Static magnetic order in metallic triangular antiferromagnet Ag$_2$MnO$_2$ detected by muon-spin spectroscopy

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The magnetic nature of the triangular antiferromagnet Ag$_2$MnO$_2$, which exhibits two magnetic transitions at $T_{m1} \sim 80$ K and $T_{m2} \sim 30$ K, has been studied with positive muon-spin rotation and relaxation ($\mu$SR) spectroscopy using a polycrystalline sample in the temperature ($T$) range between 300 and 1.8 K. Weak transverse-field $\mu$SR measurements suggest the appearance of a random internal magnetic field at $T$ below $T_{m1}$, while zero-field ($\mu$SR) measurements indicate the existence of static internal magnetic fields below $T_{m2}$. Furthermore, two components with $\sim 10$ times different precession frequencies but almost equivalent amplitudes in the ZF spectrum indicate the formation of a static but complex antiferromagnetic order below $T_{m2}$. The overall magnetic behavior is therefore clarified that the disordered moments appear below $T_{m1}=T_{m2}^\text{on}$, whereas static short-range antiferromagnetic order completes below $T_{m2}=T_{m2}^\text{off}$.

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I. INTRODUCTION

Layered transition-metal dioxides AMO$_2$ [which consist of alternating stacks of A and MO$_2$ planes, where A$^+$ stands for an alkali (Li$^+$, Na$^+$, K$^+$, Rb$^+$, and Cs$^+$), Ag$^+$ or (Ag$_x$)$_+$ ion and $M^{2+}$ for a transition-metal ion, and in which Li$^+$, Na$^+$, K$^+$, Rb$^+$, and Cs$^+$ ions with larger ionic radius. In particular, Ag$_2$NiO$_2$ has been thought to be a typical 2D system with $T_N=19.9$ K (Ref. 30) and specific-heat measurements ($T_N=19.7$ K).31] Among several combinations between Ag$_2$ and $M$ for AMO$_2$, only Ag$_2$NiO$_2$ and Ag$_2$MnO$_2$ have, to authors’ knowledge, been prepared thus far. We have therefore naturally extended our $\mu$SR experiments to Ag$_2$MnO$_2$. Disilver manganese dioxide Ag$_2$MnO$_2$ belongs to a monoclinic system with $a=0.5178$ nm, $b=0.2875$ nm, $c=0.8815$ nm, and $\beta=102.3^\circ$ at ambient $T$ (Ref. 20) (see Fig. 1), although it is difficult to obtain more detailed structural data because the $c$-
plane of the powder sample is easily aligned by pressing. In spite of its metallic conductivity down to 2 K due to a quarter-filled Ag $5s$ band, as in the case of Ag$_2$F (Ref. 32) and Ag$_2$NiO$_2$, the specific-heat $C_p$ measurements indicated a transition at $T_m1 = 80$ K, while the susceptibility ($\chi$) measurements showed a spin-glass like transition at $T_m2 = 22$ K under the magnetic field ($H$) of 1 kOe. Also, both the paramagnetic (PM) Curie temperature ($\Theta_p$) and the effective magnetic moment ($\mu_{eff}$) of Mn ions are estimated as $-370$ K and 4.93 $\mu_B$, where the latter value is equivalent to the spin-only value for the Mn$^{3+}$ ions with $S=2$ and $g=2.0 [\mu_{eff} = g\sqrt{S(S+1)}\mu_B]$. Very recently, Yoshida et al. proposed the possibility of the chirality transition on the 2DTL MnO$_2$ plane below $T_m1$. However, the microscopic magnetic nature of Ag$_2$MnO$_2$, to authors’ knowledge, has been less investigated by neutron, NMR, ESR, and $\mu^+\text{SR}$ measurements thus far.

Among the several magnetic measurement techniques available, $\mu^+\text{SR}$ is particularly sensitive to the local magnetic environment because the $\mu^+$ mainly feels $H_{int}$ due to its nearest neighbors and is specially sensitive to the short-range magnetic order, which sometimes appears in low-dimensional systems, while both neutron-scattering and $\chi$ measurements mainly detect long-range magnetic order; in particular, if the correlation length is very short, neutron-diffraction peaks broaden and eventually disappear.

Here, we report on both weak (relative to the spontaneous internal fields in the ordered state) transverse-field (wTF) and zero-field (ZF) $\mu^+\text{SR}$S for a polycrystalline Ag$_2$MnO$_2$ sample at temperatures between 1.8 and 300 K. The former method is sensitive to local magnetic order via the shift of the $\mu^+$ spin precession frequency in the applied field and the enhanced $\mu^+$ spin relaxation, while ZF $\mu^+\text{SR}$ is uniquely sensitive to weak local magnetic disorder in samples exhibiting quasistatic paramagnetic moments.

