# Ground state of the geometrically frustrated system Y(Sc)$Mn_2$ studied by muon spin relaxation

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We performed positive muon spin relaxation measurements of the geometrically frustrated system $Y_{0.97}Sc_{0.03}(Mn_{1-x}Al_x)_2$. A spin freezing with a static magnetic field of 0.05 T was found at 2.5 K in the $x=0$ compound while the $x=0.1$ compound freezes at 45 K with the internal field of 0.17 T. In the $x=0.02$ compound, the relaxation rate shows a marked thermal hysteresis; the maximum rates occur at 10 K on cooling and at 20 K on heating, whereas the sudden changes in the initial asymmetries occur at 40 K in both processes. The results for the $x=0$ compound are interpreted as a freezing of minority paramagnetic Mn moments induced by imperfections such as Sc and/or muon charge while majority Mn moments form singlet tetrahedra.

## I. INTRODUCTION

The ground state of highly frustrated spin systems has been controversial since Wannier[1] demonstrated macroscopic degeneracy in the ground state of the triangular lattice antiferromagnet. Anderson[2] proposed an attractive idea that a quantum effect produces resonating valence bonds resulting in a spin liquid state. Much theoretical work has been devoted to verify these ideas, especially in relation to strongly correlated electron systems such as oxide superconductors. Experimental trials to reveal the ground state of the triangular lattice antiferromagnet have been blocked by phase transitions into three-dimensional ordering due to subsidiary interactions such as weak interplane coupling.

Besides two-dimensional frustrated systems such as the triangular lattice antiferromagnet, there exist a number of three-dimensional geometrically frustrated systems composed of antiferromagnetic tetrahedra.[3] In these systems tetrahedra made of four triangles extend three dimensionally by sharing corners or edges. The 16*c* sites in the spinel structure, the pyrochlore structure, and the Laves phase structure are examples of lattices composed of corner-shearing tetrahedra, while the fcc lattice consists of edge-sharing tetrahedra. Compared to triangular lattices, there have been much fewer studies of the strongly frustrated tetrahedral systems.[4]

Most studies of spin frustration have been confined to insulators. The corner-sharing tetrahedral systems include a number of intermetallic compounds with electrical conductivity. The magnetic interaction via conduction electrons extends three dimensionally to greater distance than the nearest-neighbor spacing so that the situation is rather complicated in low dimensional lattices. It is likely that conduction electrons give less difficulty in the case of three-dimensional frustrated systems. In contrast to antiferromagnetic insulators, itinerant electron systems have an extra degree of freedom to release the spin frustration by reducing the magnitude of the magnetic moments in addition to the angular orientation of moments.[5]

$YMn_2$ is an intermetallic compound with the Laves phase ($C15$) structure, where Mn atoms form a corner-sharing tetrahedral lattice. At 100 K, the itinerant electron character of paramagnetic $YMn_2$ changes into a localized spin character with magnetic moments of $2.7\mu_B$ in the first-order transition. Below the temperature a helical magnetic ordering appears[6] accompanying a tetragonal distortion and a 5% swelling of the lattice.[7] The instability of magnetic moments in $YMn_2$ results from strong frustration in the itinerant *d*-electron system close to a magnetic-nonmagnetic transition. A Hubbard Hamiltonian deduced from the Blume-Capel model could provide an insight into the stability of the magnetic moments in itinerant antiferromagnets $RMn_2$.[5] This model demonstrated successfully the existence of the mixed phase with nonmagnetic sites in addition to magnetic and nonmagnetic phases.

The magnetic ordering is suppressed by applying a relatively weak hydrostatic pressure.[8] Alternatively, the 3% substitution by smaller Sc atoms for Y atoms also suppresses the structural and magnetic transitions due to chemical compression.[9] $Y_{0.97}Sc_{0.03}Mn_2$ is an itinerant electron system with a high specific-heat $\gamma$ value in excess of 140

 

mJ/K$^2$ mol.[10] The magnetic susceptibility of $Y_{0.97}Sc_{0.03}Mn_2$ is almost temperature independent above 100 K but increases divergently with decreasing temperature at low temperatures.[11] The Curie-Weiss fraction of the magnetic susceptibility yields $p \sim 0.2\mu_B$ and $\theta \sim 7$ K so that it may be attributable to minority magnetic moments induced by some imperfections. No magnetic ordering has been detected by NMR down to 0.07 K.[12]

