μSR investigation of magnetically ordered states in the A-site ordered perovskite manganites

**RBaMn$_2$O$_6$ (R = Y and La)**

Y. Kawasaki,1,* T. Minami,1 M. Izumi,1 Y. Kishimoto,1 T. Ohno,1 K. H. Satoh,2 A. Koda,2,3 R. Kadono,2,3 J. L. Gavilano,4 H. Luetkens,5 T. Nakajima,6 and Y. Ueda7

1Institute of Technology and Science, The University of Tokushima, Tokushima 770-8506, Japan
2Graduate University for Advanced Studies (SOKENDAI), Tsukuba 305-0801, Japan
3Muon Science Laboratory, Institute of Materials and Structure Science, High Energy Accelerator Research Organization (KEK), Ibaraki 319-1106, Japan
4Laboratory for Neutron Scattering, PSI and ETHZ, Villigen CH-5232, Switzerland
5Laboratory for Muon-Spin Spectroscopy, Paul Scherrer Institut, Villigen CH-5232, Switzerland
6AMRI, National Institute of Advanced Industrial Science and Technology, Tsukuba 305-8565, Japan
7Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan

DOI: 10.1103/PhysRevB.86.125141 PACS number(s): 75.25.Dk, 76.75.+i

**I. INTRODUCTION**

In the last few decades a great deal of experimental and theoretical research efforts have been devoted to perovskite manganites with chemical formulas of $R_{1-x}^3A_{x}^{3+}$Mn$_3$O$_7$ ($R^{3+}$ = rare earth ions, $A^{3+}$ = alkaline-earth ions) because they show a rich variety of fascinating electromagnetic properties, such as the colossal magnetoresistance (CMR), charge and orbital ordering (COO), and metal-insulator transition. Recently, a great deal of attention is given to A-site ordered perovskite manganites, materials with an ordered arrangement of two cations, $R^{3+}$ and $A^{3+}$, at the A site of the perovskite structure.1,2

$RBaMn$_2$O$_6$, an A-site-ordered manganite is formed by substituting 50% of rare earth by barium ions in $RMnO_3$.3,4 The crystal structure of $RBaMn$_2$O$_6 is schematically shown in Fig. 1. It has a layered ordering of $R^{3+}$ and $Ba^{+2}$ ions along the c axis, resulting in a stacking of two-dimensional structures with the sequence of $-RO$-MnO$_2$-BaO-MnO$_2$ planes. The structure of $RBaMn$_2$O$_6 at room temperature has a tetragonal $ap \times bp \times cp$ cell ($ap = bp$) with no tilt of MnO$_6$ octahedra for $R =$ La, Pr, and Nd (dashed lines in Fig. 1), while for $R =$ Sm-Y the structure has a larger unit cell of $\sqrt{2}ap \times \sqrt{2}bp \times 2cp$ with a tilt of MnO$_6$ octahedra.3,5 Here the three lattice constants $ap$, $bp$, and $cp$ are associated with the cell of the corresponding A-site disordered $R_{0.5}Ba_{0.5}$Mn$_3$O$_7$.2 The $\sqrt{2}ap \times \sqrt{2}bp \times 2cp$ unit cells for $R =$ Sm, Eu, and Gd are orthorhombic but those for $R =$ Tb, Dy, Ho, and Y are monoclinic.5-9

The ordering of $R^{3+}$ and $Ba^{+2}$ ions dramatically changes the phase diagram of the disordered $R_{0.5}Ba_{0.5}$Mn$_3$O$_7$.2 The remarkable features of the A-site ordered $RBaMn$_2$O$_6 are the following. (i) The COO transition temperature $T_{CO}$ is very high for $R =$ Sm-Y, while magnetic glassy states are common in $R_{0.5}Ba_{0.5}$Mn$_3$O$_7$ except for $R =$ La. (ii) In the phase diagram of temperature $T$ vs the ratio of ionic radius $r_i$ ($\cong r_{\text{La}}/r_{\text{Ba}^{+2}}$), there is an abrupt phase transition near $r_i =$ 0.785 between a COO and ferromagnetic metal (FM) states with critical temperatures $T_{CO}(r_i)$ and $T_C(r_i)$, respectively. Near the phase boundary no drastic changes in $T_C$ or $T_{CO}$ are observed. The second feature of $RBaMn$_2$O$_6 is considered to be a key characteristic for the development of new materials that exhibit CMR at room temperature.10,11