FIG. 1. (Color online) Crystal structure of monoclinic Ag$_2$MnO$_2$. The unit cell is plotted by broken lines.

FIG. 2. (Color online) Variation in the wTF-$\mu^+\text{SR}$ time spectra with $T$ for the Ag$_2$MnO$_2$ powder sample. The solid lines show the fitting results using Eq. (1).

II. EXPERIMENT

A powder sample of Ag$_2$MnO$_2$ was prepared at the ISSP of the University of Tokyo by a solid-state reaction technique using reagent grade Ag metal and MnO$_2$ powder as starting materials. A mixture of Ag and MnO$_2$ was heated at 650 °C for 24 h in an oxygen flow. Then, the calcined powder was ground thoroughly and fired again at 750 °C several times. A more detailed description of the preparation and characterization of the powder is presented in Ref. 20.

Susceptibility ($\chi$) was measured using a superconducting quantum interference device (SQUID) magnetometer (MPMS, Quantum Design) in the temperature range between 400 and 5 K under magnetic field $H=55$ kOe. For the $\mu^+\text{SR}$ experiments, the powder was pressed into a disk of about 20 mm diameter and 1 mm thickness and subsequently placed in a muon-veto sample holder. The $\mu^+\text{SR}$ spectra were measured on $\mu$E1 (Dolly) surface muon beam line at PSI in Switzerland. The experimental setup and techniques were described elsewhere.

III. RESULTS

A. wTF $\mu^+\text{SR}$ below 100 K

In order to first roughly assess the nature of the magnetic transition at $T_m1$ and $T_m2$, Fig. 2 shows the wTF spectra obtained in a field of 30 Oe at 100, 60, 50, 40, and 30 K on Ag$_2$MnO$_2$. As $T$ decreases from 100 K, the oscillation amplitude due to the wTF gradually decreases indicating the appearance of a spontaneous internal field $H_{int}$. The wTF-$\mu^+\text{SR}$ spectra were therefore fitted using a combination of a slowly relaxing precessing signal and a fast relaxing nonoscillatory signal. The first component is due to wTF and the latter due to $H_{int}$.

$$A_0P_{TF}(t) = A_{TF} \cos(\omega_{TF} t + \phi_{TF}) \exp(-\lambda_{TF} t)$$

$$+ A_{fast} \exp(-\lambda_{fast} t),$$

where $A_0$ is the initial ($t=0$) asymmetry, $P_{TF}(t)$ is the muon-spin-polarization function, $\omega_{TF}$ is the muon Larmor frequency corresponding to the applied wTF, $\phi_{TF}$ is the initial phase of the precessing signal, $\lambda_{TF}$ and $\lambda_{fast}$ are the exponen-
FIG. 3. Temperature dependences of (a) normalized $A_{TF}$ and $A_{fast}$, (b) TF relaxation rates ($\lambda_{TF}$), (c) fast relaxation rate ($\lambda_{fast}$), (d) TF frequency ($\omega_{TF}/2\pi$), and (e) susceptibility ($\chi$) for the powder Ag$_2$MnO$_2$ sample. The data were obtained by fitting the wTF spectrum using Eq. (1). $\chi$ was measured in both zero-field-cooling (ZFC) and field-cooling (FC) modes with $H=100$ Oe.

tial relaxation rates, and $A_{TF}$ and $A_{fast}$ are the asymmetries of the two components of the $\mu^*$SR signal.

Figure 3 shows the $T$ dependences of normalized $A_{TF}$ and $A_{fast}$ ($N_{TF}=A_{TF}/A_0$, $n=$ TF and fast) [Fig. 3(a)], $\lambda_{TF}$ [Fig. 3(b)], $\lambda_{fast}$ [Fig. 3(c)], $\omega_{TF}/2\pi$ [Fig. 3(d)], and $\chi$ [Fig. 3(e)]. Upon decreasing $T$ below 100 K, $N_{TF}$ [which corresponds to the volume fraction ($V_p$) of PM phases in the sample] shows a clear decrease below 70 K and reaches 0 at 30 K. The $T$ [at which $N_{TF}=0$ below $T_{m2}$ ($=30$ K), at which the $\chi(T)$ curve obtained in the zero-field-

FIG. 4. (Color online) ZF-$\mu^*$SR time spectra in the $T$ range between 40 and 120 K for the Ag$_2$MnO$_2$ powder sample. The solid lines show the fitting results using Eq. (2).

