μSR study of A-site ordered perovskite manganite LaBaMn$_2$O$_6$

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Abstract. We have investigated magnetic properties of the A-site ordered perovskite manganite LaBaMn$_2$O$_6$ by muon spin relaxation. In the ferromagnetic metal (FM) phase below $T_C \sim 330$ K, no muon spin oscillation is observed in the time spectra, where the muon spin relaxation significantly deviates from a single exponential form. These results may suggest a large spatial distribution of spin dynamics, i.e. the inhomogeneous magnetic nature in the FM phase of LaBaMn$_2$O$_6$, which has been also observed in the FM phase of A-site disordered perovskite manganites. It is, therefore, suggested that there exists the spatial distribution of spin dynamics in the FM phase of the perovskite manganites irrespective of the order/disorder at the A-site of perovskite structure.

1. Introduction

In these several decades, the perovskite manganites $R_{1-x}A_xMnO_3$ ($R$ = rare earth element, $A$ = Ca, Ba, Sr) have been investigated intensively, because they show a rich variety of fascinating electromagnetic properties, such as a colossal magnetoresistance (CMR), charge and orbital ordering (CO) and metal-insulator transition. Recently, the A-site ordered perovskite manganites $R$BaMn$_2$O$_6$ have been synthesized in half-doped manganites ($x = 0.5$). The crystal structure of $R$BaMn$_2$O$_6$, shown in the inset of Fig. 1, has a layer-type ordering of $R$ and Ba ions along the $c$-axis, resulting in the two dimensional structure with the stacking sequence of -RO-MnO$_2$-BaO-MnO$_2$- planes. Interestingly, the order of $R$ and Ba elements at the A site of a perovskite structure dramatically changes their physical properties from the A-site disordered manganites, $R_{0.5}$Ba$_{0.5}$MnO$_3$, with the same chemical composition [1, 2, 3, 4, 5, 6].

One of the features peculiar to the A-site ordered $R$BaMn$_2$O$_6$ is their much higher CO and magnetic ordering temperatures compared with the A-site disordered manganites. For example, the CO transition temperature of YBaMn$_2$O$_6$ is as high as 500 K, while $Y_{0.5}$Ba$_{0.5}$MnO$_3$ has only a spin glass phase below 30 K. The increase in the ordering temperature is considered to
originates mainly from the ordering of $R$ and Ba in the structure, which results in the absence of electrostatic potential disorder. The very high ordering temperature in $R$BaMn$_2$O$_6$ is considered to be a key feature for the development of new materials that exhibit CMR at room temperature [7, 8].

In this report, we focus on another A-site ordered perovskite manganite, LaBaMn$_2$O$_6$. This compound shows a transition from a paramagnetic metal (PM) to a ferromagnetic metal (FM) phase around $T_C \sim 330$ K. The transition temperature is enhanced from 280 K in La$_{0.5}$Ba$_{0.5}$MnO$_3$. Noticeably, it is found by the neutron diffraction measurements that a part of the FM phase transforms to antiferromagnetic insulating (AFI(CE)) phase below $\sim 200$ K and that the AFI(CE) phase coexists with the FM phase at the ground state [3]. The NMR study has revealed from the Mn nuclear spin-spin relaxation rate that the magnetic properties of the FM phase in LaBaMn$_2$O$_6$ is the same type as that of the disordered form La$_{0.5}$Ba$_{0.5}$MnO$_3$ [6]. It seems that the magnetic properties of the FM phase are not affected by the absence of disorder at the A site in LaBaMn$_2$O$_6$, although the transition temperature is enhanced. In order to get further information on magnetic properties of the FM phase, we have performed $\mu$SR experiments on LaBaMn$_2$O$_6$ in zero field.

2. Experimental

Powder sample of LaBaMn$_2$O$_6$ for the present $\mu$SR measurements is the same one used in the previous NMR experiments [6]. This sample was prepared by a solid-state reaction and subsequent sintering process in pure Ar gas, followed by annealing in flowing O$_2$ gas. The details of the preparation method are described in the literature [2]. The crystal structure and magnetic properties of this sample are confirmed to be consistent with the conclusions based on the previous data [1, 2, 3, 4, 5, 6]. From the Mn-NMR spectra, majority of the sample is confirmed to be A-site ordered, although it contains disordered La$_{0.5}$Ba$_{0.5}$MnO$_3$ with several percent [6]. The $\mu$SR data presented here have been taken at the M20 muon channel at TRIUMF (Vancouver, Canada) and on $\pi$A muon channel at KEK (Tsukuba, Japan). All the $\mu$SR data have been measured in zero field.

