7 Supporting Information

Calculations based on susceptibility data revealed a best fit with two exchange coupling constants. One parameter describes Cr-Cr exchange at the tips of the chain, while the other parameter describes exchange between Cr ions within the body of the chain. For such a model J_a = -1.38 meV and J_b = -1.80 meV (where J_a is the exchange interaction at the tips of the chain and, J_b is the interaction within the middle of the finite chain). Although this model fits the magnetic susceptibility data very well, INS data reveals the resultant eigenvalues to be incorrect, due to the first excited state eigenvalues lying 0.15 meV too high in energy relative to the energy determined directly by INS.

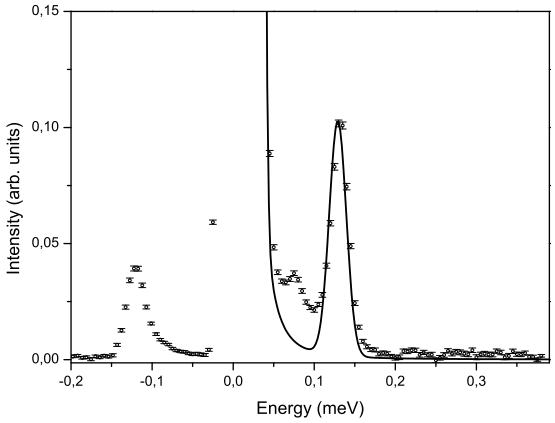


Figure 11: INS spectrum of **4** measured with a 9.0 Å setting at 1.8 K (black circles), while the black solid line shows the calculated spectra with J = -1.46 meV D_{Cr} = -0.045 meV.

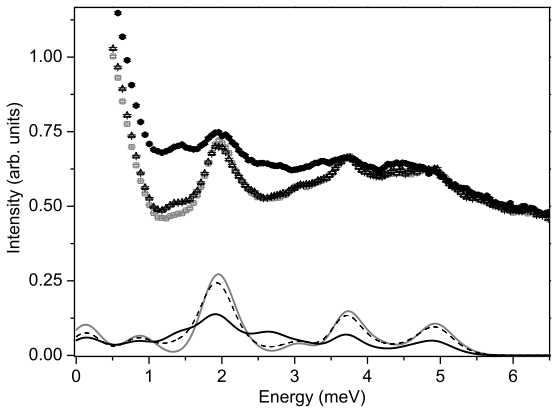


Figure 12: INS spectrum of **4** measured with a 3.2 Å setting at 1.8 K (gray open squares), 6.0 K (black open triangles) and 15.0 K (black circles). Lines show the calculated spectra where J = -1.46 meV and D_{Cr} = -0.045 meV at 1.8 K (solid gray line), 6.0 K (dashed black line) and 15.0 K (solid black line).

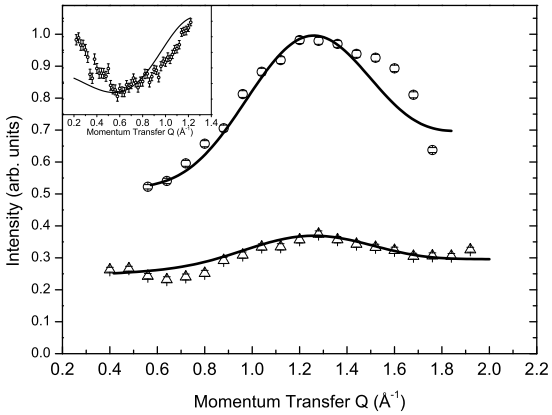


Figure 13: Intensity of 5.0 Å INS transitions as a function of Q for compound **4**. Transition **III** at 1.92 meV, 1.8 K (circles) and transition **i** at 1.42 meV, 6.0 K (triangles) are shown with calculated curves represented as solid lines. The insert shows the intensity of a 9 Å INS transition, **I** at 0.127 meV, 1.8 K (circles) as a function of Q dependence, with calculated curve represented as a black solid line. Calculations are performed where J = -1.46 meV and D_{Cr} = -0.045 meV.

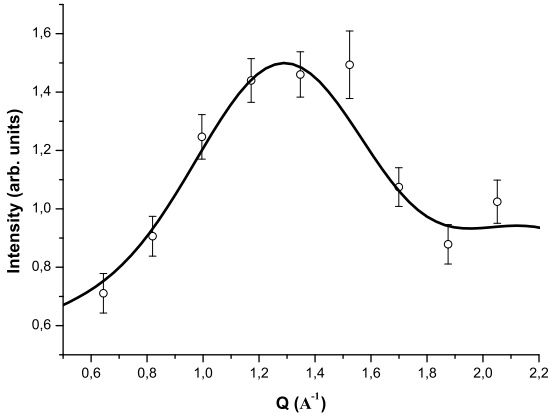


Figure 14: INS intensity of the $|1\rangle$ to $|2\rangle$ transition of **5** centered at 1.2 meV as a function of momentum transfer Q (black circles). Solid black line represents the calculated momentum transfer Q. The calculation is performed where J = -1.46 meV and D_{Cr} = -0.042 meV.