cooling mode exhibits a clear cusp, it is concluded that static order completes below $T_{m2}$. In contrast, $N_{TF}$ has a nonzero value below $T_{m1}$ and increases with decreasing $T$ and finally levels off around 0.5 below 30 K. The fact that $N_{TF}-0.5$ even at 5 K indicates the presence of a large $H_{int}$ below $T_{m1}$ as will be described later. The relaxation of the TF component, $\lambda_{TF}(T)$, shows a monotonic increase below $T_{m1}$ accompanying the decrease observed in $N_{TF}$, while $\lambda_{fast}(T)$ appears mainly below $T_{m2}$ and increases with decreasing $T$ indicating the evolution of $H_{int}$ below $T_{m2}$. From the viewpoint of wTF $\mu^*$SR, the two magnetic transitions ($T_{m1}$ and $T_{m2}$) are therefore assigned to be the onset ($T_{N}^{m1}$) and end point ($T_{N}^{m2}$) of the one AF transition with a transition width ($\Delta T_N$) of about 40 K. The midpoint of the transition, at which $N_{TF}=0.5$, is also estimated as $\sim 45$ K.

Although the $C_p$ measurements showed the peak at the magnetic transition at $T_{m1} (=T_N^{m1})$, the entropy change ($\Delta S_N$) from the phase below $T_{m1}$ to the high-$T$ paramagnetic phase was estimated as 0.36 JK$^{-1}$ mol$^{-1}$, which is significantly smaller than the expected value for the $S=2$ system (13.38 JK$^{-1}$ mol$^{-1}$). This is very consistent with the conclusion from the wTF measurements; that is, disordered moments appear below $T_{m1}$ because the small $\Delta S_N$ supports the absence of a conventional second-order AF transition at $T_{m1}$, below which the whole sample enters into the long-range AF ordered phase.

B. ZF $\mu^*$SR between 40 and 120 K

In order to elucidate the magnetic behavior above $T_N^{m1}$, in particular near $T_N^{m1} \sim 70$ K, we carried out ZF-$\mu^*$SR measurements in the $T$ range between 40 and 120 K (see Fig. 4). In spite of the absence of an oscillatory signal above $T_N^{m1}$, the relaxation rate is found to increase rapidly with decreasing $T$. Although it is difficult to fit the spectrum using a simple exponential relaxation function [exp$(-\lambda T)$] in the whole $T$ range between 40 and 120 K, the ZF-$\mu^*$SR spectrum is well fitted by a power exponentially relaxing signal,

$$A_p P_{ZF}(T) = A_p \exp[-(\lambda_{PE})^\beta].$$  

(2)

Here, “a power exponentially relaxing signal” has been observed for many dense-moment disordered magnetic
Accompanying the change in system\textsuperscript{33,34} in a paramagnetic state. The fit results are shown in Fig. 5 together with $\chi^{-1}$. As $T$ decreases from 120 K, $\beta \sim 1$ (simple exponential) down to 70 K then $\beta$ decreases monotonically with further lowering $T$, dropping below $\beta = 0.5$ (root exponential for a dilute-disordered magnet),\textsuperscript{35} to finally reach its lowest value ($1/3$ for a dense-disordered phase).\textsuperscript{36} This means a gradual increase in the number density of localized magnetic moments, which are detectable within the muon time scale with decreasing $T$ below $T_N^\text{end}$. Accompanying the change in $\beta$, $\lambda_{\text{FE}}$ also increases very rapidly below 70 K with decreasing $T$ indicating a broadening of the distribution of $H_{\text{int}}$. This result is therefore consistent with the scenario that disordered moments appear below $T_N^a$ and develop with decreasing $T$, and finally static order completes below $T_N^\text{end}$.

Since the slope of the $\chi^{-1}(T)$ curve was found to start to deviate downward from the linear relationship below around 120 K [see the inset of Fig. 5(c)], a FM interaction would contribute in determining the magnetic order in the MnO$_2$ plane as will be discussed later.

FIG. 5. Temperature dependences of (a) $\beta$, (b) $\lambda_{\text{FE}}$, and (c) $\chi^{-1}$ for the Ag$_2$MnO$_2$ powder particularly above $T_N^\text{end}$. The ZF data were obtained by fitting using Eq. (2), whereas the wTF data using $A_{\text{TF}} \cos(\omega_{\text{TF}} t + \phi_{\text{TF}}) \exp[-(\lambda_{\text{FE}}) \beta]$. $\chi$ was measured in both ZFC and FC modes with $H=10$ kOe. The inset of (c) shows the $\chi^{-1}(T)$ curve below 400 K.

