Magnetic structure of a new quantum magnet SrCuTe$_2$O$_6$

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SrCuTe$_2$O$_6$ consists of a 3-dimensional arrangement of spin-$\frac{1}{2}$ Cu$^{2+}$ ions. The 1st, 2nd and 3rd neighbor interactions respectively couple Cu$^{2+}$ moments into a network of isolated triangles, a highly frustrated hyperkagome lattice consisting of corner sharing triangles and antiferromagnetic chains. Of these, the chain interaction dominates in SrCuTe$_2$O$_6$ while the other two interactions lead to frustrated inter-chain coupling giving rise to long range magnetic order at suppressed temperatures. In this paper, we investigate the magnetic properties in SrCuTe$_2$O$_6$ using muon relaxation spectroscopy and neutron diffraction and present the low temperature magnetic structure.

Interesting magnetic behaviour in Heisenberg spin systems originates from a network of some elementary motifs such as triangles or tetrahedra, where spins at their vertices interact with each other via antiferromagnetic (AF) interactions. The frustration in such systems often leads to exotic ground states such as spin liquids [1,2] and spin ice states [3,4] where long-range magnetic order (LRO) is suppressed to low temperatures or completely eliminated. In the case where order still occurs it can provide insights into the underlying physics and the new states arising from the frustration. There are many experimental examples for the three dimensional (3D) networks of corner-shared tetrahedra (pyrochlore and spinel structures) such as Gd$_2$Ir$_2$O$_7$ [5,6]. 3D networks of corner-shared triangles are relatively less explored despite the expectation of novel ground states. The simplest possibility of the latter is known as a hyperkagome lattice and has been observed in the compound Na$_4$Ir$_3$O$_8$ where every Ir$^{2+}$ spin is involved in two triangles. Although initial studies suggested a highly frustrating magnetic lattice with QSL behaviour [9], a glassy magnetic ground state has been observed in the muon relaxation studies [10,11].

PbCuTe$_2$O$_6$ is an example of a highly connected hyperkagome lattice, also known as the hyper-hyperkagome lattice, formed by the highly frustrated first and second nearest neighbour (NN) interactions between Cu$^{2+}$ spins [12]. Experimental and theoretical studies of this compound reveal evidence for quantum spin liquid behaviour down to 20 mK, a rare observation in three dimensional magnetic lattices [12,13], confirming the strong frustration in the system. However, density functional theory calculations also suggest significant non-frustrated third and fourth NN magnetic interactions in PbCuTe$_2$O$_6$ whose role in the QSL phase diagram is less understood.

SrCuTe$_2$O$_6$ is a promising quantum magnet, isostructural to PbCuTe$_2$O$_6$, that can give insights into the hyper-hyperkagome frustration mechanism responsible for the QSL ground state. SrCuTe$_2$O$_6$ crystallizes in cubic symmetry at room temperature (space group $P4_32$ [12]) with the magnetic spin-$\frac{1}{2}$ Cu$^{2+}$ ions occupying a single Wyckoﬀ site. The Cu$^{2+}$ ions are coupled together by exchange interactions $J_1$, $J_2$ and $J_3$. These three interactions couple them into isolated equilateral triangles, a hyperkagome lattice and uniform chains (running parallel to the a, b and c axes) respectively. If these interactions are antiferromagnetic they can give rise to a frustrated network of spin-$\frac{1}{2}$ chains. DC susceptibility of SrCuTe$_2$O$_6$ yields a negative Curie-Weiss temperature of $\theta_{CW}$ $\sim$35.4 K revealing predominantly antiferromagnetic exchange interactions [16,17], and shows a broad maximum at 32 K. This feature has been attributed to a one-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnetic chain revealing $J_3$=45 K [16,17] as the dominant interaction. However, two sharp features occur in the susceptibility at lower temperatures $T_{N_1}$=5.5 K and $T_{N_2}$=4.5 K, where a sharp $\lambda$-type anomaly is also observed in the heat capacity, indicating the onset of

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magnetic transitions in the system. These anomalies reveal non-negligible frustrated inter-chain coupling due to the finite $J_1$ and $J_2$ \[^1\O 1\]^1. In addition, the compound exhibits magneto-dielectric coupling at $T_{N1}$ and $T_{N2}$ \[^2\] attributed to the non-centro-symmetric nature of the structural symmetry. Furthermore, specific heat, magnetization and dielectric constant measurements as a function of applied magnetic field reveal a complex phase diagram with an additional field induced phase \[^16\] \[^17\].

