Spin freezing and dynamics in the quasi-2D triangular-lattice antiferromagnets TGa_2S_4 (T = Fe, Ni)

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Summary

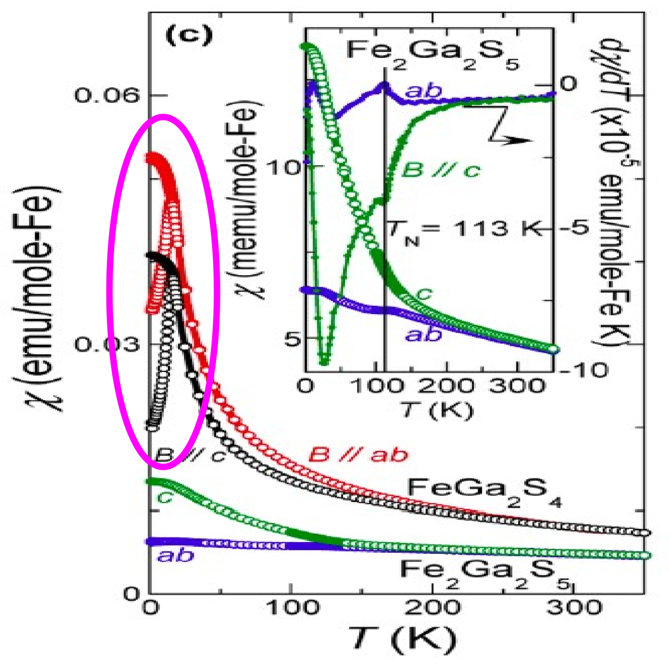
Results of muon spin relaxation (μSR) and dc magnetic susceptibility measurements on the 2D triangular-lattice antiferromagnet $FeGa_2S_4$ are reported and compared with previous data from $NiGa_2S_4$. In $FeGa_2S_4$ μSR data indicate a critical slowing-down of magnetic fluctuations and a transition at $T^* \approx$ 31 K, which is twice the bifurcation temperature $T_f \approx$ 16 K from magnetic susceptibility measurements. As the applied field increases, T_f decreases, consistent with a spin glass-like freezing at this temperature. This could suggest a viscous spin liquid state for $T_f < T < T^*$, as has been attributed to $NiGa_2S_4$ with $T_f \approx$ 3 K from ac susceptibility and $T^* \approx$ 8.5 K from both NMR and μSR. The inhomogeneous dynamic muon spin relaxation rate λ_d scales for both compounds (Fig. 3 below), suggesting a common mechanism for their spin dynamics. Exponential critical slowing down of spin fluctuations [$\lambda_d \propto T^{3/2}\exp(-T_0/T)$], expected in a 2D Heisenberg antiferromagnet, is observed in both compounds. The similar spin dynamics in $NiGa_2S_4$ (S = 1) and $FeGa_2S_4$ (S = 2) suggest that they are essentially classical ($S = \infty$) and may therefore involve Z_2 vortex excitations (Kawamura *et al.*).