Among the series of $RBaMn$_2$O$_6, $YBaMn$_2$O$_6$ has the highest ordering temperatures; a structural change from a triclinic to a monoclinic at $T_i \approx 520$ K, charge and orbital order at $T_{CO} \approx 480$ K, and an antiferromagnetic spin order with $T_N$ just below 200 K.12-15 The distinct hysteresis at $T_i$ and $T_N$ both in the susceptibility and the electrical resistivity indicate these transitions to be first order, while the transition at $T_{CO}$ was suggested to be second order.6 These transition temperatures are far higher than the spin-glass transition temperature of about 50 K in the disordered form $Y_{0.5}Ba_{0.5}$Mn$_3$O$_7$. The high $T_{CO}$ is considered to be due basically to the stacking order of $Y^{3+}$ and $Ba^{+2}$ in the structure, which results in the absence of electrostatic potential disorder.

The ordering patterns of orbital, charge, and spin in $YBa$_2$Mn$_2$O$_6$ are also closely related to the peculiar structure to the A-site ordered perovskite.13 The charge order below $T_{CO}$ is suggested to be a checkerboard type in the MnO$_2$ plane. The orbital ordering pattern in the MnO$_2$ plane is the so-called

1098-0121/2012/86(12)/125141(8) 125141-1 ©2012 American Physical Society
The rearrangement of the stacking pattern have performed muon-spin-relaxation (μSR) on YBaMn$_2$O$_6$ and LaBaMn$_2$O$_6$. We stress that these two compounds provide a unique opportunity to study different aspects of the A-site order, which result in dramatic changes of the physical properties in the Y compound, and a uniform ground state may be attributed directly to the A-site order. As explained above the situation is very different for the La compound.

Our results reveal an inhomogeneous magnetic FM phase for the A-site ordered LaBaMn$_2$O$_6$. This is indicated by the absence of muon spin oscillation in the μSR time spectra and the significant deviation from a single exponential form in the relaxation. The spatially inhomogeneous spin dynamics in the FM phase may be naturally associated with intimate intermixing of different magnetic phases, such as in the case of the Griffiths phase. Within this magnetically inhomogeneous phase, an antiferromagnetic state develops below 150 K, which displays well defined static internal magnetic fields, but reaches only 30% of the volume fraction at low temperatures. In the AFI(CE) state of YBaMn$_2$O$_6$, the magnetic properties extracted from the μSR results agree with the expectations for a charge-ordered antiferromagnet, that is, the observed muon spin oscillating frequencies are consistent with the checkerd-type charge order and the antiferromagnetic spin order proposed by the neutron diffraction study, where $e_g$ electrons of Mn$^{3+}$ ions are well localized.

II. EXPERIMENTAL PROCEDURE

Powders of RBaMn$_2$O$_{5+x}$ ($R = Y$ and La) were obtained by a solid-state reaction of R$_2$O$_3$, BaCO$_3$, and MnO$_2$. After repeating the sintering process in pure Ar gas, the obtained ceramics were annealed in flowing O$_2$ gas, resulting in the A-site ordered perovskite RBaMn$_2$O$_6$. The details of the preparation method are described in the literature. The crystal structure and magnetic properties of both samples were measured and found to be consistent with the published data.

The samples for the present μSR measurements were used in the previous $^{55}$Mn-NMR experiments. In the $^{55}$Mn-NMR spectrum, one can clearly distinguish between the A-site ordered LaBaMn$_2$O$_6$ and the disordered La$_{0.5}$Ba$_{0.5}$MnO$_3$ from their resonance frequencies. We confirmed that the majority of the material is A-site ordered for both samples; the degree of A-site order is about 92% for LaBaMn$_2$O$_6$. As for YBaMn$_2$O$_6$, we did not observe $^{55}$Mn-NMR signal originating from impurity or secondary phases, although the x-ray powder diffraction detected minor impurity of BaMnO$_{3-δ}$ (less than 1%).

The μSR data for YBaMn$_2$O$_6$ have been taken by using surface muon at the M20 muon channel at TRIUMF (Vancouver, Canada) and on GPS at PSI (Villigen, Switzerland). For the μSR measurements on LaBaMn$_2$O$_6$, we have employed the M20 muon channel at TRIUMF and the π A muon channel at KEK (Tukuba, Japan). All the μSR data presented here have been measured in zero field (ZF).

