Experimental report

02/09/2022

Proposal: **DIR-259** **Council:** 4/2021

Title: A DETERMINATION OF SPIN PAIRING STATE IN KAGOME SUPERCONDUCTOR CsV3Sb5

Research area: Physics

This proposal is a new proposal

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Experimental team: Philippe BOURGES

Local contacts: Anne STUNAULT

Samples: CsV3Sb5

Instrument	Requested days	Allocated days	From	To
D3	6	11	28/09/2021	04/10/2021
			08/10/2021	13/10/2021

Abstract:

Report on the proposal (DIR-259)
Title: A DETERMINATION OF SPIN PAIRING STATE IN KAGOME SUPERCONDUCTOR CsV3Sb5

Experimental team: Philippe Bourges, Chunruo Duan, Mason Klemm, Anne Stunault, Peng-Cheng Dai

Abstract: In the proposal DIR-259, we planned to study with polarized neutron the superconducting symmetry in the new topological superconductors, CsV_3Sb_5. Recent muons spin resonance results point out a time-reversal symmetry breaking in that material. It was then important to determine if the spin pairing state is a singlet or a triplet state. The experiment shows that no electronic magnetic moments are contributing to the observed magnetization which can be entirely explained by nuclear moments of I=7/2 of ^{51}V.

The kagome lattice which is constructed by corner-sharing triangles lead to a heated discussion about exotic properties arising from its geometrical frustration. Recently, superconductivity (SC) up to 3 K was observed in a set of kagome materials AV_3Sb_5 (A = K, Cs, Rb) [1]. Large anomalous hall effect [2], topological charge density wave (CDW) [3], twofold von Hove singularities [4], and two-domain SC were reported [5]. Therefore, the kagome metals AV_3Sb_5 (A = K, Cs, Rb) provide an exciting platform to study the interplay between correlations, magnetism, topology, and SC. Recent muon spin rotation experiments have confirmed the presence of a time reversal symmetry breaking (TRSB) below charge ordering temperature [6]. More importantly, the time reversal symmetry breaking field persists within the superconducting state [6]. This basically means that superconductivity in the system may not be s-wave conventional superconductor, as it is difficult to imagine how a s-wave superconductor with isotropic gap function can induce time reversal symmetry breaking field in the superconducting state.

The existence of magnetism induced by orbital effect is also supported by the results from scanning tunneling microscopy (STM) measurements. A robust 2x2 superlattice structure was observed and it exhibits an intensity reversal in real space, signaling charge ordering, across the Fermi level. The intensity of the charge ordering indicates a chiral CDW order, and its unusual response to the magnetic field suggests that the TRSB caused by spin-orbital magnetism [3].

On D3, the CsV_3Sb_5 sample has been mounted to access various Bragg peaks: (0,0,2),(0,0,4)… We have applied a vertical magnetic field (up to 9T) to magnetize the material. Experiments have been performed first in an orange cryostat. Through standard flipping ratios analysis, a very weak magnetic moment has been detected above 1.5K (Fig. 1). The fact that the moment is very weak shows that the material has very little magnetic electronic response down to 1.5K. Next, it does not change across the superconducting state (Fig. 1). A field of only 3T was applied below Tc in order to stay below Hc2 at 1.5K.

Surprised by this intriguing result, we mounted next the sample in a dilution fridge to cool down a 0.1K where Hc2 is larger, so we could apply a larger magnetic field. The figure 2 shows the results versus temperature. A large ferromagnetic component is then observed which is scaling with the applied field, H: $F_M = \chi H$ where χ is the uniform magnetic susceptibility. Further, χ is found proportional to 1/T (Figure 2) as one would expect for a purely paramagnetic moments with no correlations between spins. Further, the measured moments do not follow any form factor expected for electronic vanadium spins. We also do not see anisotropy of the uniform susceptibility in contrast to what would be expected for electronic spins.

All of that makes us next to consider what could be the contribution of the nuclear moments of I=7/2 of ^{51}V. At our surprise, a detailed quantitative analysis shows that the magnetization can be entirely explained by the contribution of these nuclear moments magnetized by the applied field. No electronic magnetic moments have been actually observed in this experiment and, unfortunately, no information could be provided about the superconducting spin symmetry in the new topological superconductors, CsV_3Sb_5 from this experiment.

