A μSR magnetic study of frustrated $FeSc_2S_4$ and $MnSc_2S_4$

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Magnetic susceptibility, specific heat [1], X-ray and neutron scattering data [2] showed that the normal spinels $FeSc_2S_4$ and $MnSc_2S_4$ remain cubic to low temperatures. The magnetic ions occupy the A site, its tetrahedral coordination causing strong geometrical magnetic frustration. Thus $MnSc_2S_4$, despite the relative large magnetic moment ($\mu_{eff} = 5.8\mu_B$) of the Mn^{2+} ion, becomes antiferromagnetic only at $T_N = 2\,K$ (frustration parameter $f = 11$). Above T_N the susceptibility follows a Curie–Weiss law ($\theta = 23\,K$). In $FeSc_2S_4$ the Fe^{2+} ion is Jahn–Teller (J–T) active. The small J–T splitting allows rapid spin fluctuations between the CEF states causing in addition orbital frustration and long-range magnetic order is altogether suppressed (frustration parameter on the order 10,000). Around 10 K a small deviation from Curie–Weiss ($\theta = 45\,K$) behavior is seen. $MnSc_2S_4$ and $FeSc_2S_4$ are discussed as a spin-liquid and a spin-orbital-liquid material, respectively [1,2].

μSR spectroscopy in zero (ZF) and longitudinal (LF) field was carried out on powder samples of the two thiospinels, mainly to gain information on the fluctuation rates of the atomic magnetic moments via the temperature dependence of the muon spin relaxation rates. The Mn and Sc ions also possess sizable nuclear magnetic moments. Their influence on muon spin relaxation can be suppressed by application of a weak (5–10 mT) LF. Most of the data have been obtained with specially prepared samples. Control measurements were made on the probes used for neutron scattering. Significant differences were not found.

Down to 125 K we observe in $FeSc_2S_4$ a weakly exponentially relaxing signal $A(t) = A_0 \exp(-\lambda t)$ where $A(t)$ is the measured signal strength (asymmetry) and A_0 is the initial ($t = 0$) asymmetry. This is the typical μSR

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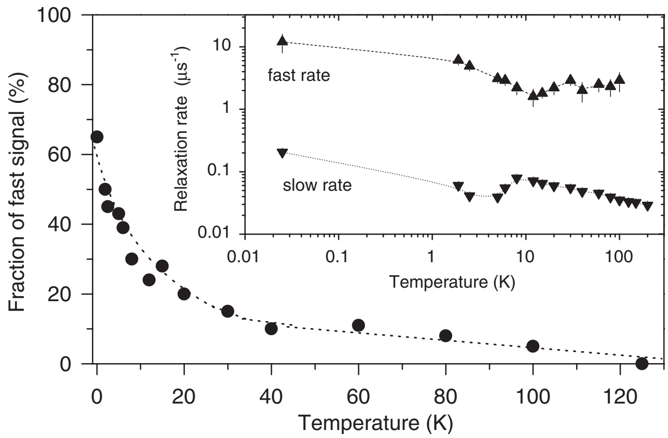

Fig. 1. Volume fraction of the fast relaxing sub-signal of $FeSc_2S_4$ as function of temperature. The inset shows the temperature dependencies of the relaxation rates of the fast and slow sub-signals.

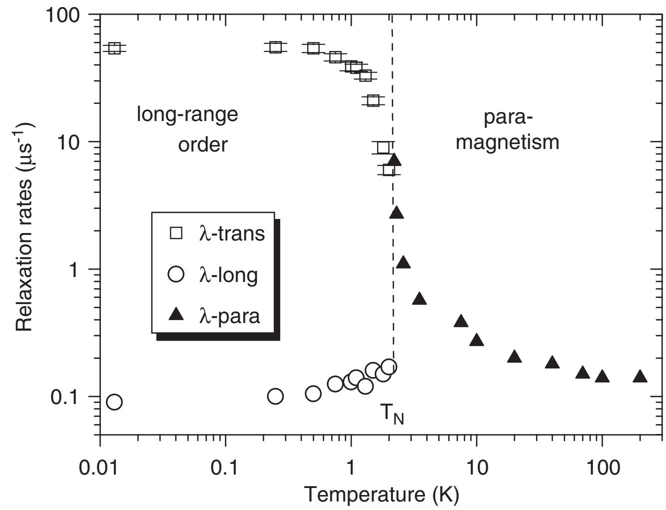

Fig. 2. Temperature dependencies of the transverse, longitudinal (ordered state) and paramagnetic muon spin relaxation rates in $MnSc_2S_4$.