III. RESULTS AND DISCUSSION

A. YBaMn$_2$O$_6$

We show three examples of the ZF-μSR time spectra of YBaMn$_2$O$_6$ above and below $T_N = 195$ K in Fig. 2. A spontaneous Larmor precession is observed in the data below $T_N$, characteristic of a static magnetically ordered phase. The Fourier transform FFT of the time spectra below $T_N$ are shown...
in Fig. 3, where four well-resolved frequencies are evident at low temperatures. The time spectra above $T_N$ were fitted using $G_z(t) = A \exp(-\lambda t)$, where $A$ and $\lambda$ are the initial total asymmetry and the relaxation rate, respectively. Below $T_N$, the functional form of the ZF-$\mu$SR time spectra includes oscillating terms

$$G_z(t) = A_1 \exp(-\lambda_1 t) + \sum_i A_{2,i} \exp(-\lambda_{2,i} t) \cos(2\pi v_i t + \phi),$$

(1)

describing the effect of the static internal fields. $A_{2,i}$ ($\sum_i A_{2,i} = A - A_1$) and $v_i$ are the $i$th oscillating components of amplitude and frequency, respectively. $\phi$ is the initial phase, the same for all the oscillating terms. The associated relaxation rates, longitudinal and transversal, are $\lambda_1$ and $\lambda_{2,i}$, respectively. The red solid lines in Fig. 2 represent the best fits to the data by using Eq. (1).

The parameters (a) $A$, $A_1$, (b) $\lambda$ and $\lambda_1$, and (c) $v_i$ obtained for various temperatures are summarized in Fig. 4, where $A$ and $\lambda$ ($A_1$ and $\lambda_1$) correspond to $T > T_N$ ($T < T_N$). A slight but clear hysteretic behavior in these parameters for the data measured on cooling (filled symbols) and on heating (open symbols) reveals first-order features of the antiferromagnetic phase transition. The situation, however, is complicated because the transition also reveals critical fluctuations [see Fig. 4(b)] which are normally associated with second-order type phase transitions. All this points to a weak first-order type phase transition.24 We note that the first-order features may be related to a possible structural transition. The antiferromagnetic order is accompanied by a change in the COO superstructure.13

The initial total asymmetry $A \simeq 0.22$ has a very weak temperature dependence above $T_N$. $A_1$, representing the signal from muons polarized along the direction of the internal field, decreases due to the antiferromagnetic spin order just below $T_N$. We define the transition temperature as $T_N = 183$ and 173 K for the warming and cooling process, respectively, from the inflection point of $A_1$. The ratio $A_1/A \simeq 1/3$ is expected if all the implanted muons are exposed to static local fields in a polycrystalline sample. $A_1/A$ is, however, slightly larger than 1/3 below $T_N$ and reaches 1/3 only below 50 K. This
result suggests that a very small fraction of the sample, most probably an minority phase \( \text{BaMnO}_3 \) or \( \text{Y}_{0.5}\text{Ba}_{0.5}\text{MnO}_3 \), orders magnetically below \( \sim 50 \) K. Therefore, the muon spin oscillation with \( \nu_1 = 101.1 \) MHz [see Figs. 3 and 4(c)], that appears only below 50 K, is considered to be extrinsic. The existence of a minority phase is also indicated by an anomaly in \( \chi(T) \) at the same temperature.\(^{25}\) The broad peak in the relaxation rate just below \( T_N \) may be due to a critical slowing down of Mn magnetic moments accompanying the antiferromagnetic order.

Below \( T_N \) one detects in the \( \mu \)SR time-spectra oscillations with frequencies \( \nu_1 = 20.2 \) MHz, \( \nu_2 = 24.8 \) MHz, and \( \nu_3 = 88.9 \) MHz. They reveal quasi-static internal magnetic fields of \( B = (2\pi/\gamma)\nu \), where \( \gamma = 2\pi \times 135.5 \) MHz/T is the muon gyromagnetic ratio and \( B \) is a local static magnetic field at muon site. The inferred local fields are 0.15, 0.18, and 0.66 T at 2.5 K, respectively. The high values of the frequencies just below \( T_N \), \( \nu_1/T_N/\nu_1(0) \approx 0.6 \) reveal first-order type features of the magnetic transition.