By paramagnetic scattering of polarized neutrons, a marked spin correlation is observed even at 300 K at $Q = 1.5\,\text{Å}^{-1}$ corresponding to twice the nearest Mn spacing which indicates a strong antiferromagnetic spin correlation between neighboring Mn moments.[13] The observed spin correlation is not relevant to antiferromagnetic short-range order of localized magnetic moments but soft mode of spin fluctuations. The scattering intensity decreases as temperature is lowered; the amplitude of the antiferromagnetically correlated spins was evaluated to be $1.3\mu_B$ at 8 K. Our recent measurements did not detect any change in the neutron-scattering spectrum down to 1.35 K. Inelastic neutron-scattering experiments on a single crystal of $Y_{0.97}Sc_{0.03}Mn_2$ revealed that the spin-fluctuation spectrum is strongly anisotropic near (1.25 1.25 0) but has no magnetic scattering at any reciprocal lattice points at 10 K.[14] The results are interpreted in terms of short-lived four-site collective spin singlet tetrahedron.

These remarkable behaviors were interpreted in terms of quantum spin liquid[15] but Lacroix and co-workers[16] attributed them to the giant spin fluctuations characteristic to the heavy fermion system close to magnetic-nonmagnetic transition induced by strong geometrical frustration.

The partial substitution of nonmagnetic Al atoms for Mn atoms gives rise to a remarkable change in magnetism. For example, $Y_{0.97}Sc_{0.03}(Mn_{0.9}Al_{0.1})_2$ has a much larger magnetic susceptibility and shows a field cooling irreversibility below 40 K suggestive of spin glass.[15] An anomalous volume expansion below 150 K implies the growth of the localized magnetic moments below that temperature. In contrast to the case of $Y_{0.97}Sc_{0.03}Mn_2$, the neutron-scattering amplitude of zero energy transfer at $Q = 1.6\,\text{Å}^{-1}$ grows markedly as temperature is reduced to 10 K due to spin-glass freezing.[17] Whether the Al substitution in $Y_{0.97}Sc_{0.03}Mn_2$ changes the electronic state gradually without phase boundary or not, has not been elucidated. In any case, the Al-substituted compound is an excellent reference system for the study of frustration in $Y_{0.97}Sc_{0.03}Mn_2$.

Here we report the results of muon spin relaxation ($\mu$SR) measurements which were carried out to reveal the ground state of the strongly frustrated system $Y_{0.97}Sc_{0.03}Mn_2$ by comparing with the Al substituted compounds. We expected the observation of the muon relaxation due to the soft modes of the zero-point spin fluctuations at the lowest temperature. $YMn_2$ and its related compounds have been studied by $\mu$SR technique so far but the measurements are limited to a rather high-temperature region.[18,19] A part of the present study has been already published[20] which are in good agreement with the results above 3.5 K reported by Kalvius and co-workers.[19]

## II. EXPERIMENTAL PROCEDURE

Polycrystalline specimens of $Y_{0.97}Sc_{0.03}(Mn_{1-x}Al_x)_2$ with $x=0$, $x=0.02$, and $x=0.1$ were prepared from their constituent metals with 99.9% purity by arc melting in an argon atmosphere followed by annealing at 800 °C for one week. Only fresh specimens were used for the measurements because they get easily spoiled during long storage. The quality of the prepared specimens was checked by means of x-ray diffraction and magnetic susceptibility measurements.

