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Modelling diffuse magnetic scattering in a “partial spin-liquid” kagomé compound

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$\text{Fe}_4\text{Si}_2\text{Sn}_7\text{O}_{16}$ has a well-ordered complex composite layered structure that incorporates perfectly hexagonal kagomé lattices of high-spin Fe^{2+} ($S = 2$) cations. Below $T_N = 3.5$ K it adopts a unique “striped” magnetic structure [1,2] that breaks hexagonal symmetry and leaves 1/3 of the spins geometrically frustrated in an apparent “partial spin-liquid” state down to at least 40 mK. Polarised neutron diffraction data collected on D7 at the ILL show persistent structured magnetic diffuse scattering below T_N , consistent with short-range interactions among 1/3 of the HS Fe^{2+} sites. We have fit the diffuse magnetic scattering intensity with a Monte Carlo “big box” approach and a magnetic interaction approach. The determined signs and relative magnitudes of the spin exchange coupling are consistent between these two approaches and with the results of high-level DFT (meta-GGA SCAN functional) calculations. Our key result is that the first, second and third-nearest neighbour exchange interactions (J_1 , J_2 , J_3) are all strongly antiferromagnetic. This may explain why $\text{Fe}_4\text{Si}_2\text{Sn}_7\text{O}_{16}$ does not adopt either of the conventional long-range-ordered kagomé states that preserve hexagonal symmetry: $q = 0$ “in-out”, in which third-nearest neighbours are ferromagnetic with respect to each other; or $q = \sqrt{3} \times \sqrt{3}$ “spiral”, in which second-nearest neighbours are ferromagnetic with respect to each other. We propose that the partially ordered ground state of $\text{Fe}_4\text{Si}_2\text{Sn}_7\text{O}_{16}$, in which there are no strictly ferromagnetic relationships, is the best compromise when J_1 -3 are all strongly antiferromagnetic.

[1] CD Ling et al., Physical Review B 9 (2017) 180410

[2] S Dengre et al., Physical Review B 103 (2021) 064425

Topics

Magnetism and Condensed Matter

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