3. Results and Discussion

We show the $\mu$SR time spectra measured at 324 K, 300 K and 240 K in Fig.1. In the FM phase below $T_C \sim 330$ K, no clear muon spin oscillation is observed in the time spectra, where the time spectra significantly deviates from a single exponential form. The time spectra data have been
fitted by using the stretched exponential model expressed as

\[ G_z(t) = A \exp(-\lambda t)^n, \]

where \( A \) is the initial asymmetry and \( \lambda \) is the longitudinal muon spin relaxation rate and \( n \) is the stretched exponent. \( n = 1 \) means a single exponential relaxation, while smaller values, \( n < 1 \), suggests a distribution of relaxation rates. The best fits are represented by the red solid lines in Fig. 1.

The parameters \( A, \lambda \) and \( n \) obtained at various temperatures are summarized in Figs. 2 (a), (b) and (c), respectively. \( A \approx 0.22 \) has very weak temperature dependence in the PM phase above \( T_C \). With decreasing temperature, \( A \) starts to decrease at \( T_C \) and reaches about 1/3 of the value in the PM phase. The value of 1/3 is expected if all implanted muons are exposed to a static local field in a polycrystalline or powder sample, \( i.e., \) the volume fraction of magnetic phase is almost 100%.

In the temperature dependence of \( \lambda \), two broad peaks are observed at 280 K and 125 K. The relaxation peak at 280 K, close to the inflection point of \( A \), is considered to be due to a critical slowing down of Mn magnetic moments accompanied by the ferromagnetic order. The very broad peak around 125 K, ranging from 50 K to 200 K, suggests a crossover-like change in spin dynamics. It may be due to the change in magnetic structure from the FM one to the AFI(CE) one in the part of sample [3, 6]. Actually, oscillating components with frequency of 69.6 MHz and 31.6 MHz, which may originate from muons in the AFI(CE) phase, appear in the muon time spectra below 160 K (not shown).

The muon relaxation curve changes from an exponential (\( n = 1 \)) in the PM phase to a root exponential (\( n = 1/2 \)) in the FM phase. The small value of \( n \) may suggest a spatial distribution of spin dynamics in the FM phase, that has been observed in the A-site disordered ferromagnetic manganites La\(_{1-x}\)Ca\(_x\)MnO\(_3\) (\( x = 0.18, 0.33 \) and 0.375) and La\(_{0.85}\)Sr\(_{0.15}\)MnO\(_3\) [9, 10, 11, 12]. This result may suggest that there exists spatially inhomogeneous spin dynamics even in the FM phase of LaBaMn\(_2\)O\(_6\), although the disorder at the A-site is largely diminished in this material. The further decrease in \( n \) below 160 K may suggest an increase in distribution of spin dynamics, most probably reflecting the appearance of the AFI(CE) phase inside the FM one.

Finally, we discuss on a possible muon site in LaBaMn\(_2\)O\(_6\). It is reasonable to assume that the muon site in LaBaMn\(_2\)O\(_6\) is the position corresponding to that in YBaMn\(_2\)O\(_6\), \( i.e., \) the position
1 Å away from oxygen in the BaO plane [13]. In the tetragonal unit cell of LaBaMn$_2$O$_6$, there are three oxygen sites; O(1) at (1/2, 1/2, 0) in the LaO plane, O(2) at (1/2, 0, 0.2373(5)) in the MnO$_2$ plane and O(3) at (1/2, 1/2, 1/2) in the BaO plane at 400 K [3]. One may, therefore, consider the muon site to be at (0.755, 1/2, 1/2) in the BaO plane, indicated by open star in the inset of Fig. 1. We have calculated the magnitudes of dipolar field at the muon site in the FM phase. Here, we used the structural parameters listed in Table I of the literature [3] and the size of Mn magnetic moment 3.0 $\mu_B$ estimated from magnetization in 5 T [3]. An easy axis is assumed to be [110], corresponding to the direction of Mn moment in the sublattice of the AFI(CE) phase of YBaMn$_2$O$_6$. The calculation gives the dipolar field of 0.38 T. The corresponding muon frequency of 51.5 MHz is sufficiently observable in the resolution of our experimental condition, ruling out possibility that muon frequency is too high to be detected in the FM phase.

4. Conclusions
Magnetic properties of the A-site ordered perovskite manganite LaBaMn$_2$O$_6$ have been investigated by $\mu$SR experiments in zero field. In the FM phase below 330 K, no clear muon spin oscillation has been observed in the time spectra, while the muon relaxation significantly deviates from a single exponential form. These results may suggest a spatial distribution of spin dynamics, which has been observed in the FM phase of A-site disordered ferromagnetic manganites, although the disorder at the A-site is largely diminished in LaBaMn$_2$O$_6$. With decreasing temperature, oscillating components appear in the muon time spectra below 160 K. The appearance of the oscillating components may be due to the existence of the antiferromagnetic insulating phase inside the FM one.

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