ions adopting the spin-orbital entangled $j_{\mathrm{eff}} = 1/2$ state. Thermodynamic, magnetometric and neutron diffraction data suggest a frustrated ground state with residual magnetic order below $T_N \approx 0.22$ K and a highly complex H-T phase diagram, including several incommensurate states. We proposed a single-crystal investigation on the magnetic excitation spectrum of $(CD_3ND_3)_2NaRuCl_6$ in the fully polarized regime using the cold triple-axis spectrometer THALES with FLATCONE multi-analyzer setup. This should allow us to determine the leading terms in the effective spin Hamiltonian, constituting a key step in understanding this exotic quantum material.

16 single crystals were coaligned in the ab scattering plane for a total sample mass of 1.9g. The probe was installed with (hk0) scattering geometry in a dilution refrigerator with 10T vertical cryomagnet and cooled to $T \sim 70$ mK. All measurements were taken at $E_f = 4.06$ meV (FWHM resolution $\Delta E = 175$ μeV), fixed by the FLATCONE-setup. After checking the sample alignment & confirming the presence of magnetic bragg peaks around the K-point, the magnetic field was ramped to $\mu_0 H_{\parallel c} = 7\mathrm{T}$. In this field-polarized regime, we collected 35 constant-energy slices, rotating 90° in 0.5° steps to capture the spin-wave dispersion throughout the Brillouin zone (Fig.1, left).

After masking the inelastic bragg-tails and accounting for form factor, we average equivalent cuts and plot the QE dispersion along high symmetry directions (Fig.1, right). As expected, there is only one resolution-limited branch. The bandwidth amounts to $9JS \approx 0.50$ meV, while the gap $\Delta \approx 0.58$ meV at the K-point neatly separates magnetic scattering from the quasi-elastic line. We extract points along the dispersion from gaussian fits at constant-Q and fit the exchange parameters using SpinW. The dispersion is well described by a nearest neighbor Heisenberg model (green line) with $J_1 = 1.30(1)$ K. Further neighbor couplings are irrelevant within error bars. Inter-plane interactions must be small, since the saturation field $g_c \mu_B H_{\mathrm{sat}} \approx 0.49$ meV matches the $9JS$ expected in 2D. An anisotropic model (red line) with $J_1 = 1.31(1)$ K, $\Delta = 0.97(2)$ and $J_{\pm\pm} = 0.3^{+0.1}_{-0.3}$ K gives a marginally better fit ($\chi^2 \approx 1.8$ vs. $\chi^2 \approx 2.4$), but the dispersion is weakly sensitive to $J_{\pm\pm}$ and completely insensitive to $J_{z\pm}$. Actually, given the spin-orbital entangled ground state doublet, incommensurate order and anisotropic phase diagram, such bond-dependent terms are likely to play at least a sub-leading role. This calls for further investigations in a transverse-field geometry.

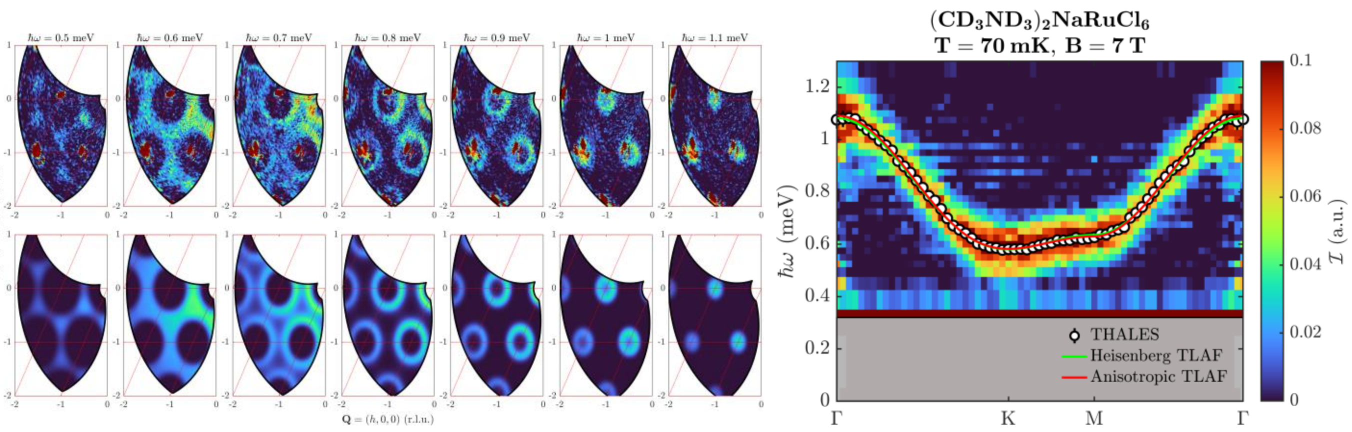

Figure 1: Left: Representative const-E slices in the field-polarized regime at 7T (top), together with the simulated FM spin-wave intensity in the same region (bottom). Right: Spin-wave dispersion along high-symmetry directions averaged over several Brillouin zone paths. Green/red lines show fits to the NN Heisenberg model / anisotropic model discussed in the text. The dispersion is nearly insensitive to bond-dependent terms.