Muion spin relaxation measurements on powder $FeTe_2O_5X$, X=Cl, Br samples

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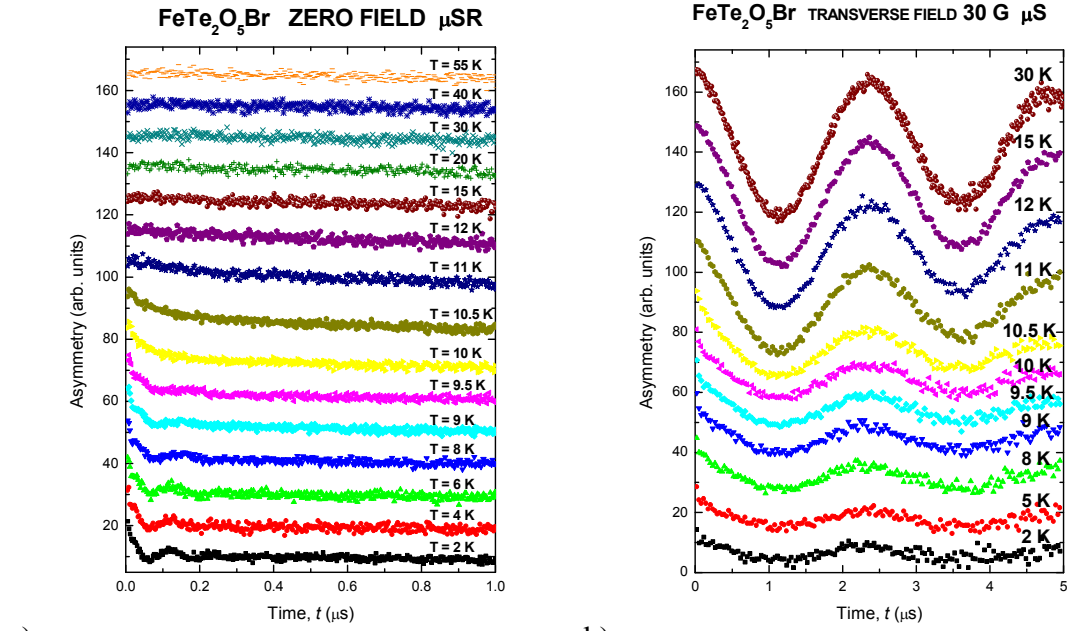
The $FeTe_2O_5X$, X = Cl, Br system grows in a layered structure with monoclinic unit cell, where individual layers are bonded by weak van der Waals forces. Every layer is built of separate $[Fe_4O_{16}]^{20-}$ tetramers, which are held together by $[Te_4O_{10}X_2]^{6-}$ entities. In each individual tetramer two chemically inequivalent Fe^{3+} sites can be distinguished. Fe^{3+}(I) ions are coupled via oxygen bridges to two nearest Fe^{3+}(II) ions. In addition, Fe^{3+}(II) ions are coupled diagonally with each other introducing some geometrical frustration into the tetramer exchange interactions.

Negative Curie-Weiss temperature Θ_{CW}= -124 K (-98 K for Br sample) implies strong antiferromagnetic (AFM) interactions between the (S = 5/2) Fe^{3+} moments. However, strongly suppressed Néel temperature T_N=12.6 K (9.7 K for Br sample) suggests that the magnetic ground state might be indeed frustrated [1]. Moreover, the establishment of AFM long range order is rather complex. In Cl sample a sharp λ-type anomaly at T_N in the specific heat measurements is followed by another anomaly at T'=11 K. On contrary no such behavior is found in Br sample. Our previous single crystal and powder neutron diffraction studies imply possible phase coexistence below T_N. However, from the neutron diffraction experiment alone, we cannot claim with a complete certainty if there exists only one magnetic phase or we really observe phase segregation.