The observation of three precession frequencies indicates multiple magnetically inequivalent stopping sites for the muon. Considering that dipolar fields dominate the local field at the muon site in \( \text{LaMnO}_3 \),\(^{26,27}\) we estimated the fields for possible muon sites about 1 Å away from oxygen ions and in the large empty space in the \( \text{MnO}_2 \) plane, and adopting the crystalline and the magnetic structure with the CE-type ordering in the \( \text{MnO}_2 \) plane with the \( \alpha\alpha\beta\beta \)-type stacking along the \( c \) axis.\(^{6,13}\) We propose a likely muon site at \((0.8,0.96,0.5)\), about 1.2 Å away from the oxygen positions in the BaO plane, where the estimated dipolar fields agree roughly with the observed local fields. The position of the muon site is indicated by blue stars in the inset of Fig. 4(c), where all the plotted stars are crystallographically equivalent. It is close to the muon site for \( \text{LaMnO}_3 \),\(^{26,27}\) but not exactly the same. One may not expect to have exactly the same muon sites, because the O-ion displacements from their ideal positions are much smaller in \( \text{YBaMn}_2\text{O}_6 \) than those in \( \text{LaMnO}_3 \). In addition, the spatial distribution of electrostatic potential are expected to be different from each other, because \( \text{Mn}^{3+} \) and \( \text{Mn}^{4+} \) have the checkerboard-type ordering in the \( \text{MnO}_2 \) plane of \( \text{YBaMn}_2\text{O}_6 \), while the valence of Mn ion is uniform for \( \text{LaMnO}_3 \). We stress that not only the magnitude of the estimated fields of 0.208, 0.232, 0.235, and 0.580 T at the muon sites agree with the experimental results but also the relative intensities are consistent with the observed intensity in the Fourier spectra (see Fig. 3). In particular, the intensity at \( \nu_2 \) is twice as large as those at \( \nu_1 \) and \( \nu_3 \). A detailed discussion on the muon site derivation is given in the Appendix.

In the case of \( \text{LaMnO}_3 \), the transferred hyperfine field vanishes by symmetry considerations of the \( A \)-type magnetic structure (ferromagnetic planes are stacked antiferromagnetically along the \( c \) axis) with a tiny canting.\(^{26}\) For \( \text{YBaMn}_2\text{O}_6 \), symmetry arguments do not imply a vanishing transferred hyperfine field in the BaO plane. Nevertheless, the estimated field at the muon site \((0.8,0.96,0.5)\) is consistent with the experimental results and the site is close to that in \( \text{LaMnO}_3 \). We conclude, therefore, that the dipolar contribution to the local field is dominant in \( \text{YBaMn}_2\text{O}_6 \). The transferred hyperfine field may account for the small difference between the estimated dipolar fields and experimentally observed values.
lower than $T_C$. We note, however, that this scenario has a difficulty. Namely, here the transition region is very broad and in such a case it is not clear that one can detect critical slowing down. Therefore, one should consider a different scenario. Namely, between 250 and 300 K part of the fast relaxing (at low temperatures) component may be in fact observed in the $\mu$SR time signal. The inferred relaxation rate then may be the result of an interplay between the increase in the fast (transversal) relaxation rate and the gradual decrease in its weight. The very broad signal centered around 125 K, ranging from 50 to 200 K, may be due to the change in a magnetic structure associated with the appearance of AFI(CE) phase at 2.5 K as follows. The sum of oscillating $\nu_1$ and $\nu_2$ being approximately temperature independent and still high at 125 K. This indicates that the antiferromagnetic spin structure arises from the ferromagnetic phase through a first-order type transition (spin re-orientation). This result is consistent with the temperature independent $^{55}$Mn-NMR frequency in the AFI(CE) phase.

One may roughly estimate the volume fraction of the AFI(CE) phase at 2.5 K as follows. The sum of oscillating
amplitudes $A_{2.1} + A_{2.2} = 0.041$ may be considered to be $2/3$ of the initial asymmetry from muons in the AFI(CE) phase. Thus, the volume fraction of the AFI(CE) phase is estimated to be $(3/2)(A_{2.1} + A_{2.2})/A \approx 30\%$, where $A \approx 0.22$ is the initial asymmetry at high temperature above $T_C$. This estimated value is in good agreement with the value of 30% obtained from the magnetization curve.21

From considerations of possible muon sites, we concluded that most likely the muons stop at the position $(0.755, 1/2, 1/2)$ for LaBaMn$_2$O$_6$, see Appendix for details. As explained in the Appendix, for this site we estimated the dipolar field assuming a uniform ferromagnetic state, and obtained 0.38 T corresponding to 51.5 MHz. This is clearly within our range of detection. But the fact that no muon spin oscillations are observed, adds support to the claim that the inhomogeneity of the FM phase is in fact intrinsic (as opposed to the scenario where well defined $\mu$SR frequencies are simply too high to be observed).

