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Low energy XY spin clusters in a pyrochlore antiferromagnet with weak disorder¹

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The spin liquid state of the Heisenberg antiferromagnet (HAFM) on the pyrochlore lattice arises from an extensive degeneracy of correlated yet disordered ground states. How this spin liquid is modified in real materials with imperfect Heisenberg exchange is a rich field of study, with many possible outcomes depending on the relevant perturbations. We have studied a single crystal of a new pyrochlore antiferromagnet, NaCaCo₂F₇. High spin ($S = 3/2$) Co²⁺ forms a fully ordered pyrochlore sub lattice, while non-magnetic Na⁺ and Ca²⁺ are intermixed on the A-site. Despite isotropic magnetic properties and large antiferromagnetic coupling ($\Theta_{CW} = -140$ K), a freezing transition is observed at temperature much lower than the exchange energy ($T_f \sim 3$ K), thus revealing the relatively weak exchange disorder induced by the mixed ion A-site. Unexpectedly, our inelastic neutron scattering measurements reveal that the frozen state is of local XY character and supports low energy XY fluctuations. Yet the system can break free from the XY states at energies above 2.5 meV \sim 30 K; at these energy scales we observe the collinear Ising configurations expected for the weak-disorder HAFM model. The frozen state in NaCaCo₂F₇ provides a new outlook on the role of disorder in selecting spin configurations from the Heisenberg pyrochlore spin liquid state.

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