response of a free paramagnet. When cooling further magnetic inhomogeneity develops. For $T \leqslant 100$ K the μSR spectra contain two sub-signals, one fast relaxing, corresponding to a strongly correlated paramagnetic state, the other slowly relaxing, belonging to the weakly correlated, i.e. free paramagnetic fraction. The type of spectral response remains unaltered down to our base temperature of 25 mK, only changes of parameters as shown in Fig. 1 are seen. A transition into a statically ordered magnetic state does not occur. Spin freezing is absent, although spin correlations increase. Measurements in strong LF (≈ 300 mT) reveal a spin fluctuation rate around 1 GHz at 100 K which slows down to $\sim$200 MHz at 2.5 K for the strongly correlated fraction. From the increase (by a factor of 2.5) by going down in temperature to 25 mK one estimates a further reduction in rate to $\sim$80 MHz. The spin system retains persistent spin fluctuation for $T \to 0$, a feature observed in other strongly frustrated magnets. Also the magnetic inhomogeneity persists, a fraction of the system continues to show free paramagnetism. For this fraction spin fluctuations are above 10 GHz at all temperatures. Around 10 K both relaxation rates show an irregularity. This is the temperature range where the deviation from Curie–Weiss behavior is seen. At present we have no good explanation.

In $MnSc_2S_4$ we observe from 250 K down to 2.3 K a single signal with exponential relaxation. The temperature dependence of the relaxation rate (λ_p) is given in Fig. 2. The sharp rise toward 2 K is characteristic for critical slowing down of paramagnetic spin fluctuations on approaching a second-order phase transition. At 2 K spectral shape is no longer exponential. Since the onset of magnetic order at this temperature is known, the spectra were fitted with the μSR response for a long-range ordered powder sample: $A(t) = A_0[2/3\ \exp(-\lambda_t t)\ \cos(\gamma_\mu B_\mu t) + 1/3\ \exp(-\lambda_l t)]$. Here $\gamma_\mu \approx 850\,\mathrm{MHz}/T$ is the muon gyromagnetic ratio and B_μ the internal field at the muon site. The first term describes the action of internal fields perpendicular to the muon spin, causing spin precession. The transverse relaxation rate λ_t (which damps the oscillatory pattern) is given mostly by the static distribution of B_μ due to differences in the arrangements of spins around the muon. The second term represents the cases where B_μ is oriented parallel to the muon spin. Hence this term does not describe a precession pattern, just exponential relaxation whose rate is directly proportional to the rate of fluctuations of B_μ coming from fluctuations of the atomic magnetic moments. In $MnSc_2S_4$ we found $\lambda_t > \gamma_\mu B_\mu$, i.e. an oscillatory pattern cannot develop. The width of the distribution of B_μ is larger than the mean magnitude of B_μ for which the inequality gives $B_\mu < 60$ mT. The large value of the field distribution width means that, although long-range order exists, the spins are considerably disordered on a local scale of a few lattice constants. No change of spectral type occurs between T_N and 13 mK (our base temperature), i.e. no second phase transition occurs. LF data exclude the coexistence of a spin-glass-like state. The temperature dependencies of the two rates are plotted in Fig. 2. The transverse rate exhibits Brillouin-like behavior reflecting the increase of the effective moment for $T \to 0$ expected in a normal ordered magnet. The longitudinal rate shows the move toward the static limit for the magnetic spins, yet the fully static situation is not reached.

References

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