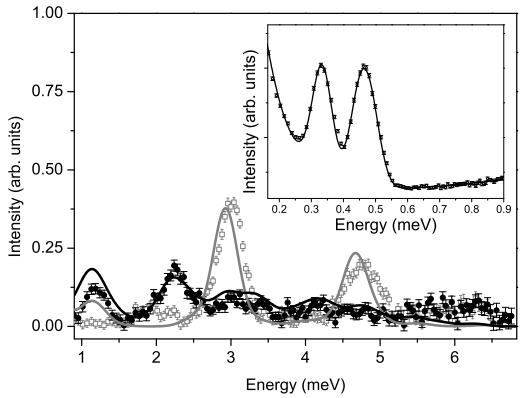


Figure 15: INS energy spectra measured with a 3.2 Å setting at 2.0 K (gray open squares) and 15 K (black circles). Solid lines represent calculated spectra. The insert shows 6.5 Å 6.0 K INS energy spectrum (back circles) with calculated spectrum (solid black line). Calculations are performed where J_a = -1.40 meV, J_b = -1.46 meV and D_{Cr} = -0.042 meV.

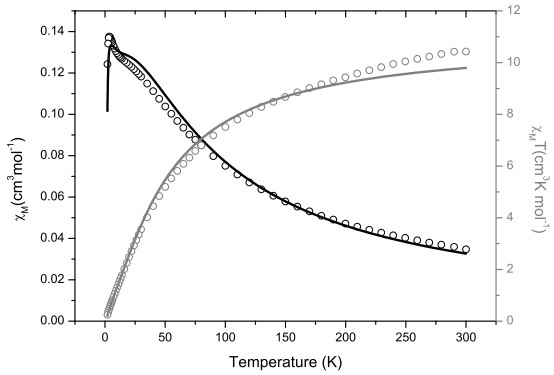
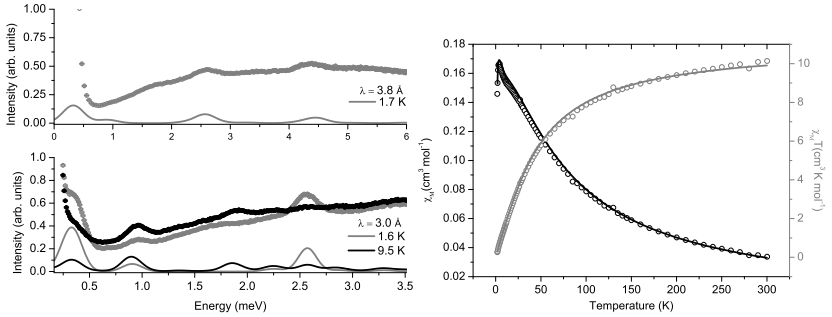


Figure 16: Susceptibility versus temperature of **6**. χ_M versus T, black open circles and $\chi_M T$ versus T, gray open circles. Solid lines represent calculations, where $g = 1.98$, $J_a = -1.40$ meV and $J_b = -1.46$ meV



(a) INS spectra (b) Susceptibility

Figure 17: (a) 3.0 Å and 3.8 Å INS spectra of **1** (circles) with corresponding calculations (lines). (b) $\chi_M T$ and χ_M verses temperature of **1** (circles), with corresponding calculations (lines). Calculation parameters J_a = -1.09 meV and J_b = -1.46 meV. INS simulations include anisotropy parameters D_{Cr} = - 0.028 meV and $|E_{Cr}| = 0.005$ meV. Susceptibility calculation includes a zeeman splitting factor of $g = 1.98$. Experimental data taken from reference [4].

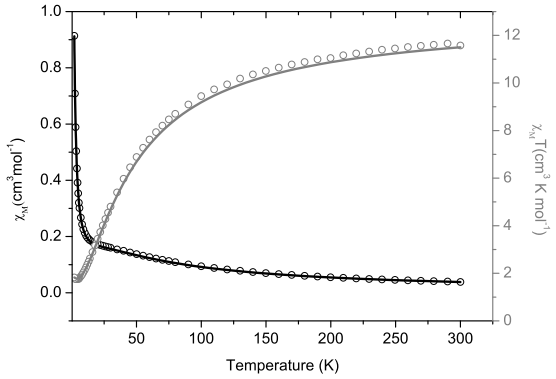


Figure 18: Susceptibility versus temperature of **2**, taken from reference [4]. χ versus T, black circles and χT versus T, gray circles. Solid lines represent calculations, where $g = 1.98$, J_b = -1.46 meV and J_a = -1.09 meV.