Although SrCuTe$_2$O$_6$ reveals interesting magneto-dielectric and magnetoelectric properties around the magnetic transitions, the origins of the magnetic order and the nature of the magnetic structure below the transition temperatures is not known. Here, we present the field-temperature phase diagram for two different directions of the single crystalline samples of SrCuTe$_2$O$_6$ that shed light on the magnetic properties of the compound. Further, we investigate the polycrystalline samples with $\mu^+$SR and neutron powder diffraction measurements and propose a model for the zero-field magnetic structure in the ordered state. The results reveal that the first neighbor triangle interaction provides the interchain coupling and is responsible for the long-range order in the system.

### I. SAMPLES & EXPERIMENTAL METHODS

Polycrystalline powder of SrCuTe$_2$O$_6$ was prepared from stoichiometric mixture of high purity powders of SrCO$_3$ (99.99%), CuO (99.995%) and TeO$_2$ (99.99%) by solid state reactions at 650°C in a vacuum furnace under Argon flow. For crystal growth, first stoichiometric amount of high purity SrCO$_3$, CuO and TeO$_2$ were mixed as above and sintered twice for 12 hours at 600°C in Argon flow with intermediate grinding. Then a feed rod (D=6 mm, L=7-8 cm) was prepared from the stoichiometric powder and densified by pressing in a Cold Isostatic press in 2000 bars and subsequent sintering at 650°C in Argon flow. Crystal growth was done using the seed-rod by the Floating zone technique in a four mirror type optical image furnace (Crystal Systems Corp., Japan). Growth was done at a rate of 1 mm/hr in Argon atmosphere at ambient pressure. The as-grown crystal is approximately 5 mm diameter and 3.5 cm in length. The as-grown crystal was checked by X-ray Laue diffraction for single crystallinity and confirmed by polarized optical microscopy to be free of inclusions. The quality of the crystal has also been analyzed for phase purity by grinding a small piece of crystal in to powder upon which X-ray diffraction was performed. These single crystals reveal a small quantity of non-magnetic impurity in the form of Sr$_2$Te$_3$O$_8$ amounting to less than 1%. The single crystals were then characterized by magnetic susceptibility, magnetization and heat capacity in the temperature range of 1.8–400 K and an external field of 0–7 T using a Physical Property Measurement System (PPMS). The sample synthesis and characterization took place at the Core Lab for quantum Materials, Helmholtz-Zentrum Berlin, Germany.

$\mu^+$SR measurements on the polycrystalline SrCuTe$_2$O$_6$ were performed at the General purpose Spectrometer at the SMuS facility in Paul Scherrer Institut down to 1.6 K in zero field. The nuclear and magnetic structure of SrCuTe$_2$O$_6$ was investigated between 20 K and 1.6 K by obtaining neutron diffraction patterns on powder sample of 10 g. An initial search for the magnetic Bragg peaks was carried out using the DMC diffractometer \[^22\] at the Paul Scherrer Institut, Switzerland using two incident wavelengths $\lambda$=2.46 Å 4.504 Å (PG002 monochromator) covering a momentum transfer $Q$ in the range of 0.2 Å$^{-1}$ < $Q$<3.7 Å$^{-1}$ and 0.35 Å$^{-1}$ < $Q$<2 Å$^{-1}$ respectively. The diffraction patterns were collected at 1.6 K, 5.2 K and 20 K. Detailed temperature dependence of the nuclear and magnetic structure on the powder sample was performed at the time-of-flight diffractometer WISH \[^23\] at the ISIS facility, UK. The patterns were collected for temperatures between 1.5 K and 15 K and momentum transfer 0.37 Å$^{-1}$ < $Q$<9 Å$^{-1}$. In both cases, the powder was loaded into a cylindrical vanadium can and the temperature was controlled using a typical orange cryostat. The patterns are refined using the Reitveld method.

| Atom | Wyckoff position | $x/a$ | $y/a$ | $z/a$ | $B_{iso}$ |
|------|------------------|------|------|------|---------|
| Te   | 24e              | 0.33775 | 0.91970 | 0.058890 | 0.46001 |
| Sr1  | 8c               | 0.05335 | 0.05335 | 0.05335 | 0.65537 |
| Sr2  | 4b               | 0.87500 | 0.87500 | 0.87500 | 0.61456 |
| Cu   | 12d              | 0.12500 | 0.77446 | 0.02445 | 0.47196 |
| O1   | 24e              | 0.57936 | 0.92944 | 0.47196 | 0.81156 |
| O2   | 24e              | 0.26670 | 0.81156 | 0.97806 | 0.49215 |
| O3   | 24e              | 0.22239 | 0.97760 | 0.12925 | 0.53796 |