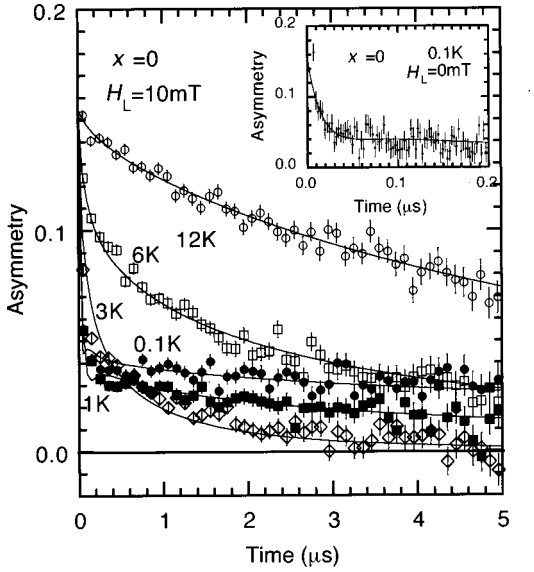


FIG. 1. Typical time spectra for the $x=0$ compound. Solid curves are the best fit to the relaxation functions in the text. The inset is the prompt part of the zero-field spectrum taken at 0.1 K.

The $\mu^+$SR measurements were performed with the continuous $\mu^+$ beam at the M13, M15, and M20 channels of TRIUMF down to 0.1 K using a gas-flow-type cryostat and a dilution refrigerator. A superconducting magnet produced longitudinal magnetic fields at the specimen up to 2 T.

## III. RESULTS

$\mu^+$SR spectra were measured below 150 K where $\mu^+$ diffusion is not appreciable. The zero-field spectra at high temperatures give the nuclear dipolar field of 0.2 mT. From the magnitude of the nuclear dipolar field computer simulation suggests the muon site to be at the 16*c* sites close to the reported site for $CeAl_2$.[21] All asymmetries of the decayed positrons observed in zero field and longitudinal fields show monotonic decreases with time and no precession spectrum was observed in the measured temperature range.

Figure 1 shows some typical time spectra for the $x=0$ compound. For the analysis of the time spectra at high temperatures, the powered exponential relaxation function

$$A(t) = A_0 \exp\{-(\lambda t)^{\beta}\}, \tag{1}$$

was applied where $\lambda$ is the relaxation rate corresponding to $1/T_1$. It is clear from Fig. 1 that the stopped muons are subject to static fields at 0.1 and 1 K.[22] Since the spectra exhibit no precession and no clear dip after rapid decrease from the initial asymmetry, it is likely that the static fields are widely distributed and attributed to randomly frozen moments.

We assumed the Lorentzian distribution of the random fields because any rounding of the zero-field time spectrum

around the prompt time is not appreciable at 0.1 K as seen in the inset of Fig. 1. For the Lorentzian distribution of the static internal field,

$$p(H)=\frac{\gamma_\mu^3}{\pi^2}\frac{4\pi a|H|^2}{(a^2+\gamma_\mu^2|H|^2)^2}, \tag{2}$$

Kubo[23] derived the relaxation function as follows:

$$A(t)=A_0\left[\frac{1}{3}+\frac{2}{3}(1-at)\exp(-at)\right]. \tag{3}$$

If we leave the numerical factor $\frac{1}{3}$ which is fraction of the tail component as a free parameter $g$ and take into account random slow fluctuations with averaged fluctuation rate λ, then Eq. (3) may be rewritten approximately as

$$A(t)=A_0[g+(1-g)(1-at)\exp(-at)]\exp\{-(\lambda t)\}^{0.5}. \tag{4}$$

The solid curve in the inset is for $a=44\,\mu s^{-1}$ corresponding to $H_{\rm int}=0.05$ T. The internal field is so strong in the Al substituted compounds that the observation of the prompt drop of asymmetry is beyond the time resolution.

Using Eqs. (1) and (4), an analysis was performed for all the compounds. The results are shown in Fig. 2. At high temperatures, the time spectra show exponential relaxation, or $\beta=1$, from the full asymmetry in a longitudinal field of 10 mT applied to decouple the static nuclear dipolar fields. The temperature dependence of λ measured in a longitudinal magnetic field of 10 mT is shown in Fig. 2(a) for the three compounds. As the temperature is reduced, the exponents $\beta$ decrease gradually from 1 to 0.5 and the relaxation rate λ increases toward broad maxima at 2.5 and 45 K in the $x=0$ and 0.1 compounds, respectively. A pronounced thermal hysteresis was observed for the $x=0.02$ compound; λ shows a maximum at 20 K on heating after rapid cooling of the sample to 3 K, whereas the maximum shifts to 10 K when the spectra were measured on cooling. Since no long-range order exists, the broad maxima should be due to random freezing of the Mn moments. The maximum temperature will be denoted as $T_g$. In contrast to the previous results,[19] we did not notice clear indication of short-range ordering above $T_g$ although the initial asymmetry decreases by 10% above $T_g$ when the samples are cooled from 100 K.

