From one- to two-magnon excitations in the S = 3/2 magnet β-$CaCr_2O_4$

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Decades of theoretical, numerical and experimental studies have sought quantum systems, beyond the classical picture of conventional magnetism, focusing on low-dimensional S = 1/2 materials. The characteristic features of such systems are the lack of long-range magnetic order and the presence of deconfined fractional spin-1/2 excitations called spinons. Owing of the exact solution of the spin- chain model, theoretical and experimental works have led to a fairly comprehensive understanding of spin dynamics in the simplest quantum system, the Heisenberg antiferromagnetic chain of S = 1/2 spins. As spinons are created in pairs, the excitation spectrum for such spin chain is characterised by a continuum of excitations. Introducing a magnetic coupling between individual S = 1/2 chains forces spinon excitations to confine into bound states, called magnons, thus leading to a dimensional crossover between a quantum one-dimensional to a semi-classical three-dimensional regime. Yet, what happens when increasing the spin fractional quantum number is not known. Here we present neutron scattering experiments for a weakly coupled S = 3/2 chain compound, β-$CaCr_2O_4$. In the ordered state, the low-energy spin fluctuations resemble large-S linear spin-waves from the incommensurate groundstate. However, at higher energy, these semi-classical and harmonic dynamics are replaced by an energy and momentum broadened continuum of excitations. Applying kinematic constraints, required for energy and momentum conservation, sum rules of neutron scattering and comparison against exact diagonalization calculations, we show that the dynamics at high energy resemble low-S one-dimensional quantum fluctuations. β-$CaCr_2O_4$ therefore represents a unique example of a magnet at the border between classical Néel and quantum phases, hosting dual behaviors. As studies on low-dimensional magnets with intermediate spin values are scarce, we show that this system provides a rich playground to explore the physics beyond the usual quantum/classical dichotomy.