

# $\mu$ SR Measurements of the Magnetism of $\text{Sr}_3\text{Fe}_2\text{O}_7$

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**Abstract.** We present a preliminary report of zero and transverse field  $\mu$ SR measurements in a high quality single crystal of fully oxygenated  $\text{Sr}_3\text{Fe}_2\text{O}_7$ [1], the insulating  $3d$  magnetic analogue of the well studied  $\text{Sr}_3\text{Ru}_2\text{O}_7$ , in its charge-disproportionated state, through the magnetic transition to the incommensurate phase at  $T_N \sim 115$  K. A very broad distribution of static fields below  $T_N$  yields a large ( $2/3$ ) missing fraction. Rapidly damped zero field oscillations indicate a large internal field at the muon, estimated to be 0.46 T at 0 K. In the paramagnetic state, we find two muon precession frequencies at 0.25 T applied field, reminiscent of the Mössbauer spectra.

## 1. Introduction

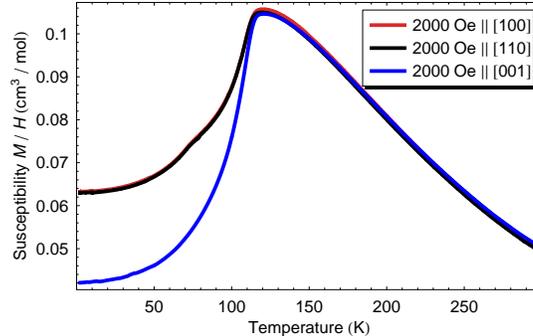
The Ruddlesden-Popper series of mixed alkaline-earth transition-metal oxides, with general formula  $\text{A}_{n+1}\text{M}_n\text{O}_{3n+1}$ , exhibit a wealth of interesting properties, such as superconductivity, colossal magnetoresistance, and charge, orbital, and spin order. Among these, the fully oxygenated  $n = 2$  compound  $\text{Sr}_3\text{Fe}_2\text{O}_7$  offers unique insight into the differences between the magnetic properties of the manganates, ruthenates, and ferrates.  $\text{Sr}_3\text{Fe}_2\text{O}_7$  crystallizes in a tetragonal layered perovskite structure (space group  $I4/mmm$ ) and its crystal structure contains double sheets of  $\text{FeO}_6$  octahedra. Mössbauer spectroscopy indicates charge disproportionation of Fe (nominally  $2\text{Fe}^{4+} \rightarrow \text{Fe}^{3+} + \text{Fe}^{5+}$ , but realistically the charge difference is likely substantially less) at  $T_{charge} = 340$  K[2]. The layered crystal structure of this compound gives rise to interesting quasi-two-dimensional magnetic properties. The analogous (metallic) ruthenate compound  $\text{Sr}_3\text{Ru}_2\text{O}_7$  has been studied extensively, both in relation to the well-known related  $\text{Sr}_2\text{RuO}_4$  superconducting phase, as well as for its metamagnetism. This has been interpreted as a magnetically tuned quantum critical transition[3], with a high field state that may be an electronic nematic analogous to quantum Hall systems[4]. In contrast,  $3d$  Fe has stronger electron interactions (Hubbard  $U$ ) and smaller bandwidths, making  $\text{Sr}_3\text{Fe}_2\text{O}_7$  nonmetallic at ambient pressure below  $T_{charge}$ <sup>8</sup>, but exhibits interesting low dimensional magnetism that may bear some relation to the analogous  $4d$  ruthenate. The two dimensionality is expected to result

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<sup>8</sup> It conducts above  $\sim 15$  GPa and, at ambient pressure, above  $T_{charge}$ .

in a strongly 2D correlated paramagnetic state, with long range order brought about by small 3D couplings. Here we present preliminary  $\mu$ SR results on the low field magnetic state of fully oxygenated  $\text{Sr}_3\text{Fe}_2\text{O}_7$  in the range from 300 K to 2 K.

## 2. Experiment



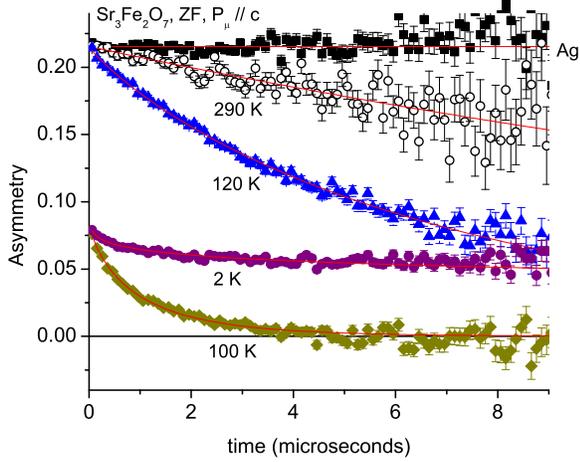
**Figure 1.** The molar magnetic susceptibility of  $\text{Sr}_3\text{Fe}_2\text{O}_7$  as a function of temperature in a magnetic field of 0.2 Tesla applied along three different high symmetry directions of the tetragonal single crystal. The two in-plane directions are nearly identical.