In order to resolve the nature of low temperature magnetic ordering and confirm coexistence of two different magnetic phases below T_N we performed series of muion spin relaxation (muSR) experiments.



a) b)

Fig. 1: a) Zero-field and b) low transfers-field spectra of Br sample

In Fig. 1 we show relaxation spectra obtained for Br sample in the absence and in a small transverse magnetic field. In order to resolve the zero-field spectra we used three component fit, i.e. corresponding to three different muion sites, with three different local magnetic fields:

$$I = \sum_{i=1}^{3} A_i (2/3 \cos(2\pi\nu_i) \exp(-\lambda_{T_i} t) + 1/3 \exp(-\lambda_{L_i} t))$$

The obtained parameters are shown in Fig. 2a,2b. The results imply that below the transition temperature, $T_{Br} \sim 10$ K, first two components correspond to the muion sites, where local magnetic field is well defined, i.e., within magnetically ordered state. On the other hand, the third component most likely correspond to the muion site, where relaxation is very fast, i.e., this site does not feel long range magnetic order. The intensity asymmetry ration imply that volume ratio between the ordered and disordered phase is approximately 2:1. We obtain complementary information from transverse field measurements (Fig. 2c, 2d), which indicate that short range magnetic correlations extend well above the ordering temperature, i.e., up to ~ 20 K.

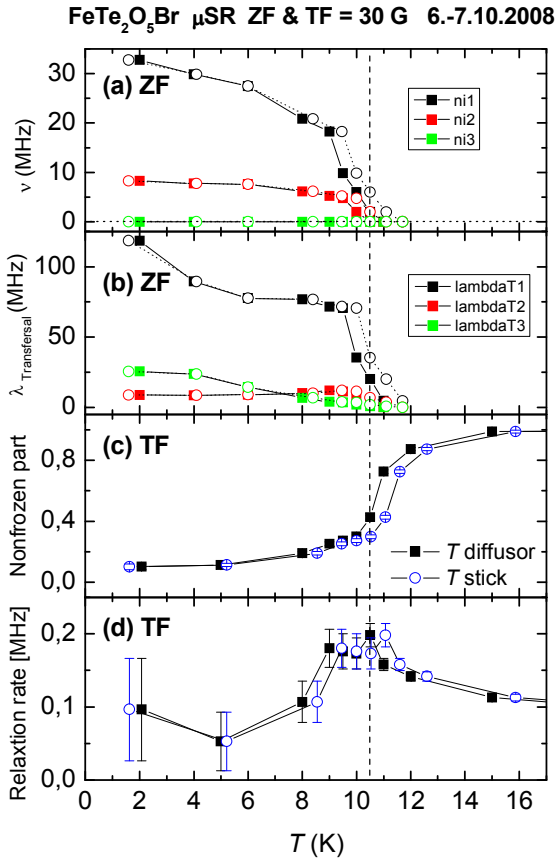
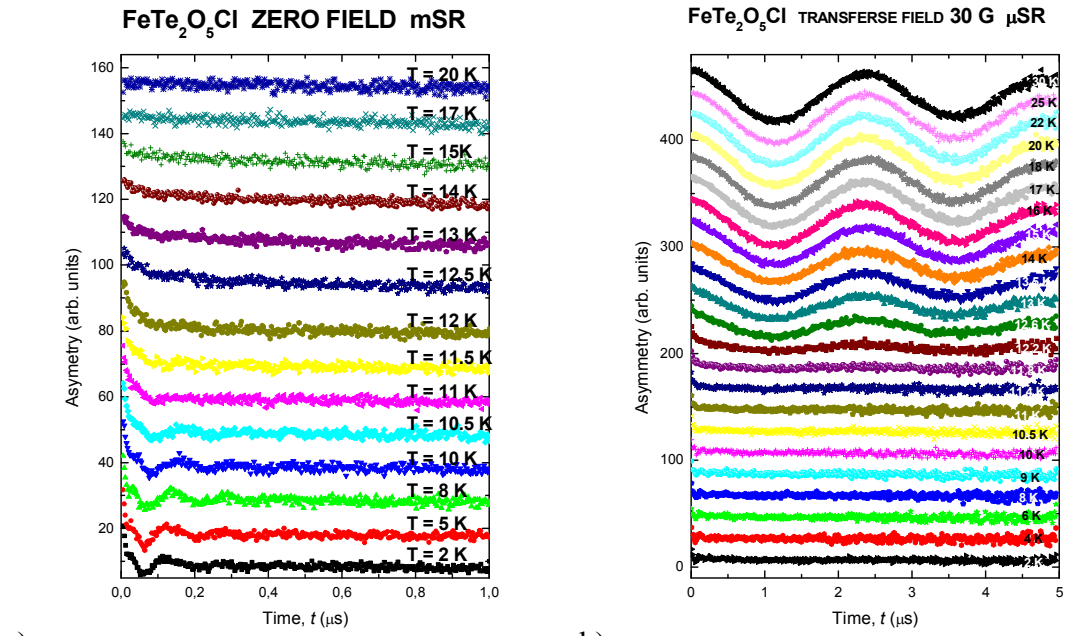