Finally, we present an alternative approach for the analysis of the data between 150 K < $T$ < $T_C$. The two-exponential model takes into account a two-phase percolation proposed for ferromagnetic manganites below $T_C$.30 It leaves out some of the subtleties, but it is simple to understand the meaning of the fitting parameters. Here the time-spectrum data is expressed by

$$G_x(t) = A_f \exp(-\lambda_f t) + A_s \exp(-\lambda_s t),$$

where $\lambda_f$ and $\lambda_s$ are the relaxation rates for two exponentials, respectively. $A_f$ and $A_s$ are the corresponding $\mu$SR amplitudes. Here $A = A_f + A_s$ and $\lambda_f \ll \lambda_s$. The smaller value of the relaxation rate $\lambda_s$ indicates the faster fluctuation rate of local field at the muon site.

In Fig. 9 we show the temperature dependence of fractional amplitude of $A_s$, that is, (a) $A_f/A$, (b) $\lambda_f$, and (c) $\lambda_s$ in the two-exponential model for the FM phase of LaBaMn$_2$O$_6$.

relaxation rates $\lambda_f$ and $\lambda_s$ become larger upon cooling down to 150 K.

The relaxation rate $\lambda_s$ is one order of magnitude larger than $\lambda_f$. Similar results in the two-exponential model have been reported for the ferromagnetic conductor La$_{0.62}$Ca$_{0.38}$MnO$_3$ and the insulating ferromagnet La$_{0.82}$Ca$_{0.18}$MnO$_3$.27,30 Heffner et al. associated the relaxations of $\lambda_f$ component with overdamped spin waves, characteristic of a disordered ferromagnetic metal, and that of $\lambda_s$ component with relatively insulating regions of the sample. The damping of spin waves may be caused by an inherent disorder due to the random replacement of La atoms by Ca. In our case there is no such a random replacement, but similar mechanism may be also responsible for the observed relaxation of $\lambda_f$ component due to possible random arrangements of the layers as discussed above.

In the ferromagnetic conductor La$_{0.62}$Ca$_{0.38}$MnO$_3$ $A_s$ decreases ($A_f$ increases) upon cooling below $T_C$, while $A_s$ is almost independent of temperature below about 170 K for the insulating ferromagnet La$_{0.82}$Ca$_{0.18}$MnO$_3$ that shows a transition to a CO state below around 60 K.27,30 The temperature independent $A_s$ below around 250 K for LaBaMn$_2$O$_6$, which is qualitatively similar to that for La$_{0.82}$Ca$_{0.18}$MnO$_3$, indicates that the relatively insulating region survives even in the FM phase. The weak metallic nature of the FM phase in LaBaMn$_2$O$_6$ may be consistent with the appearance of AFI(CE) phase inside the FM phase at low temperature.

IV. CONCLUSION

In conclusion, our zero-field $\mu$SR data reveal striking differences in the magnetically ordered phases of YBaMn$_2$O$_6$ and LaBaMn$_2$O$_6$. For the former, a homogeneous state is observed, as expected for an A-site ordered manganite. For the latter, unexpectedly one finds inhomogeneous magnetic phases below room temperature.
It is not clear what triggers these important differences in our two materials. On the other hand, the A-site order triggers important changes in the thermal and transport properties of the Y compound, but not in the La compound. The inhomogeneous magnetic phase of the La compound is reminiscent of the findings in \( (La_{1-x}Pr_x)_2Sr_{1.8}Mn_2O_7 \), where an inhomogeneous magnetic phase is thought to be triggered by peculiar stacking faults leading to intergrowth of different magnetic layers. But this mechanism does not seem to apply for the Y compound.