We have synthesized and oxygen-annealed large ( $\text{cm}^3$  sized) single crystals of  $\text{Sr}_3\text{Fe}_2\text{O}_7$ . The oxygen content was confirmed by thermogravimetric analysis and refinement of neutron diffraction[1]. The DC magnetization indicates an antiferromagnetic transition at Néel temperature  $T_N \sim 115$  K, e.g. see Fig. 1. The single crystal boule was mounted on a low background sample frame with thin aluminized mylar tape. A  $c$ -axis facet on the sample was aligned within a few degrees of the beam direction in the LAMPF spectrometer on the M15 beamline at TRIUMF. Two measurements were performed: 1) a zero applied field (ZF) experiment with the muon spin polarized along the beam direction parallel to the crystalline  $c$ -axis and 2) a precession experiment in an applied transverse field (along  $c$ ) with the muon spin rotated (TF). Temperature was varied using a helium gas flow cryostat. Calibration measurements were made on a high purity Ag blank sample before the measurement.

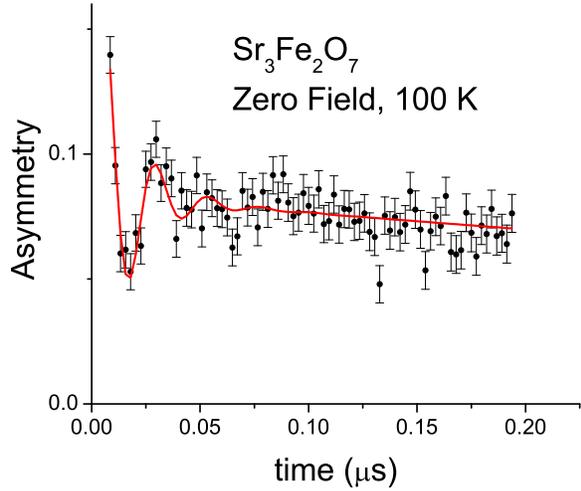
## 3. Results and Discussion

The ZF data shows an increasing relaxation rate as the sample is cooled towards  $T_N$ , consistent with a slowing down of spin fluctuations, see Fig. 2. Below  $T_N$  there is a very fast relaxing component, that exhibits measurable, but extremely rapidly damped oscillations over a narrow range of temperature (Fig. 3). Below 75 K, this signal is lost as its relaxation becomes too fast. There is also a non-oscillating slower relaxing component that, at low temperature, accounts for about 1/3 of the total asymmetry in the paramagnetic state. This signal, likely corresponding to muons stopping in a local longitudinal field, can be fit to a single exponential in the vicinity of  $T_N$ , but below 75 K is better described by a stretched exponential,  $\exp(-\sqrt{t}/T_1)$ . The relaxation rates from such fits are shown in Fig. 4a). Interestingly, the relaxation rate does not peak close to  $T_N$  but rather continues to increase well below the transition, eventually slowing below 50 K.

We fit the oscillating signal with a rapidly damped exponential and could extract the frequency reliably at only two temperatures, Fig. 4b). This signal corresponds to muons in



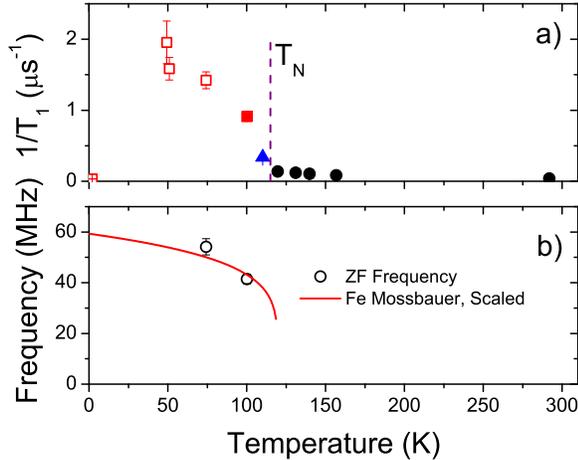
**Figure 2.** Temperature dependence of the zero field  $\mu$ SR spectra. Below  $T_N$  there is a large missing fraction. The nonrelaxing calibration spectrum in Ag at 290 K is shown for comparison.