Motivation

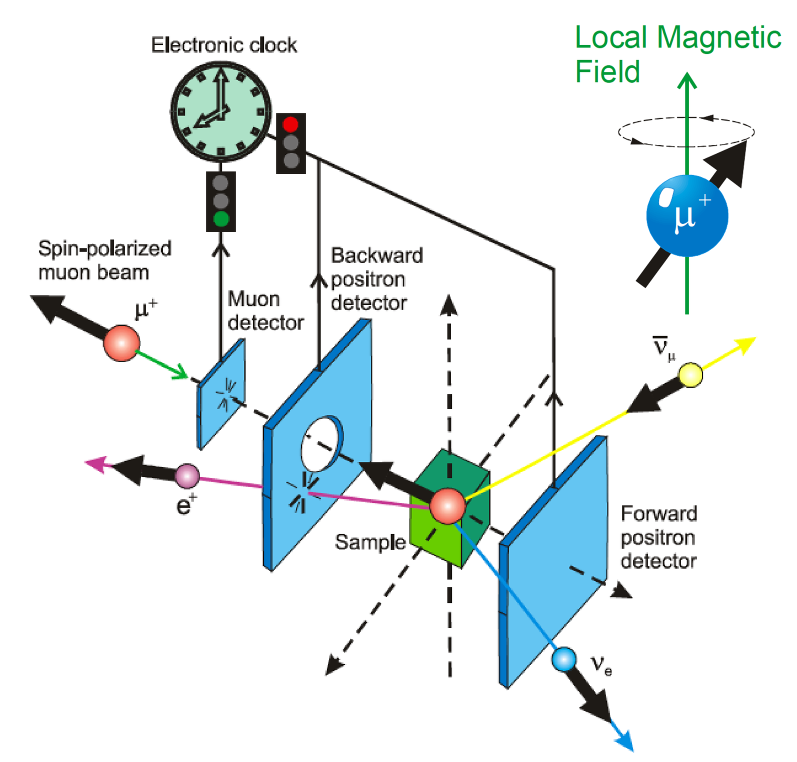
$NiGa_2S_4$ was first studied by Nakatsuji et al. to search for novel ground states in an exact triangular lattice. The dc magnetic susceptibility indicates a transition at $T \approx$ 8.5 K, which is also the temperature where μSR experiments observe critical slowing down and the onset of quasistatic spin freezing.

In the isostructural compound $FeGa_2S_4$, dc magnetic susceptibility measurements exhibit a ZFC-FC bifurcation at $T \approx$ 16 K (figure), which could be a freezing temperature similar to the case of $NiGa_2S_4$. However, recent studies indicate the ZFC-FC bifurcation at 8.5 K in $NiGa_2S_4$ is due to an impurity effect, and spin-glass-like freezing is observed at a lower temperature.

Is 16 K the critical slowing down temperature in $FeGa_2S_4$?



Experimental setup (figure stolen from Jeff & Jess)



Depolarization function:

$$A(t) = A_s \exp[-\Lambda_s t)]\, J_0(\omega_\mu t) + A_d \exp[-(\lambda_d t)^K]$$

Results and Discussion

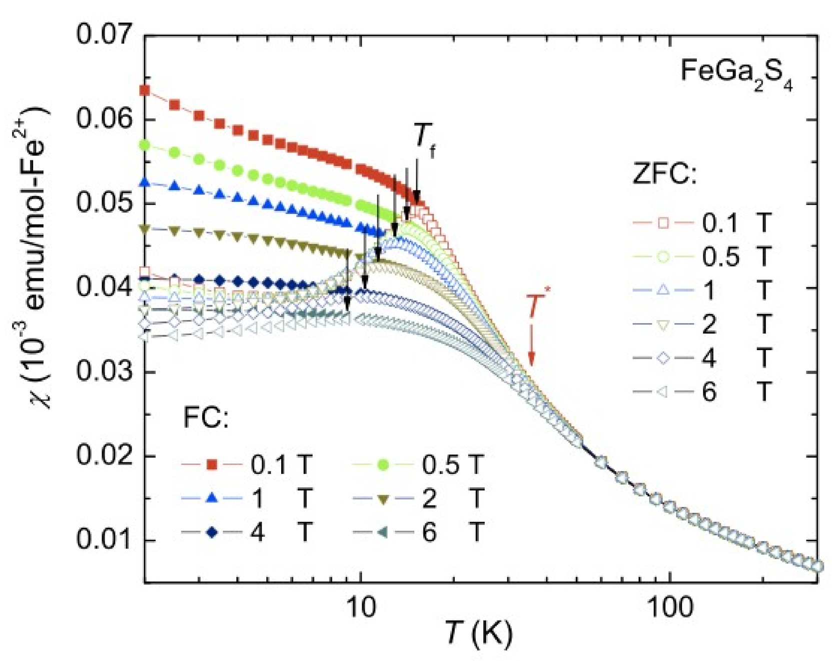


Figure 1: Temperature dependence of magnetic susceptibility $\chi = M/H$ in $FeGa_2S_4$ from 2 K to 300 K for various applied magnetic fields. T_f and T^* are the spin freezing temperature and critical slowing-down temperature, respectively.

