Magnetic moments and ordered states in pyrochlore iridates $Nd_2Ir_2O_7$ and $Sm_2Ir_2O_7$ studied by muon-spin relaxation†

R. Asih,[*1,*2] N. Adam,[*1,*3] S. S. Mohd-Tajudin,[*1,*3] D. P. Sari,[*1,*2] K. Matsuhira,[*4] H. Guo,[*5] M. Wakeshima,[*6] Y. Hinatsu,[*6] T. Nakano,[*2] Y. Nozue,[*2] S. Sulaiman,[*3] M. I. Mohammed-Ibrahim,[*3] P. K. Biswas,[*7] and I. Watanabe[*1,*2,*3,*8]

Pyrochlore iridates, $R_2Ir_2O_7$ (R227, R = Nd, Sm, Eu, Gd, Tb, Dy, or Ho), which have the relatively large spin-orbit coupling (SOC) inherent in Ir 5d electrons and a d-f exchange interaction, have been suggested to show peculiar electronic properties.[1)] R227 also shows metal-insulator transitions (MITs) at $T_{\rm MI}$, which seems to be accompanied by magnetic transitions. $T_{\rm MI}$ gradually decreases from 141 K for R = Ho to 117 K for R = Sm with the increasing ionic radius of the trivalent R ion. $T_{\rm MI}$ suddenly drops to 33 K for Nd227, and no MIT is observed in Pr227, which shows metallic behavior.[2)] Owing to difficulties in observing Ir magnetic ordering by means of neutron studies in these compounds, muon-spin relaxation (μSR) studies have been proven to directly confirm the appearance of long-range magnetic ordering (LRO) in some of the R227 compounds.[3–5)]

In this study, we investigate the magnetic orderings and structures of Nd227 and Sm227, which are particularly important as they lie in the boundary of MIT. In the case of Nd227, we observed the additional LRO of Nd moments below 10 K and found a saturated internal field at the muon site ($H_{\rm int}$) of approximately 530 G at 1.5 K, which confirmed indications suggested by previous neutron scattering[6)] and μSR studies.[5)] In the case of Sm227, spontaneous muon-spin precession was observed below $T_{\rm MI}$ = 117 K, which indicated the appearance of LRO of Ir moments below $T_{\rm MI}$. The parameters obtained from the fitting to the zero-field μSR data on Sm227 are shown in Fig. 1. The solid line in Fig. 1(a) indicates the temperature dependence of the resistivity, showing a clear transition at $T_{\rm MI}$. The internal field at the muon site, $H_{\rm int}$, rapidly increases just below $T_{\rm MI}$ and saturates at the temperature region between 60 K and 20 K. With further decrease in temperature, $H_{\rm int}$ decreases below approximately 10 K. As shown in Fig. 1(b), the relaxation rate λ_1 is relatively constant in the paramagnetic region at $T > T_{\rm MI}$ and then increases below $T_{\rm MI}$, forming a peak at approximately 10 K, which indicates a slowing-down behavior toward an LRO below this temperature. This, therefore, suggests the appearance of the additional LRO of Sm moments below 10 K. Increases in the damping rate of the muon-spin precession are also observed below 10 K and near the $T_{\rm MI}$, as indicated in Fig. 1(c), which further indicates a broadening of the distribution of $H_{\rm int}$. Dipole-field calculations at the possible muon stopping site show that the all-in all-out spin structure most convincingly explained the present μSR results with the lower limits of the magnetic-ordered moments determined as 0.12 μ_B/Ir^{4+} and 0.2 μ_B/Nd^{3+} in Nd227 and 0.3 μ_B/Ir^{4+} and 0.1 μ_B/Sm^{3+} in Sm227. Further analysis indicated that the spin coupling between Ir and Nd/Sm moments was ferromagnetic for Nd227 and antiferromagnetic for Sm227.

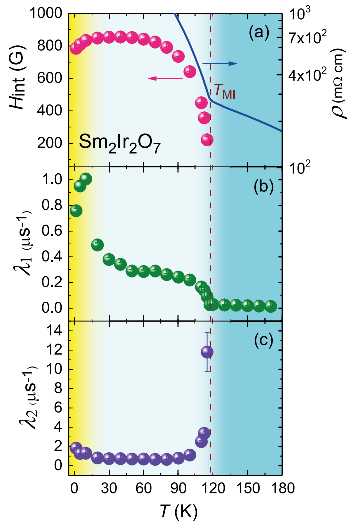

Fig. 1. Temperature dependences of (a) internal field at the muon sites $H_{\rm int}$, (b) slow relaxation rate λ_1, and (c) damping rate of the muon-spin precession λ_2. The broken line indicates $T_{\rm MI}$ = 117 K. The area $T > T_{\rm MI}$ indicates the paramagnetic region. The temperature dependence of the resistivity of $Sm_2Ir_2O_7$ is displayed in (a) by the solid line as a reference MIT.

† Condensed from the article in J. Phys. Soc. Jpn. **86**, 024705 (2017)

*1 RIKEN Nishina Center
*2 Department of Physics, Osaka University
*3 School of Distance Education, Universiti Sains Malaysia
*4 Graduate School of Engineering, Kyushu Institute of Technology
*5 Department of Physics, Zhejiang University
*6 Department of Chemistry, Hokkaido University
*7 Bulk μSR Group, Paul Scherrer Institut
*8 Department of Condense Matter Physics, Hokkaido University

References

1) X. Wan *et al.*, Phys. Rev. B **83**, 205101 (2011).
2) K. Matsuhira *et al.*, J. Phys. Soc. Jpn. **80**, 094701 (2011).
3) S. Zhao *et al.*, Phys. Rev. B **84**, 180402(R) (2011).
4) S. M. Disseler *et al.*, Phys. Rev. B **84** 174441 (2012).
5) H. Guo *et al.*, Phys. Rev. B **88**, 060411(R) (2013).
6) K. Tomiyasu *et al.*, J. Phys. Soc. Jpn. **81**, 034709 (2012).