C. ZF $\mu$SR below 40 K

Following upon the $\mu$SR experiments above 40 K, we measured the ZF-$\mu$SR spectrum in order to investigate the detail of $H_{\text{int}}$ below $T_N$. Figure 6 shows ZF-$\mu$SR time spectra in an early time domain (below 200 ns) in the $T$ range between 1.8 and 35 K for a powder sample of Ag$_2$MnO$_2$. One can clearly see two heavily damped oscillatory components with different frequencies below 30 K; that is, at 1.8 K, one component exhibits the first minimum at around 6 ns and the second minimum at around 18 ns, while the other component shows the first minimum at around 55 ns and the second broad minimum at around 170 ns. This unambiguously establishes the existence of static magnetic order in the sample. However, comparing the result on Ag$_2$NiO$_2$ [see Fig. 6(b)],\textsuperscript{22} the ZF spectrum for Ag$_2$MnO$_2$ is found to be heavily damped even at 1.8 K. This suggests that static order is unlikely long ranged but likely short ranged, although $\mu^*$SR yields no information on correlation length of magnetic order. As $T$ is increased from 1.8 K, the minimum positions shift toward longer times reflecting the expected decrease in

FIG. 6. (Color online) Temperature dependence of the ZF-$\mu$SR time spectra of a powder sample of (a) Ag$_2$MnO$_2$ and (b) Ag$_2$NiO$_2$ (Ref. 22). Each spectrum is offset by (a) 0.05 and (b) 0.2 for clarity of the display. The solid lines in (a) represent the fitting result using Eq. (3). Since the ZF time spectrum for Ag$_2$MnO$_2$ is very strongly damped, it is difficult to get a reliable Fourier transform spectrum.
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Above 35 K, the oscillatory components disappear and the ZF spectrum is just relaxing with time.

We therefore use a combination of two exponentially relaxing cosine oscillation signals and a slow exponential relaxing nonoscillatory signal for fitting the ZF spectrum,

\[ A_0 P^Z(t) = A_{AF1} \cos(\omega_{AF1}^{\mu} t + \phi) \exp(-\lambda_{AF1} t) + A_{AF2} \cos(\omega_{AF2}^{\mu} t + \phi) \exp(-\lambda_{AF2} t) + A_{tail} \exp(-\lambda_{tail} t), \]

where \( \lambda_{AF1} \), \( \lambda_{AF2} \), and \( \lambda_{tail} \) are the relaxation rates, \( \omega_{AF1}^{\mu} \) and \( \omega_{AF2}^{\mu} \) are the muon precession frequencies due to \( H_{int} \) \( \left[ \omega_{AF1}^{\mu}/2\pi = f_{AF1} = 13.554 \text{ kHz/Oe} \right] \), and \( \phi \) is the initial phase of the precession. The fitting yields \( |\phi| \) ranges below 28° well below \( T_N^{end} \) due to a wide field distribution of \( H_{int} \). Although we used a common \( \phi \) for the two cosine oscillations, the fit using two independent \( \phi \)'s also provides a similar result, that is, \( |\phi| \approx 28^\circ \). This suggests commensurate AF order below \( T_N^{end} \) because the cosine fit for an IC field should yield \( \phi = -45^\circ \) for the both cosine functions,37,38 as muons locate at the sites that are commensurate to the lattice. The \( A_{tail} \) signal corresponds to the “1/3 tail” caused by the AF field component parallel to the initial muon-spin polarization \( \{S_{\mu}(0)\} \).

Figures 7(a)–7(d) show the T dependence of the muon precession frequencies \( (f_{AF}^{\mu} = \omega_{AF}^{\mu}/2\pi) \), the normalized \( A_{AF1} \), \( A_{tail} \), and \( A_{TF} \), the exponential relaxation rates \( \lambda_{AF1} \) and \( \lambda_{AF2} \), and the ratio of the precession frequencies and amplitudes of the two oscillatory signals, i.e., \( T_N \) and \( A_{AF1}/A_{AF2} \) for the powder sample of \( Ag_2MnO_2 \). As \( T \) decreases from \( T_N^{end} \), both \( f_{AF1} \) and \( f_{AF2} \) increase with decreasing slope \( (df_{AF}^{\mu}/dT) \) and roughly level off at their highest values below about 20 K. Here, the \( f_{AF1}^{\mu}(T) \) curve seems to be very similar to the \( f_{AF2}^{\mu}(T) \) curve, although \( f_{AF1}^{\mu} \) is larger by one order of magnitude than \( f_{AF2}^{\mu} \) in the whole \( T \) range below \( T_N^{end} \). This means that the two \( f_{AF1}^{\mu} \)'s are not caused by two different AF phases but by two different muon sites in the AF ordered lattice.

The \( A_{AF1}(T) \) and \( A_{AF2}(T) \) curves are almost independent of \( T \) except at the vicinity below \( T_N^{end} \). The population of the two muon sites is therefore not altered with \( T \). This is in good agreement with the fact that muons form a stable bond with \( O^{2-} \) ions at low \( T \). Furthermore, \( A_{tail} \) is roughly a constant \( (~0.2) \) at \( T \) below \( T_N^{end} \), which is consistent with the behavior of the 1/3 tail, although the magnitude of \( A_{tail} \) is smaller than the predicted value for a powder sample (1/3).