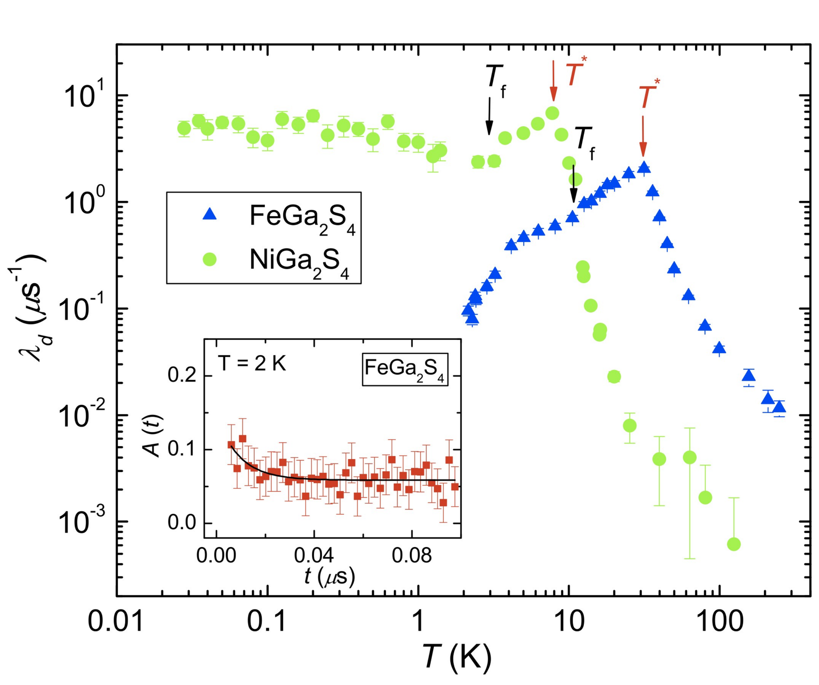


Figure 2: Temperature dependence of dynamic muon spin relaxation rate λ_d in $FeGa_2S_4$ and $NiGa_2S_4$. Inset: representative early-time asymmetry data. The damped oscillation at early times is not visible due to the spectrometer dead time.