#### TABLE I. The Reitveld refined coordinates and isotropic thermal parameters of SrCuTe$_2$O$_6$ at 7 K.
in the Fullprof package [24] and magnetic symmetry analysis was performed using a combination of BasiReps and Bilbao crystal server software packages [25]. Fig. 1 shows the neutron powder diffraction of the nuclear structure taken at 7 K at the WISH diffractometer. The refinement agrees with the non centro-symmetric cubic structure space group: $P4_{1}32$, consistent with previously reported results [16, 17] at room temperature. The lattice constant at 7 K is found to be 12.4373(2) Å. The refined values of the coordinates and thermal factors are listed in Table. II.

II. RESULTS

A. Magnetic properties of single crystal

Figure 2a shows the zero-field-cooled dc-magnetic susceptibility of the polycrystalline and single crystal samples in a bias field of $H=0.05$ T revealing several important clues to the magnetic state of the system (1.8 K-400 K). At high temperatures, the inverse susceptibility is linear (fig. 2b) and can be fitted to paramagnetic Curie-Weiss (CW) behaviour: $\chi = \chi_{\text{core}} + \chi_{\text{vv}} + \frac{C}{T-\theta_{\text{CW}}}$, where $\chi_{\text{core}}=-1.54 \times 10^{-4}$ is the diamagnetic contribution from the core non-magnetic ions $\text{Te}^{4+}$ ions and $\chi_{\text{vv}}$ refers to Van Vleck paramagnetism. In order to obtain reliable values of the Curie-Weiss temperature $\theta_{\text{CW}}$, we have varied the lower bound of the temperature range of the fits from 100 K to 200 K. The best fits are obtained for 140 K-400 K and the resulting fit parameters $\chi_{\text{vv}}$, Curie-Weiss constant $C$, $\theta_{\text{CW}}$ along with the derived $\mu_{\text{eff}} = 3Ck_BN_A/\mu_B$ and $g$–factor are tabulated in Table. II. The values of $\theta_{\text{CW}}$ are: $-28 \pm 0.3$ K, $-28 \pm 1$ K, $-26 \pm 1$ K for polycrystalline, crystalline (100) axis and the crystalline (110) axis respectively. Within the sensitivity of the measurement and of demagnetization effects due to the shape of the crystal, the single crystal susceptibility in both crystalline directions follows that of the polycrystalline sample hence confirming the isotropic nature of the Cu$^{2+}$ spins in SrCuTe$_2$O$_6$. Furthermore, the negative $\theta_{\text{CW}}$ values confirm the predominant antiferromagnetic interactions in the system. The effective moment calculated from the Curie-Weiss constant is $\sim 1.85\mu_B$ which is very close to the full moment of the free Cu$^{2+}$ spin. Accordingly, the derived $g$–factor is close to 2.1 in the three measurements assuming spin-1/2. We find that the $\theta_{\text{CW}}$ values are smaller than the previously reported $\theta_{\text{CW}} = -35$ K in polycrystalline samples [16, 17]. The discrepancy could be attributed to the sensitivity of the $\theta_{\text{CW}}$ to the fitted temperature range.

In the intermediate temperature range, all the three data sets exhibit a broad hump around $\sim 32$ K indicative of short-range magnetic correlations, characteristic of 1D Heisenberg spin-$\frac{1}{2}$ chain compounds. The solid
grey lines in fig. [2]a are a fit (T>15 K) to the high-temperature series expansion for the DC susceptibility of a spin-\(\frac{1}{2}\) Heisenberg antiferromagnetic chain [20] [21]:

\[
\chi = \chi_{\text{core}} + \chi_{vv} + \frac{N_A \mu_B^2 g^2}{4k_B T} \times \frac{1 + 0.08516 x + 0.23351 x^2}{1 + 0.73382 x + 0.13696 x^2 + 0.05356 x^3}
\]

