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Evidence for an exotic magnetic transition in the triangular spin system $FeGa_2S_4$

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We report positive muon spin relaxation measurements on the triangular lattice magnetic system $FeGa_2S_4$. A magnetic transition that, to the best of our knowledge, has not been previously detected by specific-heat and magnetic susceptibility measurements is found in zero field at $T^* \simeq 30$ K. It is observed through the temperature dependencies of the signal amplitude and the spin-lattice relaxation rate. This transition is therefore not a conventional magnetic phase transition. Since persistent spin dynamics is observed down to 0.1 K, the ground state cannot be of the canonical spin-glass type, which could be suggested from hysteresis effects in the bulk susceptibility below $T_f \simeq 16$ K. These results are compared to those found for the isomorph $NiGa_2S_4$. It is argued that the fate of the transition, which has been interpreted in terms of the Z_2 topological transition in this latter system, is probably different in $FeGa_2S_4$.

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The search for new states of matter is at the forefront of condensed-matter research, and geometrically frustrated magnetic systems provide a fruitful playground.[1–3] One of the main goals is to find and characterize magnetic systems for which long-range magnetic order is absent at low temperature despite strong exchange interactions. Two-dimensional Heisenberg antiferromagnets are good candidates since magnetic order can only occur at temperature $T = 0$. In the case of the equilateral triangular lattice, it is now believed that the system would exhibit 120° spin order.[4–6] However, symmetry-breaking energy terms and exchange interactions between the layers or inside the layers beyond the nearest-neighbor may lead to magnetic order at finite temperature or quantum spin-disordered ground states.[7] Only a few two-dimensional triangular-lattice systems are believed not to display magnetic long-range order: two organic and two inorganic compounds.[8] While recent interesting results have been reported,[9] the restricted amount of organic material that can be produced imposes serious limitations on the measurements that can be performed, precluding, e.g., inelastic neutron scattering experiments. This is not the case for the inorganic transition metal sulfides $NiGa_2S_4$ and $FeGa_2S_4$, which are available in large quantities. These two insulators, which crystallize in the space group $P\bar{3}m1$, consist of magnetic Ni^{2+} and Fe^{2+} layers, respectively, characterized by strong intralayer magnetic bonds in comparison to very weak interlayer couplings. The Ni and Fe ion sites form an equilateral triangular lattice in each layer.

While $NiGa_2S_4$ has been studied with a wide range of macroscopic and microscopic experimental techniques,[10–17] fewer studies including x-ray diffraction, magnetic susceptibility, resistivity, and specific-heat measurements have been reported for $FeGa_2S_4$.[18,19] The available data suggest that the two magnetic systems are similar. They have comparable lattice parameters. Their Curie-Weiss temperatures θ_{CW} are quite large, 80 and 160 K for $NiGa_2S_4$ and $FeGa_2S_4$, respectively. Upon cooling below 100 K, the dc susceptibility χ_{dc} of the two compounds develops weak easy-plane anisotropy (which is larger for $FeGa_2S_4$) but no in-plane anisotropy. $\chi_{dc}(T)$ shows a bifurcation between zero-field-cooled (ZFC) and field-cooled (FC) data at 8.5–9 and 16 K (i.e., $\simeq\theta_{CW}/10$) for $NiGa_2S_4$ and $FeGa_2S_4$, respectively. The magnetic contributions to the specific heat $C_M(T)$ of the two compounds are field insensitive with a T^2 dependence at low temperature, and they match one another after appropriate rescaling.[20] While C_M/T is negligible for $T \to 0$ K in $NiGa_2S_4$, an appreciable value is measured for $FeGa_2S_4$. Finally C_M/T exhibits an unusual double-peak structure: a first rounded maximum at ≈10 K for the two systems, and a second broad maximum centered around 100 and 60 K for $NiGa_2S_4$ and $FeGa_2S_4$, respectively.

The microscopic magnetic properties of $NiGa_2S_4$ are experimentally well established. Neutron scattering experiments[10] show that $NiGa_2S_4$ displays incommensurate quasistatic short-range magnetic correlations. These correlations manifest themselves in a spontaneous field below $T^* \simeq 9$ K in muon spin rotation and relaxation (μSR) experiments.[14,15] Concerning temperatures above T^*, while the longitudinal-field μSR relaxation function is exponential at high temperature, it becomes subexponential or "stretched exponential" for $T \to T^*$ and below. This shape indicates the presence of spatial inhomogeneity in the relaxation rate. On the premise that topological Z_2 vortices manifest themselves in the vicinity of the temperature where $C_M(T)$ displays a broad maximum,[21] the transition at T^* was tentatively associated with the dissociation of these vortices.[14] In addition, the stretched exponential relaxation just above T^* was interpreted as the signature of vortex unbinding, which is expected to give rise to magnetic disorder.