Fig. 2: Analysis of zero-field measurements (a) and (b). (a) relaxation frequencies of three components, (b) transversal relaxation rate for three components. Analysis of low transverse-field measurements: (c) nonmagentic part - nonfrozen part and (d) relaxation rate of the nonfrozen part. Circles correspond to the temperature measured on stick, supporting the sample, while full squares correspond to the temperature measured at He diffuser.

On Cl sample we obtained very similar results Fig. 3 and 4. Again the spectra can be simulated with three components, where two of them correspond to long-range ordering part and the third one, which indicates the presence of short-range ordering even below $T_{Cl} = 11$ K. The volume ratio between the ordered and disordered phase is in this case close to 8:1. However, in addition to well resolved transition at T_{Cl} there seem to be a trace of the second transition at ~ 12.5 K (Fig. 4). This might imply that the first anomaly in specific heat measurements (at 12.6 K) indicate ordering of rather small fraction of the sample (~20 %) and that only below T_{Cl} larger part of the sample undergoes magnetic transition.



a) b)

Fig. 3: a) Zero-field and b) low transfers-field spectra of Cl sample

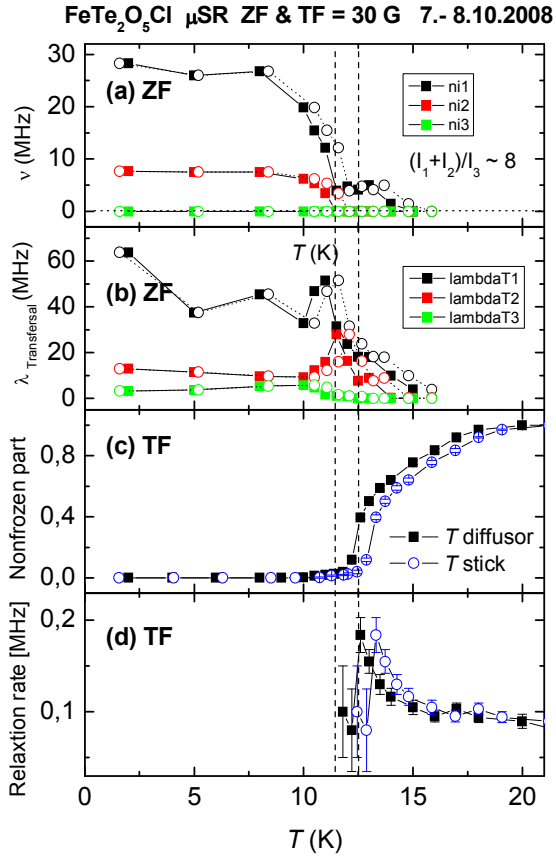


Fig. 4: Analysis of zero-field measurements (a) and (b). (a) muion relaxation frequencies of three components, (b) transversal relaxation rate for three components. Analysis of low transverse-field measurements: (c) nonmagentic part - nonfrozen part and (d) relaxation rate of the nonfrozen part. Circles correspond to the temperature measured on stick, supporting the sample, while full squares correspond to temperature measured at He diffuser.

We believe these measurements contributed a significant part to our understanding of the magnetic behavior of the system. However, in order to determine the exact magnetic structure, we must combine all the knowledge obtained from previous neutron diffraction studies and completely fresh NMR results, which are yet to be analyzed.

After complete analysis of the results, we plan to publish an article in a refereed journal.

[1] R. Becker et al., *J. Am. Chem. Soc*. **128**, 15469 (2006).