where \(J_{\text{chain}}\) in \(x = J_{\text{chain}}/T\) is the chain interaction which is also the 3rd nearest-neighbour interaction in case of SrCuTe\(_2\)O\(_6\). The g-factor and \(\chi_{vv}\) are also fitted within this model and the resulting parameters are tabulated in Table. III. The model yields a chain interaction \(J_{\text{chain}} \approx 49\) K and a g-factor of \(\sim 2.2\) in the single crystal. The observed g-factor, although slightly higher than the fully isotropic spin system, it is consistent with the values obtained from high temperature Curie-Weiss behaviour. In Heisenberg systems the Curie-Weiss temperature is the weighted sum of all the relevant magnetic interactions:

\[
\theta_{cw} = - \frac{S(S+1)}{3k_B} (2J_1 + 4J_2 + 2J_3)
\]

taking \(J_3 = 49\) K, the triangle-based inter-chain couplings in SrCuTe\(_2\)O\(_6\) sum to \(J_{\text{inter}} = J_1 + 2J_2 = 8\) K suggesting that they are antiferromagnetic and frustrated. As a result, SrCuTe\(_2\)O\(_6\) exhibits magnetic transitions at the temperatures \(T_{N1} = 5.5\) K, \(T_{N2} = 4.5\) K which are much lower than the Curie-Weiss temperature. They are revealed as peaks in the first derivative of the susceptibilities plotted in fig. [2]. To confirm the presence of magnetic transitions, heat capacity of the single crystal has also been measured. As shown in the fig. [3] the phonon contribution (\(C_{\text{phonon}}\)) of the high temperature heat capacity (T> 50 K) is very well described by a sum of two Debye integrals allowing the extraction of the dominant magnetic contribution at low temperatures.

The obtained magnetic quantity \(C_{\text{mag}}/T\), where \(C_{\text{mag}} = C_p - C_{\text{phonon}}\), shows a broad peak at \(\approx 16.2\) K (see lower inset of fig. [3]). This is a characteristic feature observed in Heisenberg spin-1/2 antiferromagnetic chains [21] [22] which relates to the chain interaction \(J_{\text{chain}}\) as:

\[
\frac{T_{\text{max}}}{C_{\text{mag}}/T} \approx 0.3072
\]

**TABLE II.** The Curie-Weiss temperature, effective moment, and the g-factor as derived by Curie-Weiss fit to the high temperature magnetic susceptibility (T>140 K) of the powder sample and single crystal sample aligned parallel to external field along the (100) and (110) directions.

| Sample | \(\chi_{vv}(\times 10^{-5})\) (cm\(^3\)/mol) | C (cm\(^3\)·K/mol) | \(\theta_{cw}\) (K) | \(\mu_{eff}\) (\(\mu_B\)) | g-factor |
|--------|---------------------------------|-----------------|-----------------|-----------------|--------|
| Powder | 4.49±0.01                       | 0.41±0.008      | 28.44±0.3       | 1.82            | 2.1    |
| (100)  | 6.95±0.05                       | 0.43±0.003      | 27.94±1         | 1.87            | 2.16   |
| (110)  | 5.38±0.06                       | 0.426±0.0003    | 26.15±1         | 1.85            | 2.13   |

**TABLE III.** The chain interaction strength and g-factor as derived by fitting the magnetic susceptibility above \(T_{N1}\) (T>15 K) of the powder sample and single crystal sample aligned parallel to external field along the (100) and (110) directions.

| Sample | \(\chi_{vv}(\times 10^{-5})\) (cm\(^3\)/mol) | g-factor | \(J_{\text{chain}}\) (K) |
|--------|---------------------------------|--------|-----------------|
| Powder | 3.85±0.1                        | 2.12±0.005 | 49.1±0.02 |
| (100)  | 3.41±0.1                        | 2.19±0.006 | 49.84±0.02 |
| (110)  | 1.59 ± 0.11                     | 2.18±0.006 | 50.09±0.02 |

**FIG. 4.** a-b) Magnetization of SrCuTe\(_2\)O\(_6\) at 1.6 K measured in pulsed field and DC field at 2 K applied along the two crystalline directions (100) and (110) respectively. Insets: derivatives of magnetization measured in dc-field at 2 K. c-d) Magnetization curves measured at several temperatures in the dc-field for the two crystalline directions and, e-f) show the evolution of the derivatives of the magnetization indicating new field-induced anomalies.

giving \(J_{\text{chain}} = 52.5\) K, in close agreement with the results from susceptibility. Two \(\lambda\)-like anomalies are observed at lower temperatures \(T_{N1} = 5.5\) K and \(T_{N2} = 4.5\) K (top inset of fig. [3]). These transitions are consistent with the previous reports in the polycrystalline samples.

