


(※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。)

 <b>MLF Experimental Report</b>	提出日 Date of Report November 20, 2016
課題番号 Project No. 2015A0055  実験課題名 Title of experiment: Challenge for detecting oxygen diffusion in solids with $\mu$ SR  実験責任者名 Name of principal investigator: Jun Sugiyama  所属 Affiliation: Toyota Central Research & Development Laboratories, Inc.	装置責任者 Name of responsible person Prof. Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D1 実施日 Date of Experiment May 26-27, 2016

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)  
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.
Powder samples of 1) $\text{La}_{1.54}\text{Sr}_{0.46}\text{Ga}_{2.98}\text{Mg}_{0.02}\text{O}_{7+d}$ 2) $\text{La}_{9.333}\text{Si}_6\text{O}_{26}$

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。)
<p>Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.</p> <p>Powder samples of <math>\text{La}_{1.54}\text{Sr}_{0.46}\text{Ga}_{2.98}\text{Mg}_{0.02}\text{O}_{7+d}</math> [1] and <math>\text{La}_{9.333}\text{Si}_6\text{O}_{26}</math> [2] were prepared by a conventional solid-state reaction technique. Then, each powder sample (about 1 g) was packed into a titanium cell sealed with a gold O-ring. The window of the cell was also Ti foil with 25 <math>\mu\text{m}</math> thickness.</p> <p>Figure 1 shows the crystal structure of melilite-type <math>\text{La}_{1.54}\text{Sr}_{0.46}\text{Ga}_{2.98}\text{Mg}_{0.02}\text{O}_{7+d}</math> (Me-24), which exhibits relatively high O ionic conductivity at high temperatures.</p> <p>Figure 2 shows ZF-spectra for Me-24 obtained at 303, 573, and 873 K. This clearly shows a dynamic behavior at high temperatures. Then, we fitted the ZF- and LF-<math>\mu</math>SR spectra with a dynamic Kubo-Toyabe function. Figure 3 shows the temperature dependences of <math>\Delta</math> and <math>\nu</math>. <math>\Delta</math> is found to be temperature-independent up to 200 <math>^{\circ}\text{C}</math> and starts to decrease with temperature above 200 <math>^{\circ}\text{C}</math> [Fig. 3(a)]. On the other hand, <math>\nu</math> gradually increases with temperature above room temperature [Fig. 3(b)].</p>

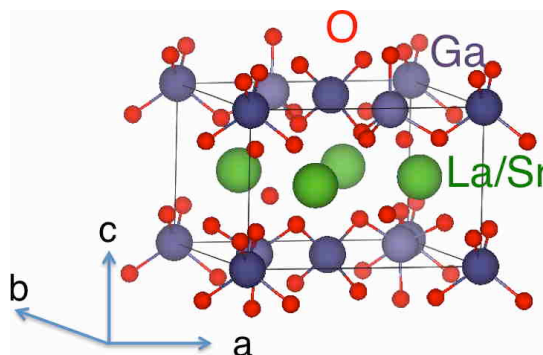


Fig. 1 Crystal structures of melilite-type  $\text{La}_{1.54}\text{Sr}_{0.46}\text{Ga}_3\text{O}_7$ .

Since the full asymmetry was observed even at room temperature, implanted  $\mu^+$  s naturally locate at the vicinity of  $O^{2-}$  so as to make a stable O- $\mu$  bond. This implies that the changes in  $\Delta$  and  $\nu$  are caused by  $O^{2-}$ -diffusion.

In order to know the origin of the temperature dependence of  $\nu$ , Fig. 4 shows the relationship between  $\nu$  and  $1/T$  and between ionic conductivity ( $\sigma_0$ ) and  $1/T$ . It is found that a remarkable increase in  $\nu$  with temperature occurs at roughly the same temperature range, at which  $\sigma_0$  starts to increase with temperature, i.e.  $1/T < 1.5$ . However, in order to confirm the possibility that  $\mu^+$  SR detects oxygen diffusion, we definitely need to obtain more reliable data for Me-24 and to add the data for the other good O-ion conductors.

[1] X. Kuang et al., Nature Mater. **7**, 498 (2008).

[2] K. Fukuda et al., Chem. Mater. **25**, 2154 (2013).

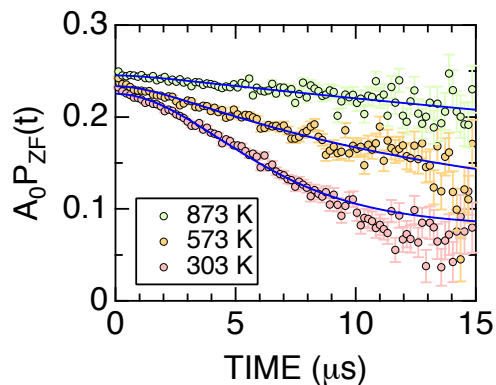


Fig. 2 ZF-spectra for Me24 obtained at 303, 573, and 873 K.

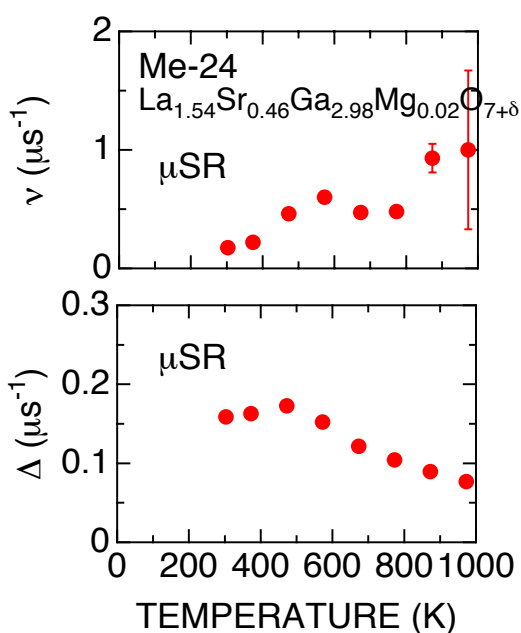


Fig. 3 The temperature dependences of  $\nu$  and  $\Delta$  for Me-24.

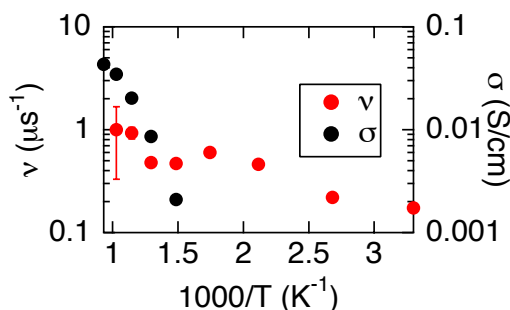


Fig. 4 The relationship between  $\nu$  and  $1/T$  and between ionic conductivity ( $\sigma_0$ ) and  $1/T$  for Me-24.