This is probably due to the preferred orientation of the pressed powder sample.20 On the contrary, since the c plane tends to align by pressing because of the 2D structural nature of \( Ag_2MnO_2 \), the smaller \( A_{tail} \) for the pressed powder than that for a random powder (1/3) would imply that \( H_{int} \perp c \) as \( S_{\mu}(0) \) is perpendicular to the basal plane. Note that all the three ZF signals appear below \( T_N^{end} \), at which \( A_{TF} \) reaches 0. This strongly supports that \( T_{int} = T_N^{end} \).

The two relaxation rates \( (\lambda_{AF1} \) and \( \lambda_{AF2} \)) are also roughly \( T \) independent below \( T_N^{end} \), suggesting that the field distribution is fixed at \( T_N^{end} \) and is not developed with decreasing \( T \). Indeed, the shape of the ZF-\( \mu \)SR spectrum at 25 K is essentially the same as at 1.8 K, whereas the minimum positions delay with increasing \( T \). In general, as \( T \) decreases,
were obtained by fitting the 14 ZF spectra below $T_N$ using common $\phi$, $A_{AF1}$, $A_{AF2}$, $A_{tail}$, and $f_{AF1}/f_{AF2}$ for Eq. (3). The solid line in (a) represents the fitting result using Eq. (4).

because $|\phi|$=28° and the rest parameters are almost T independent as seen in Fig. 7. The "global fit" provided that $\phi$ =−8 ± 5°, $A_{AF1}$=0.1027 ± 0.0008, $A_{AF2}$=0.0935 ± 0.0012 ($A_{AF1}/A_{AF2}$=1.07 ± 0.05), $A_{tail}$=0.0263 ± 0.0008, and $f_{AF1}/f_{AF2}$=1.7 ± 0.4, respectively. Figure 8 shows the T dependences of $f_{AF2}$, $\lambda_{AF2}$, and $\lambda_{AF1}$ obtained by the global fit. The $f_{AF2}(T)$ curve, which is the T dependence of an order parameter of the transition, is well fitted by the following expression:

$$f_{AF}(T) = f_{AF}(0 \text{ K}) \left(\frac{T_N - T}{T_N}\right)^{\beta_m}.$$  \hspace{1cm} (4)

This provides $f_{AF2}(0 \text{ K})$=7.66 ± 0.14 MHz, $T_N$=30.8 ± 0.5 K, and $\beta_m$=0.28 ± 0.03 [see Fig. 8(a)]. The exponent ($\beta_m$) obtained is comparable to the predictions for the 2D $xy$ model on a triangular lattice ($\beta_m$=0.25 ± 0.02),\tsci{39,40} although we need more accurate data in the vicinity of $T_N$ to determine $\beta_m$ more precisely. Also, the obtained value for Ag$_2$MnO$_2$ is 15% larger than $\beta_m$ for NaNiO$_2$ estimated by $\mu^*$SR measurements (0.24 ± 0.01) (Ref. 27) and is almost comparable to $\beta_m$ for Na$_{0.75}$CoO$_2$ (−0.28).\tsci{41}

**IV. DISCUSSION**

Although the AF spin structure has not been solved yet, here we show that we can exclude several possible AF structures from the present $\mu^*$SR results. Note that since the structural data at low T is unavailable, we assume that there are no structural phase transitions down to the lowest T measured. This is consistent with the results of the $C_p$ and $\chi$ measurements.\tsci{20}

The relationship between magnetization ($M$) and $H$ exhibits hysteresis only below $T_N$ (Fig. 9). This could imply that the Mn spins freeze below $T_N$. However, the two oscillations in the ZF spectrum (see Fig. 6), even if there are only first two minima for the both oscillations, clearly demonstrate the formation of static AF order below $T_N$. Furthermore, although the ac-$\chi(T)$ curve shows a cusp at $T_N$ as in the case of dc $\chi$, the T of the cusp is found to be independent of the ac frequency.\tsci{42} This strongly supports the static AF order for Ag$_2$MnO$_2$ at low $T$.

Moreover, the cosine fit rules out the possibility of IC-spin-density wave (SDW) order below $T_N$, which is in contrast to the case of Ag$_2$NiO$_2$. Actually, the T-independent $f_{AF1}/f_{AF2}$ and $A_{AF1}/A_{AF2}$ below $T_N$ are reasonably explained by the coexistence of two magnetically different muon sites in the C-AF ordered lattice. On the other hand, if we assume the IC-SDW field, $f_{AF1}$ and $f_{AF2}$ naturally correspond to the upper and lower limits of the field distribution. The relative field distribution width [1−($f_{AF2}/f_{AF1}$)] should thus be T independent. However, the wave vector of IC-SDW often depends on $T$.\tsci{43} This naturally implies the change in the IC field distribution with $T$ resulting in the “T dependence” of $f_{AF1}/f_{AF2}$ and $A_{AF1}/A_{AF2}$. Therefore, we decided that the phase below $T_N$ is a static but short-range C-AF ordered phase.