In order to explore the effects of magnetic field
FIG. 5. Heat capacity of SrCuTe$_2$O$_6$ at several constant magnetic field applied parallel to the crystalline (100) and (110) directions.

FIG. 6. H-T phase diagram of single crystal of SrCuTe$_2$O$_6$ with external field applied a) along (100) direction and b) along (110) direction.

on SrCuTe$_2$O$_6$, magnetization measurements were performed at various temperatures. High field magnetization at T=2 K using a pulsed magnet, as well as lower field DC magnetization measurements along the (100) and (110) direction respectively are presented in fig. 4a-b. The pulsed field measurements were normalized by the DC magnetization and reveal that the Cu$^{2+}$ moment reaches 0.5 $\mu_B$ at 56 T. Considering a linear extrapolation, the saturation field can be expected at $\approx$110 T.

At lower fields, two sets of anomalies are observed in the derivative of magnetization along (100) direction indicating possible field-induced magnetic transitions in the single crystal of SrCuTe$_2$O$_6$. As shown in the inset of fig. 4a, these anomalies occur at $\approx$ 4.2 T and 5.5 T accompanied by shoulder peaks at 3.98 T and 5.13 T. Magnetization along crystalline (110) direction at 2 K (see fig. 4b) also reveals three anomalies at $\approx$ 3 T, 4.2 T and 5.5 T. These anomalies were followed as a function of temperature for the two directions of the single crystal (see fig. 4c-d) as well as for the polycrystalline sample. The derivative of magnetization dM/dH in Fig. 4e shows that the anomalies give rise to sharp and strong peaks when the field is applied along (100) direction. With increasing temperature, the lower peak shifts to lower fields up to $T_N^2=4.5$ K whereas the higher peak (5.5 T) shows a slight shift towards higher fields and disappears above 5 K. We observe that the shoulder peaks essentially move along with the main peaks. We believe this is due to a smaller crystallite within the sample with a misaligned (100) direction.

Along the (110) direction, the peaks in the dM/dH are much weaker compared to the (100) direction, however, their position moves towards higher fields gradually up to $T_N^2=4.5$ K where the highest field peak reaches a maximum of 6 T as shown in fig. 4f. Only the highest field anomaly survives in the intermediate phase between $T_{N2}=4.5$ k and $T_{N1}=5.5$ K similar to the (100) direction.

The A-like features corresponding to $T_{N1}$ and $T_{N2}$ in the specific heat also exhibit a significant field dependence in both the directions (see fig. 5). We observe that the anomalies along (100) direction become sharper (red squares) in the external field along with the introduction...
of a new transition anomaly at lower temperatures up to 6 T. Above this field, a single, broad anomaly is seen. The phase transitions along the (110) direction (blue circles) also follow with those of the (100) direction. However, an additional transition anomaly is observed at 5 T consistent with the anomalies observed in the magnetization of the crystal along (110) direction. Combining these observations, the phase diagram is then constructed for each of the crystal directions separately along with the polycrystalline sample.

Figure 6 shows that phase diagram of the single crystalline SrCuTe₂O₆ along (100) direction identifies three possible magnetic phases in the systems. Here, phase-I refers to the magnetic ground state, phase-II is an intermediate phase and the phase-III, where heat capacity shows a broad λ, referring to ferromagnetic canting of the spins. These results are similar for polycrystalline sample and in good agreement with the previously reported results [16–18]. Whereas an additional phase-IV (and phase-V) are also observed along the (110) direction indicating additional phase transitions preferentially involving spins along (110) direction.

B. Muon Spin Relaxation

To obtain more insight into the nature of the magnetic order below the two transitions T₁ and T₂ in SrCuTe₂O₆ we further probe the material with muon spin relaxation (µ⁺SR) experiments in zero magnetic field between 2 K and 10 K. Figure 6 shows the µ⁺SR spectrum of SrCuTe₂O₆ as a function of decay time at several temperatures in the ordered state (T<T₁,N=5.5 K) and in the paramagnetic state T=6 K. At base temperature, the spectrum clearly reveals the oscillatory behavior of the asymmetry resulting from the Larmor precession of the muon spin around the local internal field set by the magnetic ordering in the system. Furthermore, the remnant relaxation at long time-scales saturates at 1/3 of the initial value of the asymmetry. These observations are a typical indications of static magnetic order in the system.

