Spin fragmentation in $Nd_2Ir_2O_7$ and its carrier-doped dependence

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Pyrochlore iridates, $R_2Ir_2O_7$ (R227, R is a rare-earth element), provide an ideal platform of strongly frustrated systems to study the interplay between electron-electron correlation (U) and spin-orbit interaction (SOI) given from $5d$ electrons of Ir^{4+}.[1] Among R227, Nd227 stands out as a fascinating system because of the additional interesting properties. Nd227 exhibits metallic behavior and undergoes metal-insulator transition (MIT) at $T_{\rm MI}=$ 33 K.[2] μSR and neutron-scattering studies on Nd227 showed the appearance of a long-range magnetic ordering (LRO) of Ir^{4+} moments below $T_{\rm MI}$ followed by an additional LRO of Nd^{3+} moments below 10 K.[3–5] With such progressive reports, however, the sizes of the magnetic ordered moments remain debatable. A reduction in the Nd^{3+} moments was found in the recent neutron study[6] compared with those estimated from the previous study[3] and crystal electric field (CEF) analysis,[7] which is argued to be attributed to a strong quantum fluctuation. This argument was also indicated from μSR results on Nd227, which show an appreciable reduction on the internal field at the muon site ($H_{\rm int}$) compared with other R227 compounds.[4,5] These results signify a possible magnetic fragmentation in Nd227, where ordered and fluctuating phases occur simultaneously. The onset of magnetic ordering on Nd227 was also reported to be suppressed by hole doping via Ca^{2+} substitution on the Nd^{3+} site, and $T_{\rm MI}$ was found to gradually decrease by increasing the Ca concentration.[8] In this study, we investigated the existence of magnetic fragmentation in Nd227 and Ca-doped Nd227, $(Nd_{1-x}Ca_x)_2Ir_2O_7$.

Longitudinal-field (LF-μSR) measurements were performed to confirm the emergence of fluctuations in the ordered phase of the compounds. Figure 1 displays the temperature dependence of the relaxation rate λ of $(Nd_{1-x}Ca_x)_2Ir_2O_7$ for $x = 0.00$, 0.05, 0.07 and 0.10 under an applied field of 3.6 T. An appreciable peak in λ was observed at higher temperatures compared with ordered and meta-transition temperatures. For $x = 0.07$ and 0.10, a clear peak was observed at about 20 K despite the fact that neither muon-spin precession nor a slowing-down behavior was observed under the zero-field (ZF) condition at this temperature. These results may indicate that Nd and/or Ir have low-lying spin fluctuations, which can be easily changed by temperatures and magnetic fields. Figure 2 shows the field dependence of the relaxation rate measured in Nd227 at 50 K (paramagnetic state), 15 K (ordered state of Ir moments), and 1.5 K (ordered state of Ir and Nd moments). λ increases with an increase in the applied field at 50 K and shows a peak around 2.5 T at 15 K, which signifies that the spectrum density of the spin fluctuations shifts down to the lower frequency side with decreasing temperature followed by the change in the dynamics of spins. At 1.5 K, λ remains about 0.6 μs^{-1} in the ZF condition, and then, it decreases exponentially with an increasing field showing Redfield-like behavior, which indicates the maintenance of the spin fluctuation even in the ordered state, *i.e.*, a magnetic-fragmentation is realized in Nd227. To further discuss the scheme of spin-fluctuations in these compounds, it is necessary to collect more data points at different temperatures and applied fields.

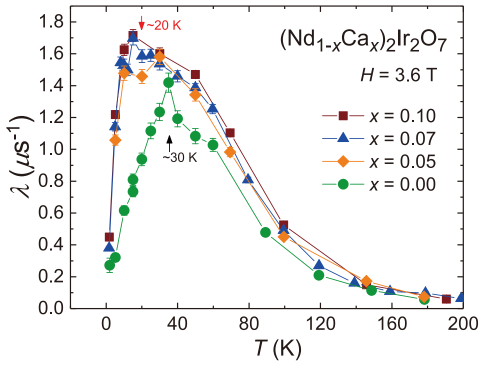

Fig. 1. Temperature dependence of the relaxation rate λ of $(Nd_{1-x}Ca_x)_2Ir_2O_7$ for $x = 0.00$, 0.05, 0.07, and 0.10 under the applied field of 3.6 T.

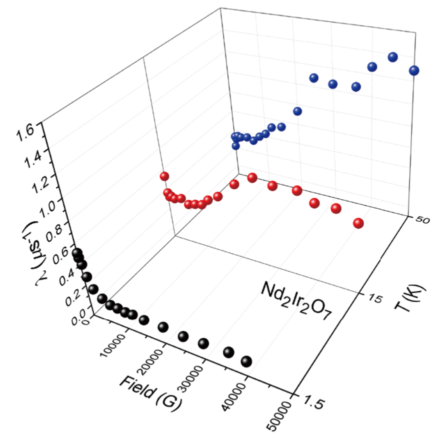

Fig. 2. Field dependence of the relaxation rate λ of $Nd_2Ir_2O_7$ at 1.5 K, 15 K, and 50 K.

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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) K. Tomiyasu *et al.*, J. Phys. Soc. Jpn. **81**, 034709 (2012).
4) H. Guo *et al.*, Phys. Rev. B **88**, 060411(R) (2013).
5) R. Asih *et al.*, J. Phys. Soc. Jpn. **86**, 024705 (2017).
6) H. Guo *et al.*, Phys. Rev. B **94**, 161102(R) (2016).
7) M. Watahiki *et al.*, J. Phys. Conf. Ser. **320**, 012080 (2011).
8) R. Asih *et al.*, RIKEN Accel. Prog. Rep. **50**, 23 (2017).