Next we discuss the case for chirality order on the 2DTL MnO$_2$ plane (see Fig. 10) proposed by the recent $C_p$ and $\chi$ measurements.\tsci{20} As is well known, $\mu^*$SR is very sensitive to the local magnetic environment. Hence, if the chirality ordered state appears in the MnO$_2$ plane, we could expect to observe the damped oscillations in the ZF spectrum as we have already obtained in Fig. 6, although the chirality ordered state was originally proposed for the phase in the $T$ range between $T_N$ and $T_N^{\text{end}}$.\tsci{20} At first, it should be noted that

**FIG. 8.** (Color online) Temperature dependences of (a) $f_{AF2}$, (b) $\lambda_{AF2}$, and (c) $\lambda_{AF1}$ for the powder sample of Ag$_2$MnO$_2$. The data were obtained by fitting the 14 ZF spectra below $T_N$ using common $\phi$, $A_{AF1}$, $A_{AF2}$, $A_{tail}$, and $f_{AF1}/f_{AF2}$ for Eq. (3). The solid line in (a) represents the fitting result using Eq. (4).

**FIG. 9.** (Color online) Magnetization ($M$) vs magnetic field ($H$) for Ag$_2$MnO$_2$ at several temperatures. Although $M$ was measured both on increasing $H$ and on decreasing $H$, the $MH$ hysteresis is only observed below 30 K.
the triangular lattice of Ag₂MnO₂ is slightly distorted from the regular triangle to the isosceles triangle as seen in the top of Fig. 10. The length of the bottom edge, which is parallel to the b axis, is shorter by ~3% than that of the rest two edges. According to electrostatic-potential calculations using the atomic positions for Ag₂NiO₂ but lattice parameters for Ag₂MnO₂, μ⁺'s are most likely to locate at the middle of the two long edges of the Mn triangular lattice but above or below the Mn plane about 1.7 Å away.

Then, simple dipole field calculations yield that there are three magnetically different μ⁺'s sites represented by solid (I), open (II), and crossed (III) circles in Fig. 10. When a magnitude of the Mn moment (μₘₙ) is 1μ₈, H_{int,1} =1965 Oe, H_{int,II}=1927 Oe, and H_{int,III}=792 Oe with the muon occupancy ratio H_{int,1}:H_{int,II}:H_{int,III}=1:1:1. If we assume that μₘₙ is comparable to μ_{eff}(=4.93μ₈), three oscillation frequencies would be observed in the ZF spectrum at f₁=131 MHz, f₁=129 MHz, and f₁=53 MHz for the chirality ordered phase [Fig. 10(a)]. In contrast, the present μ⁺SR gives two quite different frequencies (one is 11.7 times larger than the other) with almost equal number of densities [see Figs. 7(d) and 7(e)].

It is, thus, very difficult to explain the present μ⁺SR data based only on the μ⁺'s sites at the vicinity of the MnO₂ plane. Therefore, we considered two crystallographically different μ⁺ sites in the Ag₂MnO₂ lattice in order to explain the two f signals. That is, one is the site shown in Fig. 10 (IV =I, II and III) and the other is the interstitial site in the metallic Ag₂ plane (V) as in the case of metals and/or alloys. The calculations provide that H_{int,IV}/μₘₙ =1600±700 Oe/μ₈=1/2μₘₙ(H_{int,IV}/μₘₙ), H_{int,II}/μₘₙ =60±20 Oe/μ₈, and H_{int,III}/μₘₙ=30±40, respectively. The wide field distribution of the two H_{int}'s is consistent with the heavily damped oscillation in the ZF spectrum. However, even if μₘₙ=μ_{eff}, the magnitude of H_{int,III}=300±100 Oe, which corresponds to f_{int,III}=4.1±1.4 MHz, is about half of the experimental result [I_{AF1}(0 K)~7.7 MHz]. Here, considering a large estimation error for H_{int,IV}/H_{int,V} due to the wide field distribution of each H_{int}, the calculated ratio is not crucial to judge the formation of the chirality ordered phase below Tₘ. Furthermore, the μ⁺ occupancy at site IV should be accidentally equivalent to that at site V, while the vicinity of O²⁻ ions (site IV) is thought to be most stable for μ⁺. Finally, it should be noted that the ZF-μ⁺SR spectra for Ag₂NiO₂, which has almost the same crystal structure and charge distribution as Ag₂MnO₂, exhibits a very clear oscillation with two similar frequency components, i.e., 22 and 26 MHz at 1.9 K. If we assume that μ⁺'s sit at site V in Ag₂MnO₂, μ⁺'s are naturally likely to do so also in Ag₂NiO₂. However, the estimated ratio of the two frequencies is too small to explain the result of Ag₂NiO₂, and therefore μ⁺'s are most unlikely to sit at site V both in Ag₂NiO₂ and Ag₂MnO₂. These suggest that the chirality order is unlikely to explain the present result.