The initial asymmetry above $T_g$ and the amplitude of the tail asymmetry below $T_g$ are shown in Fig. 2(b) as normalized to the full asymmetry. The tail asymmetry of $\frac{1}{3}$ is the static limit.[23] The asymmetries for the three compounds decrease suddenly below 2.5, 36, and 45 K, respectively. In the heating process of the $x=0.02$ compound, the asymmetry jumps up to the full value between 40 and 50 K. It should be noted that the sudden decreases occur at the same temperatures as the peaks of the relaxation rate in the $x=0$ and 0.1 compounds whereas the $x=0.02$ compound shows the sudden change in asymmetry at two or three times as high as the peak temperatures and the asymmetry measured on cooling decreases quite gradually below the temperature indicating gradual moment freezing.

We note that the slowing down of the Mn spin fluctuations occurs over a wide temperature range in the $x=0$ compound; λ reaches $\lambda_\infty$ at around 100 K which amounts to

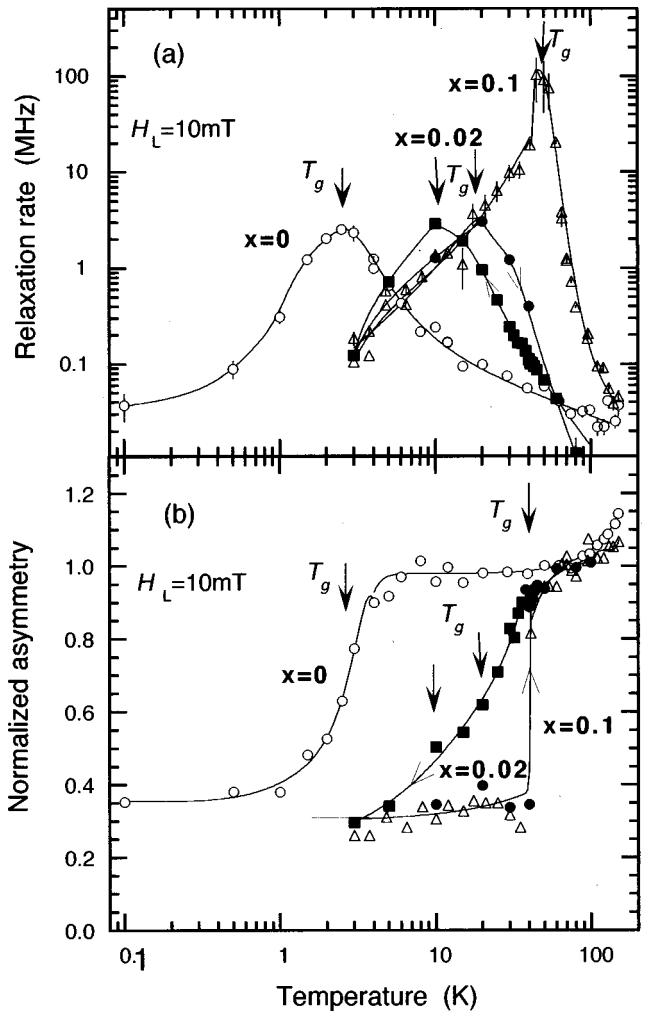


FIG. 2. Temperature dependence of (a) relaxation rate and (b) initial and tail asymmetries of $Y_{0.97}Sc_{0.03}(Mn_{1-x}Al_x)_2$ measured in longitudinal field of 10 mT. Marks ■ and ● show data for the $x=0.02$ compound taken on heating and cooling, respectively. Curves are guide to eyes.

$40T_g$. This is common in systems with strong frustration which suppresses the ordering or freezing of spins as observed in $SrCr_8Ga_4O_{19}$.[24] In contrast to this, in the $x=0.1$ compound, the slowing down occurs in the temperature range $T_g<T<2T_g$ as in conventional spin glasses.[25]

To verify the static nature of the internal field, we applied substantial longitudinal magnetic fields. Below $T_g$, the frozen moments give static random internal fields which may be decoupled by intense longitudinal magnetic fields, whereas above $T_g$, the Mn moments in the $x=0.1$ compound fluctuate very rapidly compared with the muon lifetime, giving time spectra unchanged in longitudinal fields.