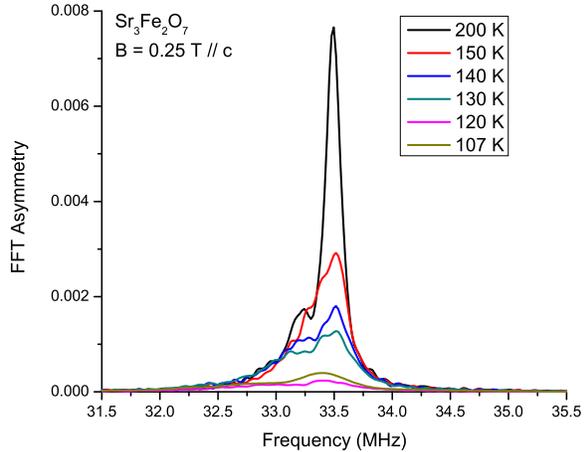
**Figure 3.** Rapidly damped zero field oscillations at 100 K are visible in the early times of the same data shown in Fig. 2.

large (4.0(2) kG at 75 K) local transverse fields, in the range seen in related Fe oxides, such as  $\text{YFeO}_3$ [5] and Co-doped  $\text{SrFeO}_3$ [6]. We note it is not the frequency that limits our ability to follow this signal (with a time resolution of a few ns), rather it is the very fast damping that eliminates it within the first 50 ns or less. The rapid damping is consistent with a magnetic state with a broad distribution of local fields, likely a consequence of the incommensurate magnetic order[1]. In order to extrapolate the field at the  $\mu^+$  to low temperature where the oscillations are lost, we fit the published temperature dependence of the Mössbauer splitting[2] to a smooth function and scaled it to the ZF frequency, see Fig. 4b), yielding a  $T = 0$  frequency of 59(3) MHz for the  $\mu^+$ . Once the details of the magnetic order are firmly established, we will be able to use the precession signal to infer the muon site in this, the simplest fully oxygenated end-member compound. This will be important for studying the full field, temperature and doping phase diagram.

Measurements in transverse field in the paramagnetic charge disproportionated state reveal a long-lived precession above  $T_N$  characterized by two distinct, but not very well-resolved, frequencies, see Fig. 5. Below  $T_N$  the precession is rapidly damped by the broad distribution of static internal fields. In preliminary analysis, the splitting appears to increase with decreasing temperature (though the linewidth is similarly increasing). Here one might expect a muon shift to scale with the susceptibility (Fig.1). It is remarkable that there are two frequencies. The obvious interpretation is that there are 2 distinct muon sites with different couplings to the surrounding Fe moments. This seems possible in the disproportionated state, where there are already 2 distinct Fe sites, but it is not clear how to obtain only two muon sites. The  $\mu^+$  sites are likely adjacent to an  $\text{O}^{2-}$ , of which 2 of the 7 are away from the Fe layer and 5 bridge adjacent Fe ions. In the simplest charge alternating structure, analogous to  $\text{CaFeO}_3$ , these would be equivalent. It would be interesting to see if this difference collapses above  $T_{charge}$ . One may expect a better resolved spectrum at higher applied field, that might be more easily tracked with temperature. The bulk  $\chi$  above  $T_N$  appears unaffected by field, so we do not anticipate field induced changes, but the TF relaxation rate might also scale with field. Note that the



**Figure 4.** a) Temperature dependence of the zero field spin relaxation rate. From single (solid symbols) and stretched (open symbols) exponential fits to the relaxation with the full (circles), 1/3 (squares), and intermediate (triangle) asymmetry. b) The ZF oscillation frequency with the Mössbauer hyperfine field for the two Fe sites from Ref. [2] fit to a smooth function and scaled to match the observed frequencies.



**Figure 5.** Temperature dependence of the fast Fourier transform of the transverse spin precession signal in an applied field of 0.25 T. The line is clearly two component, with a low frequency shoulder. The precession signal is rapidly damped as  $T$  approaches  $T_N$ .

Mössbauer shows Fe sites that differ in hyperfine field by about 50% in the magnetic state and with a very different chemical shift in the paramagnetic state. In contrast, if the structure in the TF- $\mu\text{SR}$  precession is due to two muon sites, they are rather similar.

#### 4. Summary

In summary, we have studied the magnetism of  $\text{Sr}_3\text{Fe}_2\text{O}_7$  using ZF and TF  $\mu\text{SR}$ , with the objective of studying the evolution of the magnetic phase with applied field. The preliminary data presented here establishes the  $\mu\text{SR}$  signals *in the low field regime*. Below  $T_N = 115 \text{ K}$ , we find very rapidly damped oscillations over a narrow range of temperatures. Using the temperature dependence of the Mössbauer hyperfine field as a guide [Fig. 4b)], we estimate the average static field at the muon is on the order of 0.46 T at  $T = 0$ . The non-oscillating ZF signal yields a relaxation, likely dynamic in origin, that increases on cooling towards  $T_N$  but then continues to increase below this until eventually slowing at low temperature. A better understanding of the ZF spectra will be aided by neutron data on the long range magnetism[7]. Structure in the TF precession signal in the paramagnetic phase may reflect the subtle charge disproportionation that remains unobserved by diffraction but is clear in the Mössbauer data. It would be interesting to study the precession through  $T_{charge}$  to test this.

#### 5. Acknowledgments

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