The Fourier transform (FFT) of the oscillating spectra reveals nine frequency components at base temperature as shown in fig. 6a-b and their distribution varies as the temperature increases towards T₂ (fig. 6c-g). Therefore, all the spectra below T₂ are fitted by considering a superposition of nine Gaussian-distributed internal magnetic fields to describe the precessing part of the spectrum as described in the following model:

\[
G_z(t) = A \left[ \frac{2}{3} \sum_{i=0}^{9} A_{i} \cos(2\pi \nu_{i} t)e^{-\lambda_{i} t} + \frac{1}{3}e^{-\lambda_{L} t} \right]
\]

where, A is the total asymmetry, fₙ is the sample fraction which is further separated into 2/3 Cosine-oscillating term and 1/3 non-oscillating relaxing term at long time-scales. The latter term implies the relaxation (λₜ) of those muons whose spin is longitudinal to the internal field at the time of decay and hence is indicative of the spin dynamics in the system. Consequently, λₜ peaks up at T₂=4.5 K and T₁,N=5.6 K, as shown in fig. 6a reflecting the critical dynamics at the magnetic transitions in SrCuTe₂O₆. The field distribution below T₂ is clearly separated into nine components (as explained above) with the strongest frequency occurring at ν=2.4 MHz. This refers to an internal field of 0.18 kOe with a small field distribution (gaussian width) of Δν=0.729 MHz=5 mOe. Above T₂,N, the nine frequency components collapse into a broad gaussian (fig. 5b), which can be adequately described by considering a maximum of three frequencies. This gaussian gradually moves to smaller frequencies and completely vanishes above the highest transition at T₁,N=5.6 K.

The ZF-µSR spectra clearly reveal two different magnetic phases with distinguishing internal field distributions. We can attribute the origin of these frequencies to a composite of the muon sites around three inequivalent Oxygen sites (with three Cu-O bond lengths: 1.939 Å, 1.943 Åand 3.086 Å) and three effective local spin directions of the 12 Cu moments with respect to the incoming µ⁺ spin.
C. Magnetic structure

To investigate the magnetic structure of SrCuTe$_2$O$_6$ in the ground state, i.e., below $T_{N2}=4.5$ K, several powder diffraction patterns are obtained between temperatures 1.7 K and 7 K. Representative low temperature diffraction patterns of SrCuTe$_2$O$_6$ obtained on the DMC diffractometer are plotted in fig. 9a for polycrystalline sample at the base temperature 1.7 K, in the intermediate magnetic phase at 5.2 K and in the paramagnetic state at 20 K. These patterns reveal that the nuclear structure of the SrCuTe$_2$O$_6$ remains unchanged even below the magnetic transition. The subtracted patterns below $T_{N1}=5.5$ K, in fig. 9b, clearly reveal two magnetic peaks at the $d$ -- spacing of 12.4Å, 5.6Å corresponding to (100), (210). In addition, magnetic intensities are also clearly visible on the weak nuclear peaks (300)+(221),(310) and (311) at 4.2Å, 4Å and 3.8Å $d$-spacing. The contribution of magnetic intensity on the strong nuclear peaks is ambiguous. Although the structural peaks at $(2h+1,0,0)$ are allowed for the primitive type of unit cell, the fourfold screw symmetry of space group $P4_132$ forbids these peaks while allowing only those with $h=4n$. Therefore, the magnetic propagation vector can be identified as $q_m=(000)$.

Representation analysis for the propagation vector

FIG. 8. a) Longitudinal relaxation $\lambda_L$ of the $\mu$SR spectra and, b) map of the Larmor precession frequencies, proportional to the order parameter, below the magnetic transitions in polycrystalline SrCuTe$_2$O$_6$.

FIG. 9. a) Powder neutron diffraction patterns measured at the DMC diffractometer below the magnetic transitions at 1.7 K, 5.2 K and above at 20 K. b) The difference patterns with respect to 20 K reveals several magnetic peaks.

FIG. 10. a-c) Rietveld refinement of the magnetic intensities measured at the WISH diffractometer at 1.6 K (obtained by subtracting the intensity at 7 K) using three different irreducible representations of the magnetic structure for SrCuTe$_2$O$_6$. 
(0,0,0) reveals that the reducible magnetic representations \((\Gamma_{mag})\) associated with the 12d Wyckoff position of Cu decomposes into direct sum of five irreducible representations (IRs) denoted as \(\Gamma_i\) \((i=1-5)\). We use superscript to indicate dimensionality of the IRs:

\[
\Gamma_{mag} = 1\Gamma_1^1 + 2\Gamma_1^2 + 3\Gamma_2^2 + 4\Gamma_3^3 + 5\Gamma_3^3
\]