We are aware of only one investigation of $FeGa_2S_4$ by a microscopic technique. From ^{57}Fe Mössbauer spectroscopy, a spontaneous hyperfine field has been observed at low temperature with an order-parameter-like temperature dependence.[22,23] An antiferromagnetic transition has been inferred from these data with a Néel temperature of 33 K. In this Rapid Communication we present μSR measurements which

are aimed at further characterizing the nature of the transition. We also find a magnetic transition with $T^* \simeq 30$ K $\simeq \theta_{CW}/5$ rather than 9 K $\simeq \theta_{CW}/10$ in the nickel counterpart. Persistent spin dynamics are observed in both compounds down to the lowest temperature of the measurements, i.e., 0.1 K or less.[24] Nevertheless, the two systems differ sharply in some respects. In contrast to $NiGa_2S_4$ no spontaneous field is detected in $FeGa_2S_4$, owing to a broader field distribution. In the latter compound no anomaly is observed in $C_M(T)/T$ or $\chi_{dc}(T)$ at T^*, whereas in $NiGa_2S_4$ a weak cusp is seen in $\chi_{dc}(T)$ at low fields. In addition, none of the temperatures of the two C_M maxima in the two systems scales with T^*. The observation of a magnetic transition in $FeGa_2S_4$ while no signature for it is detected in specific-heat and susceptibility data points to its exotic nature.

Data were obtained from μSR measurements carried out at the General Purpose Surface-Muon (GPS) ($2.8 \leqslant T \leqslant 160$ K) and the Low Temperature Facility (LTF) ($0.10 \leqslant T \leqslant 3.5$ K) instruments of the Swiss Muon Source (SμS), Paul Scherrer Institute (PSI), Villigen, Switzerland. Most of the spectra were recorded in zero field, with additional spectra taken with an applied external longitudinal field. Measurements were mostly performed on a powder sample prepared in Tokyo. A few spectra were also recorded on a powder prepared in Grenoble, giving results consistent with a transition at T^*.

μSR techniques and their application to the study of magnetic materials are described elsewhere.[25–27] A time-differential μSR asymmetry spectrum is given by $a_0\eta P_Z^{exp}(t)$, where a_0 is the spectrometer-dependent initial asymmetry, $P_Z^{exp}(t)$ is the muon spin polarization function [$P_Z^{exp}(t=0) = 1$],[27] and η ($0 \leqslant \eta \leqslant 1$) characterizes the so-called missing asymmetry due to rapid relaxation within the spectrometer dead time t_{dt} ($\approx$5 ns at PSI). This is typically caused by a broad distribution of static fields arising from frozen magnetism. In this case the remaining asymmetry arises from the muon spin component parallel to the static field, and its relaxation is due solely to dynamic processes (spin-lattice relaxation).[27] In the following $a_0\eta$ will be called the effective initial asymmetry and $a_0\eta P_Z^{exp}(t)$ the asymmetry.

Exploratory measurements at the GPS spectrometer indicated that the field history may have to be taken into account for $T < 60$ K. Hence, we first report zero-field measurements, for which no field was present when cooling down the sample below 60 K. In contrast to $NiGa_2S_4$, no early-time oscillation could be reliably resolved in the zero-field spectra at any temperature down to 0.1 K. Thus we have not detected any spontaneous mean field at the muon site. This and the reduced low-temperature value of η, discussed below, indicate a broad distribution of fields. A lower bound for the width of the distribution at low temperature is of order $\Delta B \approx 1/(\gamma_\mu t_{dt}) \simeq 0.2$ T ($\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1} is the muon gyromagnetic ratio). All the spectra can reasonably be described, except at short times ($t \lesssim 50$ ns) near T^* where the static fields become smaller, by the stretched exponential relaxation function

$$P_Z^{exp}(t) = \exp[-(\lambda_Z t)^\beta], \quad (1)$$

where λ_Z is the spin-lattice relaxation rate and β with $0 < \beta \leqslant 1$ is the stretching power. Two typical zero-field spectra are shown in Fig. 1. We recall that a stretched exponential relaxation function accounts for a continuous distribution of relaxation rates. But as shown for $La_{1-x}Ca_xMnO_3$ this may not be justified,[28] and a multisite model might be more appropriate. Such a description would require so many parameters (two parameters per site, i.e., a weight and an exponential relaxation rate) that the physics would be obscured, and we shall not attempt it.