The calculations were also performed for the following spin arrangements and the results are listed in Table 1: (i) Chirality ordered phase with 120° spin structure [see Fig. 10(b)]. (ii) Collinear phase, in which the spins are aligned parallel to the b axis, i.e., the FM chain is formed along the b axis, but the neighboring FM chains are antiparallel to each other. (iii) A-type AF phase. Nevertheless, there is no reasonable spin arrangement for explaining the magnitude of f_{AF1} and the ratio of f_{AF1}/f_{AF2} (~1.17) and A_{AF1}/A_{AF2} (~1.07), simultaneously.

For the A-type AF case, the H_{int} (f₂) ratio between sites IV and V is calculated as 16.3, which is the most proximate to the experimental result (11.7). We thus discuss the possibility of A-type AF order in detail. At first, we should note that even if μₘₙ=μ_{eff}, the magnitude of f_{int,IV} (~43 MHz) for A-type AF order is about half of the experimental result [I_{AF1}(0 K)~90 MHz]. The question on the occupancy ratio between sites IV and V still remains the same to the case of the chirality ordered phase as mentioned above. If we ignore these inconsistency, although Θₘ=~370 K, the FM contribution observed in the χ''(T) curve below ~120 K would support an FM arrangement of the Mn spins in the MnO₂ plane as predicted theoretically for LiNiO₂. For the A-type AF order state, past μ⁺SR work, however, reported a very clear oscillation with small damping as in the case of Na₈NiO₂ (Ref. 27) and Na₀.₇₅CoO₂. Actally, the present calculation also predicts only one f signal for μ⁺'s at site IV. Hence, a very clear oscillation is expected to be observed in the ZF spectrum, which is in contrast to the experimental result. These suggest that the A-type AF order is most unlikely to explain the present result.
TABLE I. Dipole field $H_{\text{int}}/\mu_\text{Mn}$ (top), muon precession frequency $f_\mu/\mu_\text{Mn}$ (middle), and muon occupancy ratio $R_{\text{mo}}$ (bottom) at the muon sites I–V in several ordered structures. Here, site I is crystallographically the same as sites II and III (see Fig. 10). When site I is thought to be magnetically the same as sites II and III, sites I, II, and III are represented by site IV. For such case, the interstitial site in the metallic $\text{Ag}_2$ plane (site V) is considered in order to explain the two $f$ signals. Since $H_{\text{int}}(f_\mu)$ is proportional to the ordered moment ($\mu_\text{Mn}$), its unit is given by $\text{Oe}/\mu_\text{B}$ (MHz/\mu_\text{B}). Assuming that $\mu_\text{Mn}=\mu_\text{Ag}=4.93\mu_\text{B}$, the highest $H_{\text{int}}$ ($f_\mu$) is equivalent to 4.93($\mu_\text{Mn}/\mu_\text{B}$). Here, $f_\mu=H_{\text{int}}(13.554 \text{ kHz/Oe})$. We also calculated $H_{\text{int}}$ for site IV’, which is the middle of one short edge of the Mn triangular lattice (see the top of Fig. 10) and is more unstable than site IV according to electrostatic-potential calculations. For the collinear spin structure, $H_{\text{int},IV}/H_{\text{int},IV}=7.96$ and $R_{\text{mo},IV}/R_{\text{mo},IV}=2$ (11.7 and 1.07 from the experiment), if we assume that the electrostatic potential for site IV’ is equivalent to that for site IV. For the other spin structures, muons at site IV’ give an additional field distribution width to $H_{\text{int},IV}$ but do not produce the two quite different $H_{\text{int}}$’s. Therefore, even if we consider the contribution of the muons at site IV’, it is still difficult to explain the experimental result.