(4)

Following the standard approach, the solution of the magnetic structure was searched assuming a single IR (irreducible magnetic order parameter). For the three-dimensional IRs \(\Gamma_4\) and \(\Gamma_5\), only high-symmetry combinations of the basis functions corresponding to maximal isotropy subgroups \([27]\) were tested. The low-symmetry magnetic structures require strongly first order phase transition and are unlikely from the thermodynamic point of view. The systematic absence of the \((2h,0,0)\) magnetic reflections is inconsistent with the \(\Gamma_4\) and \(\Gamma_5\) IRs, while discrimination between \(\Gamma_1\), \(\Gamma_2\) and \(\Gamma_3\) were more challenging. As the changes on the strong nuclear peaks such as \((110)\), \((111)\) and \((211)\) are not clear, these regions are excluded from the analysis while refining the magnetic structure. For this we used high intensity datasets collected on the WISH time-of-flight diffractometer. The magnetic intensity was obtained by subtracting the 7K data from the 1.5 K dataset.

Figure 10a-c show individual refinements of the magnetic peaks for IRs \(\Gamma_1\), \(\Gamma_2\) and \(\Gamma_3\) respectively. All the three representations reproduce the strongest magnetic peak \((100)\) \((d=12.438\text{Å})\) very well with the differences in fit quality appearing only at high-Q peaks such as \((221)+(300)\) \((d=3.933\text{Å})\) and \((310)\) \((d=4.146\text{Å})\) resulting in a best magnetic Bragg-factor \(2.93\) from the first IR, \(\Gamma_1\). The corresponding magnetic structure implies the cubic magnetic symmetry \(P4_{1}32\) \((\#213.63)\) with the basis and origin defined in respect of the paramagnetic space group as: \((1,0,0)\), \((0,1,0)\), \((0,0,1)\) and \((-1/4,-1/4,-1/4)\), respectively. In this magnetic structure, each of the Cu-spins is aligned along a local \((110)\) direction. Here, the third nearest neighbours of \(\text{Cu}^{2+}\) forms antiferromagnetic spin-\(\frac{1}{2}\) chains running along the three mutually perpendicular crystallographic \(a\)-, \(b\)- and \(c\)- axes. Furthermore, we observe two parallel chains per cubic direction, as shown in fig. 11a for chains along \(a\)- axis, whose spins take on two perpendicular spin directions in the \(b-c\) plane, \((011)\) and \((01-1)\). This results in a total of 6 spin directions in the ordered state of SrCuTe\(_{2}\)O\(_{6}\) so that the frustrated first nearest-neighbour interaction \(J_1\) forms coplanar \(120^\circ\) triangles as shown fig. 11b. Although these triangles are isolated from each other, spins on the vertices of the every triangle participates in coupling the three perpendicular spin-chains leading to three dimensional magnetic order in the system. These \(J_1\) rather than the hyperkagome interactions \(J_2\), are responsible for the inter-chain coupling.

We observe that the magnetic propagation vector remains unchanged even in the intermediate phase within the instrumental resolution. Therefore, the pattern in this temperature range is also refined by the same magnetic structure resulting from \(\Gamma_1\) representation at the base temperature 1.7 K showing the two chains propagating along each of the cubic axes within a single unit cell. Spins in a chain are perpendicular to those in the neighboring parallel chain in the same direction. b shows the inter-chain coupling promoted by first nearest neighbour interaction \(J_1\). c shows the temperature dependence of the ordered moment refined by considering the magnetic structure from \(\Gamma_1\) at all the temperature below 7 K.

![FIG. 11. Magnetic structure of SrCuTe\(_{2}\)O\(_{6}\) described by \(\Gamma_1\) representation at the base temperature 1.7 K showing the two chains propagating along each of the cubic axes within a single unit cell. Spins in a chain are perpendicular to those in the neighboring parallel chain in the same direction. b shows the inter-chain coupling promoted by first nearest neighbour interaction \(J_1\). c the temperature dependence of the ordered moment refined by considering the magnetic structure from \(\Gamma_1\) at all the temperature below 7 K.](https://example.com/figure11.png)
which yields a value of of $m_0 \approx 0.41 \mu_B$ for SrCuTe$_2$O$_6$ considering $J_{\text{inter}}=8$ K and $J_{\text{chain}}=50$ K. While this value is consistent with the experimental moment at the base temperature, it also confirms the presence of weak antiferromagnetic inter-chain coupling responsible for the loss of 60% of full moment expected for fully ordered Cu$^{2+}$ spin as would be found in a 3D ferromagnet. As the errorbars of the moment obtained from powder diffraction are high, we have also followed the intensity of the magnetic Bragg peak (300) in the single crystal of SrCuTe$_2$O$_6$ (right $y$-axis of fig. 11c) which clearly indicates a non-zero intensity below the first magnetic transition $T_{N1}=5.5$ K. However, no significant changes are observed at the lower transition $T_{N2}=4.5$ K.