Figure 3 shows the normalized amplitude of the tail component $g$ of the time spectra as a function of the applied longitudinal magnetic field measured at temperatures $T<T_g/10$. The strengths of the average internal field $H_{\rm int}$ were evaluated from the external field dependence of $g$ by the fit to the values calculated by the following equation:

$$g=\left\{1+\left(\frac{H_{\rm int}}{H_L}\right)^2\right\}\left\{1-\frac{H_{\rm int}}{H_L}\arctan\left(\frac{H_L}{H_{\rm int}}\right)\right\}. \tag{5}$$

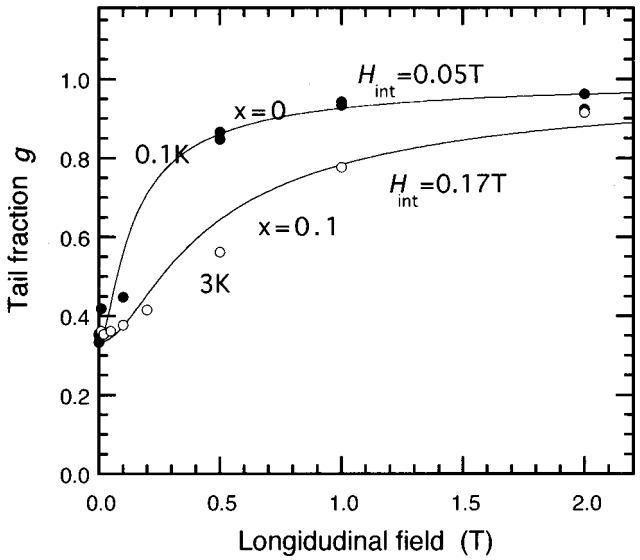


FIG. 3. Longitudinal field dependence of tail amplitudes in frozen states of the $x=0$ and $x=0.1$ compounds.

The solid curves in Fig. 3 are the longitudinal field dependence calculated for $H_{\rm int}=0.05\,{\rm T}$ and 0.17 T. The internal field in the Al substituted compound is much stronger, indicating the increase in magnitude and/or in number of magnetic moments along with the increase in $T_g$.

The values of $H_{\rm int}$ deduced from the field dependence of the tail asymmetry may be compared with 0.14 T obtained by computer simulation for dipolar field produced by the randomly frozen Mn moments of $1.3\mu_B$. This discrepancy between the observed and the calculated $H_{\rm int}$ for the $x=0$ compound is a natural consequence that the moments are originated from the soft modes of spin fluctuations. It is hard to tell if the observed internal field is produced by local magnetic order due to lattice imperfection such as Sc and muon charge, or by low-frequency component of the spin fluctuations. The internal field 0.17 T for the $x=0.1$ compound corresponds to the dipolar field from randomly frozen Mn moments of $1.6\mu_B$ which is comparable to the probable magnitude of the moments in the compound.

## IV. DISCUSSION

Contrary to our expectation from NMR and magnetic susceptibility, spin freezing was discovered at 2.5 K in $Y_{0.97}Sc_{0.03}Mn_2$ which is one order lower than $T_g$ of the $x=0.1$ compound, 45 K. The internal fields of 0.05 and 0.17 T were estimated for the $x=0$ and $x=0.1$ compounds at low temperatures, respectively. The Al substitution gives rise to much higher freezing temperatures, sharper transitions, and stronger internal magnetic fields in spite of magnetic dilution. In the $x=0.02$ compound, the freezing process of Mn spins is unusual. Although the decrease in the initial asymmetry is observed at 36 K, the relaxation rate shows the peak temperature at 10 and 20 K depending on the process of temperature change. Tail asymmetry measured on cooling decreases gradually below 36 K compared with the other compounds, while the temperature variation of the tail asymmetry on heating after rapid cooling is quite similar to that for the $x=0.1$ compound when temperature is increased.

