Experimental report

06/06/2024

Proposal: **5-53-317**

Council: 4/2023

Title: Structural and Magnetic Characterization of the Kitaev Ground State in Honeycomb SmI3

Research area: Physics

This proposal is a new proposal

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Samples: SmI3

Instrument	Requested days	Allocated days	From	To
D9	6	3	25/10/2023	30/10/2023
D3	5	10	25/09/2023	01/10/2023
			25/10/2023	30/10/2023
ORIENTEXPRESS	1	1		

Abstract:

SmI3 is a strong candidate for 2D Kitaev spin liquid physics, given that its cousins YbBr3 and ErBr3 have magnetic Hamiltonians which have suppressed Heisenberg exchange while Sm uniquely has a crystal field ground state capable of hosting the Kitaev state. We will search for structural distortions and magnetic short-range order to demonstrate the Kitaev model, while utilizing the strong octupolar magnetic form factor of Sm3+ to explore the role of multipolar degrees of freedom which could be hidden to many other techniques. The magnetic short range correlations will be used as input to a reverse-Monte Carlo model to fully solve the Kitaev magnetic Hamiltonian.

Experimental report

We carried out two experiments at D3 and one at D9. Samples we prepared in copper cans to protect from air sensitivity. Due to the use of natural Sm, we utilized nonstandard short wavelengths (0.4 Å and D9, 0.45 Å at D3).

The D9 experiment successfully mapped large areas of reciprocal space to search for diffuse scattering. We found conclusively that there is no long-range antiferromagnetic order or magnetic fluctuations. The magnetic propagation vector is constrained to k=(0,0,0), consistent with the D3 experiments.

For the D3 experiment with Cryopad, we analyzed the spin fluctuation directions for positions both on and off the ferromagnetic peaks. We found evidence for spin fluctuations on the k=(0,0,0) peaks, appearing in all fluctuation channels but without sufficient time to solve for any unique direction. In addition, there were some spurious features arising from scanning between nuclear reflections, which was unclear at the time because the resolution function was not known.

For the later D3 experiment with the liquids setup, we made complete 2D maps of the spin fluctuations, which showed where the spurious features from the Cryopad experiment came from. Most importantly, we were able to solve for the spin flip scattering component on the

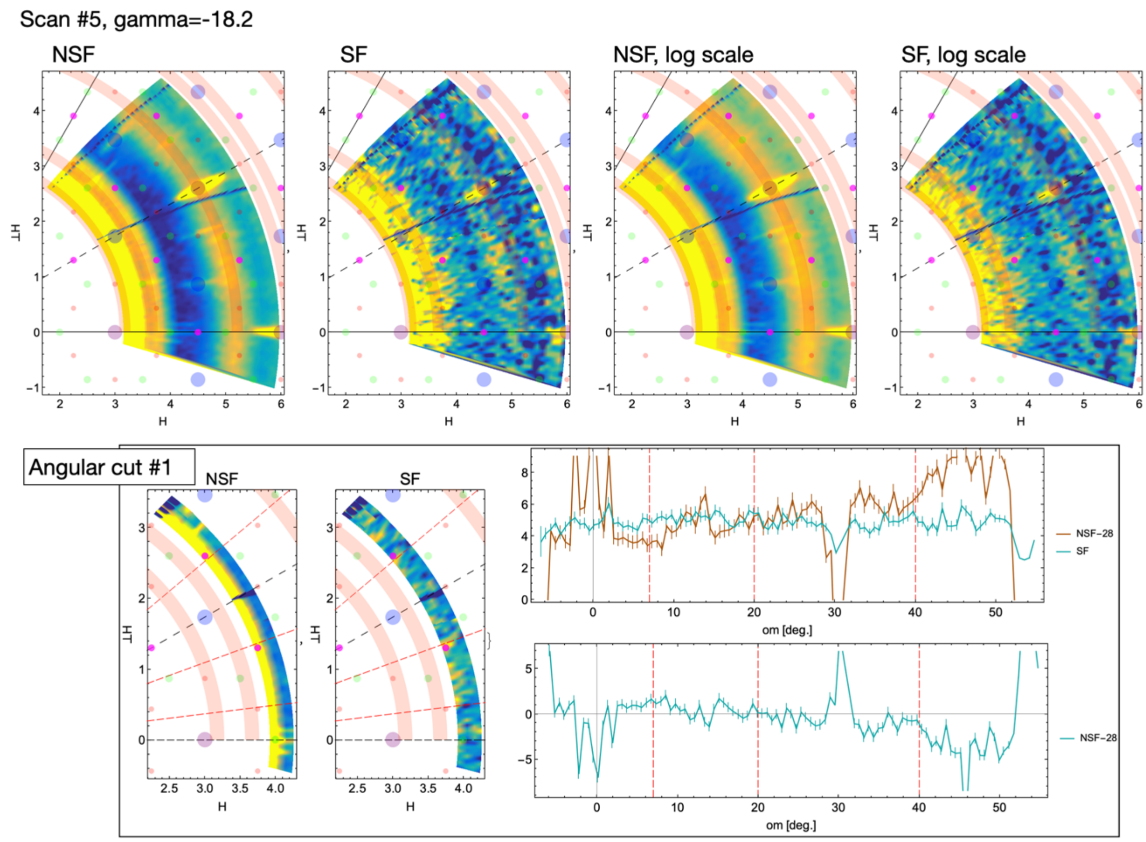

Figure 1. Liquids setup for one detector position, after polarisation correction was applied. The correction and analysis methods developed by the instrument scientists worked well. The various colored spots show different high-symmetry positions in the system. The angular cut shows no distribution of magnetic intensity that is symmetric with the crystal structure. However, the extra intensity on the nuclear peaks in the SF channel was determined to be real, through analysis of the structure factors over the entire experiment.

nuclear peaks. The importance differences with this experiment that enabled it to work were (1) use of the 2D liquids detector to fully understand the resolution function at a highly nonstandard wavelength required by Sm, and (2) the use of a small Si single crystal embedded next to the sample, which allowed for unambiguous measurements of the true beam polarization.

We found that the origin of scattering in the spin flip channel was depolarization of the neutron beam, rather than long-range magnetic correlations, indicating that the k=(0,0,0) structure and interactions are ferromagnetic. This is extremely significant, because it proves the lack of antiferromagnetic correlations in SmI_3, unlike the other candidate Kitaev materials such as the well-known $RuCl_3$. We are presently preparing a manuscript combining the results of all three experiments with other work we performed on the SmI_3 system.