**DISCUSSION**

As explained in the sec. II C, the Cu spins form a co-planar 120° structure in the ground state coupling three mutually perpendicular AF chains so that each of the spin points along a local (110) direction. This picture may also explain the origin of the 9 Larmor frequencies observed from powder diffraction. In a simplistic point of view, Cu spin in SrCuTe$_2$O$_6$ takes three orientations per triangle (120° structure) which can lead to multiples of three Larmor frequencies for the muon spin. Additionally there are three more degrees of freedom for the muon site due to the three inequivalent Oxygen sites, which together can give rise to 9 different local fields/precession frequencies. Whereas, the presence of only three frequencies in the intermediate phase could be explained by a possible canting of the spins about the (110) which in turn reduces the contrast between the magnetic (110) directions, leaving only the crystallographically inequivalent oxygen sites. However, we note that there is no indication for an incommensurate spin structure as the field distribution is always gaussian-like pointing to a preferred local internal field instead of structural changes occur at the transitions to the long-range magnetic order. These changes are likely to be much smaller in zero field such as symmetry allowed displacements which retain the nuclear space group. Hence, no visible changes were observed on the nuclear peaks in powder diffraction as that of the magnetic phase transitions observed in polycrystalline and crystalline (100) directions as a function of field. It would therefore not be surprising if antiferromagnetic order also influenced the structure so that structural changes occur at the transitions to the super-exchange pathways as proposed by Koteswararao et al. [16]. For instance the ratio of bond angles responsible for $J_2$ (Sr:92.5°, Pb:97°) and $J_3$ (Sr: 162.2°, Pb: 156°), $J_2$-angle/$J_3$-angle, is $\approx 9$% higher in PbCuTe$_2$O$_6$ compared to SrCuTe$_2$O$_6$. In addition, the extra lone-pair in PbCuTe$_2$O$_6$ might play a key role in the weaker chain interaction due to the hybridization of the Pb-O bonds, involved in the $J_3$ superexchange path (O-Pb2-O), that may have extra strain effects as in ferroelectric perovskite systems [30]. Confirmation of this needs a detailed investigation into the electronic band structure of both the systems, which is beyond the scope of this work.

Koteswararao et al. [18] find magnetoelastic effects in the form of electric polarization at magnetic transitions in SrCuTe$_2$O$_6$ in an applied magnetic field manifesting a strong coupling between magnetism and lattice. The field-induced polarization also resulted in a similar phase diagram as that of the magnetic phase transitions observed in polycrystalline and crystalline (100) directions as a function of field. It would therefore not be surprising if antiferromagnetic order also influenced the structure so that structural changes occur at the transitions to the long-range magnetic order. These changes are likely to be much smaller in zero field such as symmetry allowed displacements which retain the nuclear space group. Hence, no visible changes were observed on the nuclear peaks in powder diffraction patterns. However, heat capacity results in field (see fig. 5) reveal a sharper $\lambda$-anomaly above 3 T at $T_{N1}$, consistent with the field induced electric polarization. Therefore, investigation of magnetic structure of SrCuTe$_2$O$_6$ in an external field would give insight into the origin of the spin-lattice coupling.
SUMMARY

In summary, we have studied magnetic properties of SrCuTe$_2$O$_6$ in polycrystalline and single crystal samples and investigated the magnetic structure. The field-dependent phase diagram in single crystals reveals additional magnetic phases for the (110) direction whereas the (100) direction replicates the phase diagram of the polycrystalline sample. We propose a magnetic structure of SrCuTe$_2$O$_6$ where, $J_1$ acts as an inter-chain coupling to the AF chains formed by $J_3$ leading to three dimensional magnetic ordering in the system below $T_{N1}$.

Note: As this paper was being finalized we became aware of a similar investigation of SrCuTe$_2$O$_6$ on arXiv [31]. While there are some differences in the techniques employed, the results of that paper are in broad agreement with this paper.

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