These facts indicate that there is a wide distribution of freezing temperature in the $x=0.02$ compound. The $T_g$ corresponds to the average freezing temperature while the anomaly of the asymmetry occurs at the highest freezing temperature. A part of Mn atoms have $T_g\sim 36\,{\rm K}$ producing a large $H_{\rm int}$ at $\mu^+$ site and are quite similar to those in the $x=0.1$ compound and can be quenched by rapid cooling. On the other hand, some of Mn atoms with low $T_g$ are in the state which is relevant to $x=0$. The pronounced thermal hysteresis observed in the $x=0.02$ compound clearly shows that these two magnetic states are not continuously connected but are separated by a potential barrier of the order of $k_BT_g$. This is not surprising judging from that the transition from the nonmagnetic state to the magnetic state in $YMn_2$ is of first order.

It is evident from these results that Mn atoms in the $x=0$ and the $x=0.1$ compounds are of different nature just like those in the tetragonal and the cubic $YMn_2$ and their magnetic properties cannot be extrapolated to the other. It follows that $x=0.02$ is the critical concentration of magnetic and nonmagnetic transition.

We have measured the dc magnetic susceptibility with applied field of 10 mT. The Curie terms obtained above $T_g$ yield only Mn moments of 1.4% in $x=0$ and 5.5% in $x=0.1$ assuming $1.3\mu_B$ per Mn. This shows that strong antiferromagnetic correlation between Mn moments suppresses the temperature-dependent magnetic susceptibility even in $x=0.1$ in contrast with conventional spin glasses. We note that the ratio of the Curie term for these two concentrations are very close to that of the observed $H_{\rm int}$, 0.05 and 0.17 T which implies that the magnetically active nonsinglet moments detected in susceptibility are responsible for creating the internal fields at the muon site.

In the phase close to $x=0$, the magnetic dilution by Al raises $T_g$ appreciably due to partial release of frustration producing paramagnetic tetrahedra in addition to lattice expansion. This fact implies that the majority of Mn tetrahedra in $Y_{0.97}Sc_{0.03}Mn_2$ are singlets in the ground state whereas the remaining paramagnetic tetrahedra are frozen randomly at 2.5 K producing the internal magnetic field at the $\mu^+$ sites. The Curie-Weiss component of the magnetic susceptibility may be attributed to the paramagnetic tetrahedra which are probably associated with some imperfections of the lattice such as Sc atoms at Y sites. The extra charge of $\mu^+$ is another possible imperfection which may produce localized magnetic moment. The moments induced by the extra charge would be frozen in the same way at a temperature according to the degree of frustration of the surrounding Mn moments. Thus the more imperfections as Sc and Al give the higher freezing temperature. If this is the case, the ideal corner-sharing tetrahedral lattice of Mn atoms without imperfection would have an entirely singlet ground state.

Also in the ground state of classical spin systems, spins on each tetrahedron correlate with each other so as to vanish the total spin of each tetrahedron as shown by our Monte Carlo simulation for Ising spin.[26] However, the stopped muons would feel a finite internal magnetic field from the static Mn moments even though each tetrahedron forms antiferromagnetic spin arrangements. Accordingly, the singlet tetrahedra should be formed by coherent spin fluctuations as expected but their dynamics could not be studied by measur-

ing the muon relaxation due to the minor paramagnetic tetrahedra.

No anomaly was found in the nuclear relaxation rates of $^{45}$Sc and $^{55}$Mn in $Y_{0.97}Sc_{0.03}Mn_2$ at 2.5 K. But the line shape of NMR is expected to change at the temperature as observed in $\beta$-Mn.[27] This has not been studied yet in detail.

Recently Kondo *et al.*[28] reported heavy fermion behavior in metallic $LiV_2O_4$ which has $S=\frac{1}{2}$ moments on a corner-sharing tetrahedral lattice. They observed a tendency toward vanishing magnetic freezing with $\mu$SR for specimens with decreasing impurity concentration as detected by the Curie term. These features, i.e., metallic conductivity, large specific heat $\gamma$ value, and lower or vanishing $T_g$ for more perfect specimens, are common to the tendency found in Y(Sc)$Mn_2$.

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