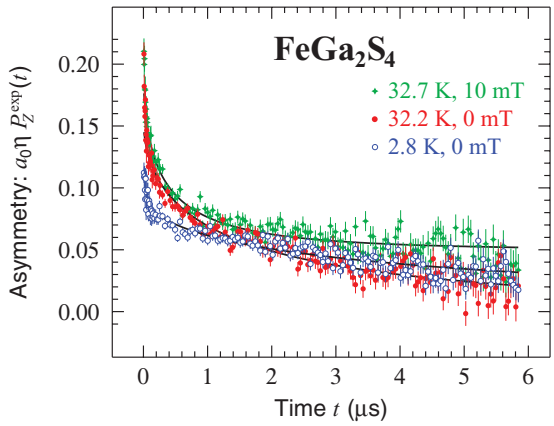

FIG. 1. (Color online) Examples of zero- and longitudinal-field μSR spectra recorded on a powder sample of $FeGa_2S_4$. Solid curves: Results of fits as explained in the text. The reduced signal amplitude, i.e., the reduced effective initial asymmetry, for the 2.8 K spectrum is clearly seen graphically. Note that these data result from the weighted subtraction of counts recorded in two opposite detectors, implying that their shape and amplitude are not dependent of any fit parameter.

Focusing first on the spectra recorded at the GPS, and assuming β to be a free parameter, we find $0.4 \lesssim \beta \lesssim 0.6$ for $2.8 \leqslant T \leqslant 25$ K and $0.3 < \beta < 0.4$ for $25 < T \leqslant 35$ K. The value above 40 K increases smoothly to reach $\beta = 1$ at $\approx$60 K. The final fits were carried out setting $\beta = 1/2$ somewhat arbitrarily for $T \leqslant 40$ K and leaving it free above that temperature. Indeed, when β approaches 0.3, $a_0\eta$ becomes unrealistically large. This points out the inadequacy of the fitting function for $t \lesssim 50$ ns. In contrast, setting $\beta = 1/2$ yields a value for $a_0\eta$ that fairly reflects the observed signal at short times. As shown in Fig. 1 the resulting description of the spectra is reasonable.

It is more difficult to analyze the spectra recorded at the LTF than at the GPS because a fraction of the muon beam stops in the silver cold finger of the dilution refrigerator. This fraction is traditionally determined from a calibration measurement performed above T^*.[27] Here this measurement cannot be done since T^* is higher than the maximum temperature available at the LTF. To circumvent this difficulty spectra with overlapping temperatures were recorded at both the GPS and LTF; the λ_Z values extracted from the GPS spectra were used to analyze the LTF data, with the effective initial asymmetry being the only free physical parameter in the fitting procedure. With this method a reliable $a_0\eta$ value was obtained from the LTF spectra. For temperatures at which only LTF spectra are available, this value was fixed during the fits. In addition, following the GPS results, $\beta = 1/2$ was assumed down to 0.1 K, leaving λ_Z as the only free physical parameter.

Figure 2 gives the temperature dependencies of $a_0\eta$, λ_Z, and β. We note the following three points. (i) The strong