**APPENDIX**

We consider a possible muon site in \( \text{YBaMn}_2\text{O}_6 \) on the basis of previous works in \( \text{LaMnO}_3 \). The crystal structure of \( \text{LaMnO}_3 \) has an orthorhombic cell with space group \( \text{Pnmb} \), where the Mn ions are located at the \((0,0.5,0)\) positions. Guidi \textit{et al.} have proposed two muon sites \((0.389,0.937,0.25)\) and \((0.5,0.5,0)\) for \( \text{LaMnO}_3 \). The first site, close to the Holzschuh site for \( \text{YbF}_2 \), is about 1.1 Å distant from an apical oxygen and farthest away from positive La ions in the LaO plane. The second site is located in the large empty space at the center of the MnO\(_2\) plane. Independently, Heffner \textit{et al.} have reported similar muon sites.

The crystal structure of \( \text{YBaMn}_2\text{O}_6 \) has a monoclinic cell with space group \( \text{P3} \). There are two Mn sites, trivalent Mn\((1)\) at \((0.018,0.005,0.242)\) and tetravalent Mn\((2)\) at \((0.481,0.518,0.244)\), and eight oxygen sites, \( \text{O}(1)\)–\( \text{O}(8) \) in the unit cell at 350 K. Muons bound to apical oxygens \( \text{O}(1) \) and \( \text{O}(2) \) in the \( \text{YO} \) plane and \( \text{O}(7) \) and \( \text{O}(8) \) in the \( \text{BaO} \) plane of \( \text{YBaMn}_2\text{O}_6 \) may be closely related to the first muon site in \( \text{LaMnO}_3 \), while the valence of Mn ion is uniform for \( \text{LaMnO}_3 \). For this muon site we find four magnetically inequivalent muon sites with the dipolar fields of 0.208, 0.232, 0.235, and 0.580 T along the \( b \) axis. The multiple magnetically inequivalent sites arise from the CE-type magnetic structure in the \( \text{MnO}_2 \) plane with the large magnetic unit cell. The transferred hyperfine field may account for the small difference between the estimated dipolar fields and the experiments as mentioned in Sec. III A. Our estimates are also consistent with the observed intensity in the Fourier transformed spectrum where intensity at \( v_2 \) is twice as large as those at \( v_1 \) and \( v_3 \).

Regarding our chosen muon site, it seems very likely that positive muons stop in the \( \text{BaO} \) plane for two reasons:

1. \( \text{Ba}^{2+} \) has smaller positive charge than \( \text{Y}^{3+} \), and
2. the average distance between \( \text{BaO} \) and \( \text{MnO}_2 \) planes is larger by 25% than that between \( \text{YO} \) and \( \text{MnO}_2 \) planes due to the distortion of \( \text{MnO}_6 \) octahedra. In addition, muons may favor a position near \( \text{O}(7) \) rather than a position near \( \text{O}(8) \) in the \( \text{BaO} \) plane, because the nearest Mn ion to \( \text{O}(7) \) is trivalent Mn\((1)\), while that to \( \text{O}(8) \) is tetravalent Mn\((2)\).

Regarding \( \text{LaBaMn}_2\text{O}_6 \), it seems reasonable to assume its muon site to be the position related to the Holzschuh site or its symmetric replica for orthoferrites. In the tetragonal cell of \( \text{LaBaMn}_2\text{O}_6 \), there are three oxygen sites: \( \text{O}(1) \) at \((1/2,1/2,0)\) in the \( \text{LaO} \) plane, \( \text{O}(2) \) at \((1/2,0,0.2373)\) in the \( \text{MnO}_2 \) plane, and \( \text{O}(3) \) at \((1/2,1/2,1/2)\) in the \( \text{BaO} \) plane at 400 K. There are no \( \text{O}^- \) deviations from the ideal positions in the \( \text{BaO} \) plane of \( \text{LaBaMn}_2\text{O}_6 \). We propose that the muon lies at \((0.755,1/2,1/2)\) in the \( \text{BaO} \) plane, 1 Å away from \( \text{O}(3) \) towards the midpoint between two adjacent Ba ions. We estimated the dipolar sums over a large spherical domain centered at the proposed muon site in the FM phase, assuming a uniform ferromagnetic state. In the estimation we used the structural parameters at 400 K and the Mn magnetic moment 3.0 \( \mu_B \) from the magnetization in 5 T, where the AFI(CE) phase is suppressed by the magnetic field. The estimated dipolar field is 0.38 T, corresponding to the muon frequency of 51.5 MHz.

---

1. \( \text{yu@pm.tokushima-u.ac.jp} \)
2. Present address: Seiryo Engineering Co., Ltd.
3. Present address: Nichia Corporation.