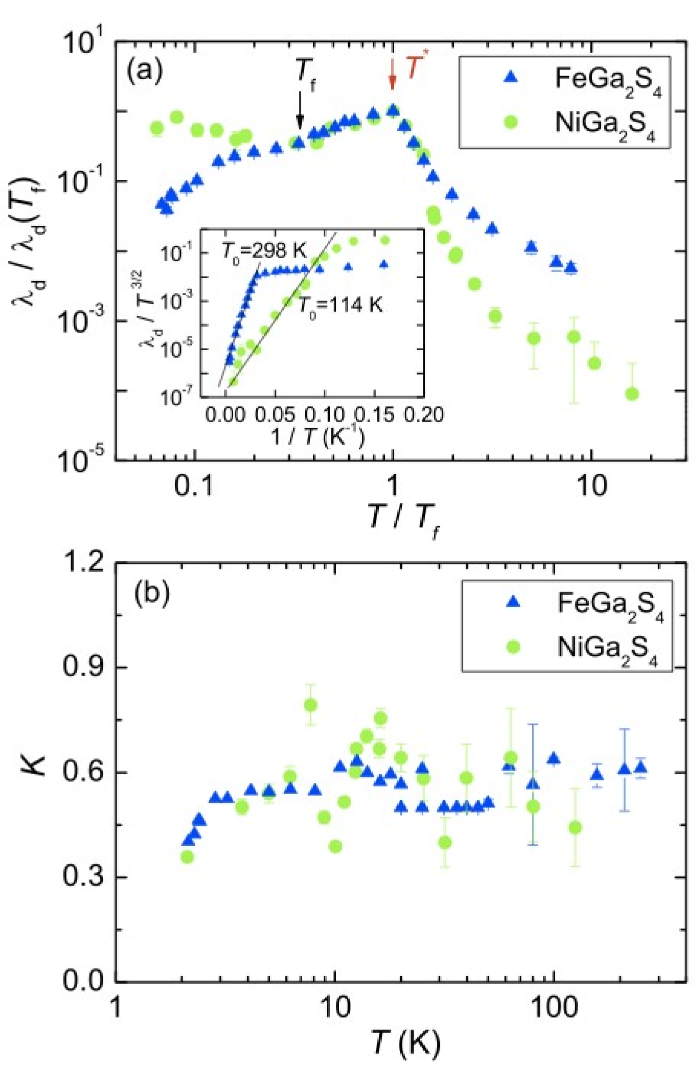


Figure 3: (a) Scaled temperature dependence of dynamic muon spin relaxation rate λ_d in $FeGa_2S_4$ and $NiGa_2S_4$. Inset: 2D critical slowing down of spin fluctuations, leading to $\lambda_d \propto T^{3/2}\exp(-T_0/T)$. (b) Stretched-exponential power K in $FeGa_2S_4$ and $NiGa_2S_4$.

Features:

1. Spin fluctuations above T^* exhibit 2D dynamic critical behavior in both compounds. (inset of Fig. 3a).
2. Comparable extended critical regimes in both compounds: $T_f/T^* \approx$ 0.4 ($NiGa_2S_4$), $\approx$ 0.5 ($FeGa_2S_4$) (Fig. 3a).
3. Similar stretching power K in both compounds (Fig. 3b).

Are Z_2 vortex excitations behind these similar spin dynamics ?

Z_2-vortex scenario:

The antiferromagnetic (AF) Heisenberg model on a 2D triangular lattice is a typical geometrically frustrated magnet. It was demonstrated by Kawamura some time ago that the model bears a topological quantum number, or the so-called Z_2 vortex. It was found that that a triangular 2DHAF exhibits a thermodynamic phase transition at finite temperature with finite spin correlation length and correlation time due to binding and unbinding of Z_2 vortices.

This thermodynamic phase transition is predicted to have an extended critical regime, as has been suggested in $NaCrO_2$, with $T_f/T^* \approx$ 0.3, and in $NiGa_2S_4$, with $T_f/T^* \approx$ 0.4. Here $FeGa_2S_4$ is found to have a critical regime with $T_f/T^* \approx$ 0.5.

A recent study of the classical AF Heisenberg model on a 2D triangular-lattice with bilinear and biquadratic exchange indicates a topological phase transition that is driven by Z_2 vortices for small biquadratic coupling $|Q|$, in which case a Z_2 vortex corresponds to a 2π rotation of the 120° spin structure around a vortex core. The thermodynamic transition temperature varies for different Q values. The observed critical slowing down of magnetic fluctuations in these systems could be a result of such a thermodynamic process.

However, in $NiGa_2S_4$ the quasistatic field below T^* follows a mean-field temperature dependence, suggesting more conventional spin freezing. In $FeGa_2S_4$ this field is much larger, and has not yet been accurately measured.

Conclusions:

Our μSR data indicate similar spin dynamics in both compounds. This and the viscous spin liquid regime or extended critical regime suggest classical Z_2 vortex excitations. In contrast, the weak and conventional low-temperature muon spin relaxation in $Fe_2Ga_2S_4$ ($\lambda_d \approx$ 0.1 μs^{-1} at 2 K and decreasing) suggests that the persistent low-temperature relaxation in $Ni_2Ga_2S_4$ ($\lambda_d \approx$ 5 μs^{-1} at 25 mK) could have a quantum origin.