<table>
<thead>
<tr>
<th>Structure</th>
<th>Site I</th>
<th>Site II</th>
<th>Site III</th>
<th>Site IV</th>
<th>Site IV’</th>
<th>Site V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chiral xy</td>
<td>$H_{\text{int}}/\mu_\text{Mn}$ ($\text{Oe}/\mu_\text{B}$)</td>
<td>1965</td>
<td>1927</td>
<td>792</td>
<td>1600±700</td>
<td>1400±700</td>
</tr>
<tr>
<td></td>
<td>$f_\mu/\mu_\text{Mn}$ (MHz/\mu_\text{B})</td>
<td>26.6</td>
<td>26.1</td>
<td>10.73</td>
<td>21±9</td>
<td>20±9</td>
</tr>
<tr>
<td></td>
<td>$R_{\text{mo}}$</td>
<td>1/3</td>
<td>1/3</td>
<td>1/3</td>
<td>1</td>
<td>1/2</td>
</tr>
<tr>
<td>Chiral 120°</td>
<td>$H_{\text{int}}/\mu_\text{Mn}$ ($\text{Oe}/\mu_\text{B}$)</td>
<td>1324</td>
<td>1268</td>
<td>2200</td>
<td>1600±500</td>
<td>1500±500</td>
</tr>
<tr>
<td></td>
<td>$f_\mu/\mu_\text{Mn}$ (MHz/\mu_\text{B})</td>
<td>17.95</td>
<td>17.19</td>
<td>29.8</td>
<td>22±7</td>
<td>20±7</td>
</tr>
<tr>
<td></td>
<td>$R_{\text{mo}}$</td>
<td>1/3</td>
<td>1/3</td>
<td>1/3</td>
<td>1</td>
<td>1/2</td>
</tr>
<tr>
<td>Collinear</td>
<td>$H_{\text{int}}/\mu_\text{Mn}$ ($\text{Oe}/\mu_\text{B}$)</td>
<td>1098</td>
<td>138</td>
<td>923</td>
<td></td>
<td></td>
</tr>
<tr>
<td>FM along $b$ but AF along $a$</td>
<td>$f_\mu/\mu_\text{Mn}$ (MHz/\mu_\text{B})</td>
<td>14.88</td>
<td>1.87</td>
<td>12.51</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$R_{\text{mo}}$</td>
<td>1</td>
<td>1/2</td>
<td>?</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$A$-type AF</td>
<td>$H_{\text{int}}/\mu_\text{Mn}$ ($\text{Oe}/\mu_\text{B}$)</td>
<td>652</td>
<td>490</td>
<td>40</td>
<td></td>
<td></td>
</tr>
<tr>
<td>FM in the plane but AF along $c$</td>
<td>$f_\mu/\mu_\text{Mn}$ (MHz/\mu_\text{B})</td>
<td>8.84</td>
<td>6.64</td>
<td>0.54</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$R_{\text{mo}}$</td>
<td>1</td>
<td>1/2</td>
<td>?</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Interestingly, a fast relaxing signal is also reported to appear in the ZF spectrum below $\sim 24$ K for NaNiO$_2$, whereas $T_N=20$ K. On the other hand, such signal does not exist above $T_N$ for Na$_{0.75}$CoO$_2$. Indeed, as $T$ decreases from 30 K, the $\chi(T)$ curve for Na$_{0.75}$CoO$_2$ exhibits a very sharp drop (a steplike decrease) at $T_N=22$ K. Therefore, the appearance of the fast relaxing signal above $T_N$, which is observed in $\text{Ag}_2\text{MnO}_2$, is unlikely to correlate the formation of $A$-type AF order.

In conclusion, although we can exclude the $A$-type AF and collinear ordered phases from the present $\mu^\ast$SR result, the magnetic structure is still not completely elucidated. In order to further understand the AF spin structure below $T_N^{*}$, $\mu^\ast$SR experiments using single crystals or aligned powder samples are required to know the magnetic anisotropy of $\text{Ag}_2\text{MnO}_2$. Furthermore, neutron-scattering measurements would provide additional crucial information on existence/absence of AF order below $T_N^{*}$, given its different spatial and time resolution from those of $\mu^\ast$SR. Moreover, a detailed structural analysis, preferably in a synchrotron-radiation source, would yield significant information on the possible slight displacement of O ions and/or the small distortion of the triangular lattice, which could be undetectable by the $C_p$ and $\chi$ measurements.

V. SUMMARY

A positive muon-spin rotation/relaxation ($\mu^\ast$SR) experiment on a powder sample of $\text{Ag}_2\text{MnO}_2$, in which Mn ions form the two-dimensional triangular lattice in the MnO$_2$ plane, has demonstrated the existence of a static but short-ranged AF order below $T_N^{*}=30$ K, while disordered moments appear below $T_N^{*}=80$ K. The strongly damped oscillatory signal in the ZF spectrum, however, indicates a wide field distribution at the muon sites even at 1.8 K due to the geometrical frustration of the triangular lattice. Although the AF spin structure is still unknown, since the $\text{Ag}_2$ plane is expected to reduce or eventually annihilate the interplane...
interaction between the adjacent MnO2 planes, further studies of the magnetic properties of the MnO2 plane particularly the ground state of the AF phase should yield significant information on the physics of these unique frustrated systems.

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42 Z. Hiroi (private communication).