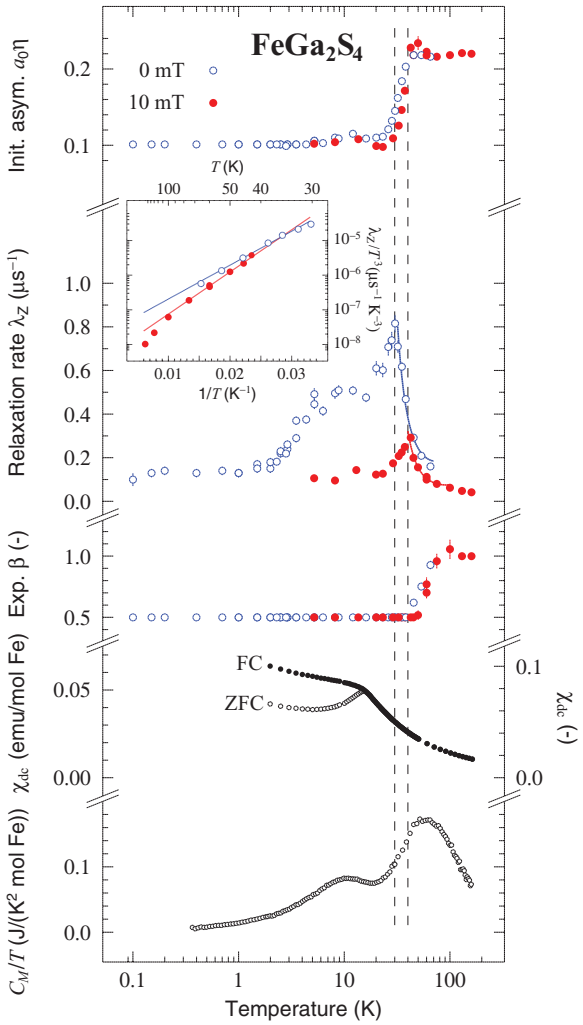

FIG. 2. (Color online) The three upper panels show the temperature dependencies of the effective initial asymmetry $a_0\eta$, the spin-lattice relaxation rate λ_Z, and the stretched-exponential power β measured in a powder sample of $FeGa_2S_4$ in zero or 10 mT longitudinal fields. The solid curves for $\lambda_Z(T)$ above T^* result from a fit to a model explained in the main text. The adequacy of the model can be judged in the inset, which displays λ_Z/T^3 vs $1/T$. The temperature dependencies of the susceptibility χ_{dc} given in cgs or *Système International* (SI) units and the ratio C_M/T of the magnetic specific heat over the temperature (Ref. 18) are shown in the lower two panels. The susceptibility has been measured in an external field of 0.1 T after a ZFC or FC protocol. For reference dashed lines are plotted at 30 and 40 K.

anomalies in $a_0\eta(T)$ and $\lambda_Z(T)$ at $T^* \simeq 30$ K point to a magnetic transition at T^*, independent of the details of the fitting procedure. This is evidence, after ^{57}Fe Mössbauer spectroscopy,[22] of such a transition in $FeGa_2S_4$. Its width is relatively large since the decrease of $a_0\eta$ with cooling begins at 40 K and ends only at ≈25 K. The increase of λ_Z by a factor 5 on cooling from 65 K to T^* is due to the slowing down of magnetic fluctuations often observed on cooling toward a magnetic transition. The drop of $a_0\eta$ is an indirect but perfectly clear indication of the onset of a spontaneous field at the muon site for a magnetic transition in a powder sample. Remarkably, there are anomalies neither in $a_0\eta(T)$ nor in $\lambda_Z(T)$ around $T_f = 16$ K, where the ZFC bulk susceptibility is maximum and shows a bifurcation with the FC susceptibility; see Fig. 2. Conversely there is no anomaly in the susceptibility at T^*. Furthermore, the temperature T^* does not correspond to any of the broad maxima observed in C_M/T. For $T \ll T^*$, $a_0\eta$ is ≈45% of the value above the transition, relatively close to the value 1/3 that would be expected for a powder sample with randomly oriented local fields. As discussed previously, the early-time signal relaxes rapidly and is lost in the spectrometer dead time. (ii) The spin-lattice relaxation channel is modeled by a stretched exponential function; the exponential relaxation expected in a homogeneous sample is only observed deep in the paramagnetic phase. This suggests that the compound is magnetically inhomogeneous: There is a distribution of spin-lattice relaxation rates.[27,29–31] (iii) Since an appreciable monotonic decay of the asymmetry is observed even at extremely low temperature, persistent spin dynamics is present. In fact, $\lambda_Z(T)$ displays a plateau below ≈2 K down to the lowest temperature (0.1 K). This behavior seems to be a characteristic of frustrated magnetic compounds either with[32–34] or without[35,36] magnetic order. Interestingly, the presence of fluctuating iron moments was also deduced from the Mössbauer spectroscopy study of $FeGa_2S_4$.[23]

Quantitatively, we can fit the prediction of a model which treats nuclear or muon spin relaxation due to critical fluctuations of a two-dimensional (2D) quantum frustrated Heisenberg antiferromagnet,[37–39] to $\lambda_Z(T)$ in the paramagnetic state. This model predicts for $T \ll T_0/2$

$$\lambda_Z \propto T^3 \exp(T_0/T), \tag{2}$$

where T_0 is proportional to the spin-stiffness constant which is related to exchange. Figure 2 shows the best fit to the data. We find $T_0 = 225$ (20) and 280 (20) K for the zero and 10-mT longitudinal field data, respectively. The exchange constant J can be estimated from the relations $k_B T_0 = 4\pi\rho_s$ (Ref. 38) and $\rho_s = [1 - 0.399/(2S)]JS^2/\sqrt{3}$ for $S = 2$:[37] We find $J/k_B =$ 8.6 (8) and 10.7 (8) K for the two values of T_0, respectively. They compare favorably with the value 13.3 (7) K derived from the Curie-Weiss temperature.