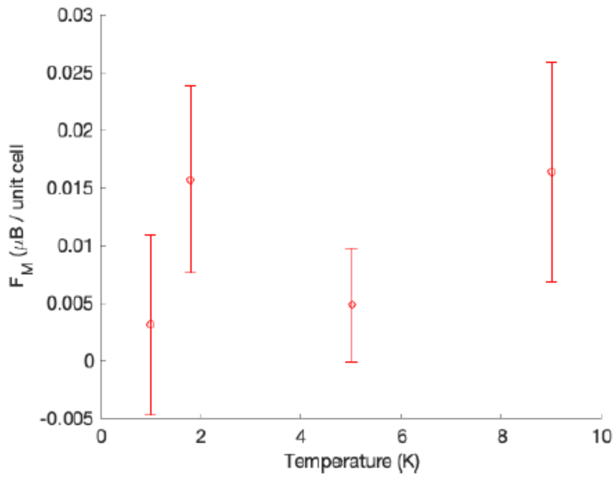

Fig 1: Tiny ferromagnetic component induced by the magnetic field (3T) in CsV_3Sb_5

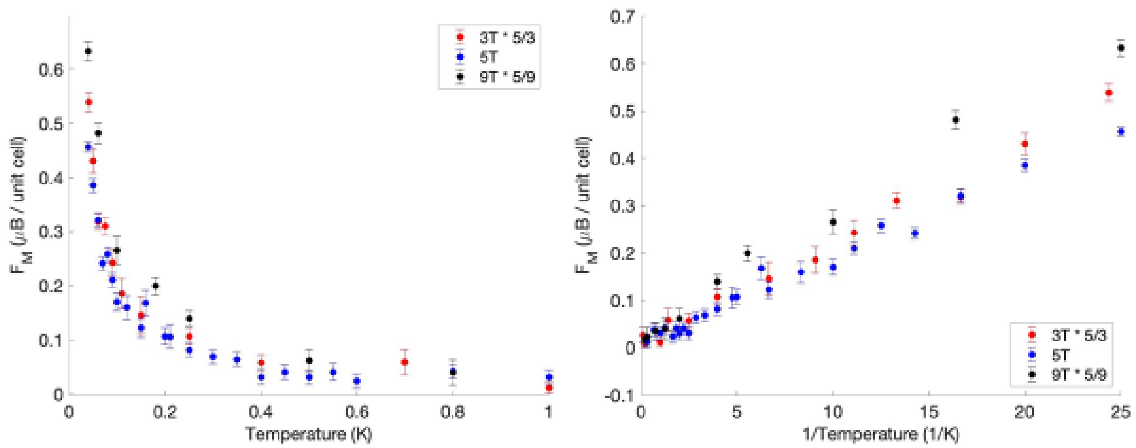

Fig 2: Ferromagnetic component below 1K induced by the magnetic field at 3, 5 and 9T in CsV_3Sb_5. All data collapse to a single curve as F_M=M/H where H is the applied field.

References:

[1] Ortiz, Brenden R., et al. "New kagome prototype materials: discovery of KV 3 Sb 5, RbV 3 Sb 5, and CsV 3 Sb 5." *Physical Review Materials* 3.9 (2019): 094407.

[2] Yang, Shuo-Ying, et al. "Giant, unconventional anomalous Hall effect in the metallic frustrated magnet candidate, KV3Sb5." *Science advances* 6.31 (2020): eabb6003.

[3] Jiang, Yu-Xiao, et al. "Discovery of topological charge order in kagome superconductor KV3Sb5." *arXiv preprint arXiv:2012.15709* (2020).

[4] Kang, Mingu, et al. "Twofold van Hove singularity and origin of charge order in topological kagome superconductor CsV3Sb5." *arXiv preprint arXiv:2105.01689* (2021).

[5] Zhao, C. C., et al. "Nodal superconductivity and superconducting dome in the topological Kagome metal CsV3Sb5." *arXiv preprint arXiv:2102.08356* (2021).

[6] C. Mielke et al., "Time-reversal symmetry-breaking charge order in a correlated kagome superconductor", arXiv: 2106.13443.

[7] A. N. Petsch et al., Phys. Rev. Lett. **125**, 217004 (2020).

[8] Ni Shunli et al, Anisotropic superconducting properties of Kagome metal CsV3Sb5, https://arxiv.org/pdf/2104.00374.pdf