Magnetic order in pyrochlore iridate Nd\textsubscript{2}Ir\textsubscript{2}O\textsubscript{7} probed by employing muon spin relaxation\textsuperscript{†}

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Pyrochlore iridates are highly suitable to investigate novel topological phases based on the network of corner-sharing tetrahedra structures and the relatively large spin-orbit coupling (SOC) inherent in Ir 5d electrons.\textsuperscript{1} The interplay between SOC and electron-electron correlations ($U$) produces characteristic electronic states. A series of $R_2$Ir$_2$O$_7$ ($R$-227, $R$ = Nd-Ho) compounds exhibit metallic or semi-metallic behavior and undergo metal-insulator transitions (MITs) at a temperature $T_{\text{MI}}$\textsuperscript{2} while Pr-227 shows metallic behavior down to 0.3 K.\textsuperscript{3}

In this study, we focus on Nd-227, which shows metallic behavior at high temperatures and undergoes a MIT at $T_{\text{MI}}$ of about 30 K, and the magnetic susceptibility shows the bifurcation below $T_{\text{MI}}$ in zero-field-cooling (ZFC) and field-cooling (FC) conditions.\textsuperscript{2} Muon spin precession is observed below $T_{\text{MI}}$, and the spectra were fitted using the following function:

$$ A(t) = A_1 e^{-\lambda_1 t} + A_2 \cos(\gamma_H H_{\text{int}} t + \varphi) e^{-\lambda_2 t} $$

where $H_{\text{int}}$ is the internal field at the muon site, $\lambda_1$ is the muon spin-lattice relaxation rate, and $\lambda_2$ and $\varphi$ are the damping rate and initial phase of the muon spin precession, respectively.

The temperature dependence of the extracted parameters is shown in Fig. 1. $H_{\text{int}}$ begins to increase below $T_{\text{MI}}$, following the Brillouin-type ordering and tends to saturate below about 20 K to a value of about 350 G. Below about 10 K, $H_{\text{int}}$ increases again. $\lambda_2$ continues to decrease below $T_{\text{MI}}$ and increases again from the same temperature at which $H_{\text{int}}$ exhibits an increase. From the inset of Fig. 1(a), it can be seen that $\lambda_1$ increases monotonically with decreasing temperature, reflecting the slowing down of the magnetic moments. A small increase is observed around $T_{\text{MI}}$. However, no critical slowing down behavior is observed, as shown in Fig. 1(b). $\lambda_1$ continues to increase below $T_{\text{MI}}$ and shows a broad peak at around 10 K.

The increase in $H_{\text{int}}$ below 10 K is consistent with the results of the neutron scattering experiment that shows the ordering of Nd$^{3+}$ moments, so it is attributed to the ordering of Nd$^{3+}$ moments from our muon spin relaxation ($\mu$SR) experiment. The decrease in $\lambda_1$ below about 10 K is then accounted for by the freezing out of the magnetic fluctuations, and the increase in $\lambda_2$ suggests that the distribution of the internal field becomes larger at the vicinity of the magnetic ordering. The ordering below $T_{\text{MI}}$ is then attributed to the ordering of the Ir$^{4+}$ moments, suggesting its close relationship with the MIT.

According to a local spin-density approximation calculation including $U$ and SOC, the magnetic structure of the Ir sublattice is the all-in/all-out type, which does not break the lattice periodicity; therefore, the Slater transition is ruled out to account for the relationship between the MIT and the magnetic transition. On the other hand, the Lifshitz-like transition in which the hole band and electron band are moved downward and upward, respectively, due to the specific magnetic structure and the large SOC of Ir 5d electrons may explain the mechanism of MIT.

The saturated internal field from the ordered Ir$^{4+}$ moments is found to be much smaller than that in the case of the other pyrochlore iridates with a magnetic insulating ground state. This implies a stronger hybridization between the Ir 5d and the O 2p electronic orbitals in Nd-227.

References