We have also recorded spectra after field cooling the sample in a $B_{ext} = 10$ mT longitudinal field; see Fig. 1. It appears that the application of the field strongly affects the spectral shape at short time. This can be interpreted as a signature of a change of the static or quasistatic field distribution at the muon site. At longer times the spectra reflect only the muon relaxation associated with the dynamics of the local field. Therefore, to gain an insight into these dynamics we fitted the experimental data after truncation of the early time part, using Eq. (1). Cutoff times ranging between 100 and 300 ns were tested and the fitting parameters were found essentially independent of this cutoff. The results are shown in Fig. 2. The field drastically suppresses λ_Z, and the temperature dependencies of the three parameters in the vicinity of T^* are somewhat shifted to higher temperature than in zero field.

The value of B_{ext} is much less than the width ΔB of the local field distribution to which the muons are submitted at low temperature. Therefore we would not expect the spectral shape at long time to be affected by B_{ext}. This field effect is reminiscent of $NiGa_2S_4$,[15] where the internal field is much less

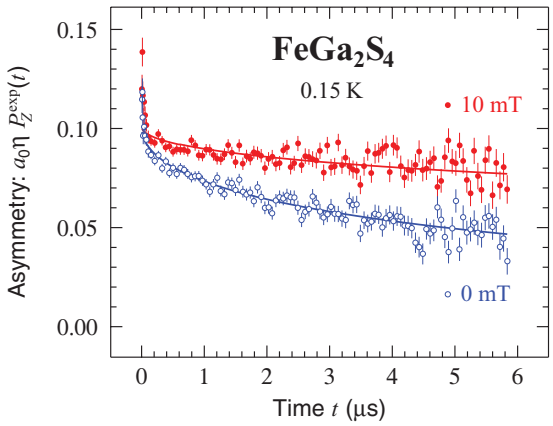

FIG. 3. (Color online) Comparison of spectra measured at 0.15 K for $FeGa_2S_4$ after zero-field cooling. The first spectrum is measured in zero field and the second after the subsequent application of a 10 mT longitudinal field.

distributed than in the current system, but still larger than 0.2 T at low temperature.

Additional measurements of the field response of $FeGa_2S_4$ have been performed at very low temperature. The 10 mT spectrum measured after a ZFC protocol and displayed in Fig. 3 again shows an important difference with the zero-field spectrum. Therefore the drastic effect of a modest field does not depend on the temperature of the field application and it is still present at 0.15 K. This is in contrast to the field insensitivity of the specific heat for a field as large as 7 T.[18]

The rapid increase of λ_Z as the sample is cooled down in zero field toward T^* is a key result of our study. In a conventional magnet it would indicate a magnetic phase transition; see, for example, Ref. 40. However, as no anomaly is observed in the magnetic specific heat and susceptibility at that temperature,[8,18] the detected anomaly in λ_Z does not correspond to a conventional phase transition. Note that a rapid increase of λ_Z as the sample is cooled down toward 9 K was also found for $NiGa_2S_4$.[14,15] The transition at T^* is not spin-glass-like, since persistent spin dynamics are found even at temperatures as low as 0.1 K.

As indicated earlier, the $NiGa_2S_4$ transition at T^* could be associated with a Z_2 topological transition. In the case of $FeGa_2S_4$ the transition which has been revealed in the present study occurs at a temperature much higher than those of the first broad peak in the magnetic specific heat ($\approx$10 K) and of the bifurcation in the ZFC susceptibility ($\approx$16 K). Thus it seems unlikely that the transitions in $FeGa_2S_4$ and $NiGa_2S_4$ are of the same nature. This conclusion is further supported by the fact that hysteresis effects for the latter compound are much smaller and occur at T^*.

In conclusion, our μSR experiments on $FeGa_2S_4$ confirm the presence of the magnetic transition at $T^* \simeq 30$ K in zero field which was previously observed by ^{57}Fe Mössbauer spectroscopy. With the data available we are unable to estimate the magnitude of the magnetic moment involved. Because of the lack of anomalies at T^* in the specific heat and in the susceptibility and because of a strong field effect, this transition is exotic. In addition, as usual for geometrically frustrated magnetic compounds and in particular for $NiGa_2S_4$, persistent spin dynamics are observed at low temperature.

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