Investigation of magnetic ordered states in the pyrochlore iridates (Nd, Ca)$_2$Ir$_2$O$_7$ probed by μSR


Pyrochlore iridates $R_2$Ir$_2$O$_7$ ($R$227, $R$ is a lanthanide element), have attracted growing interest because of their potential for realizing new topological states in the presence of strong spin-orbit coupling (SOC) and electron correlation ($U$), such as the Mott insulator, Weyl semimetal, and axion insulator. Interestingly, the electron correlation ($U$) in these compounds can be systematically tuned by changing the ionic radius of the $R$-ion ($r$). $R$227 shows systematic metal-insulator transition (MIT) at $T_{\text{MIT}}$, which gradually decreases by increasing the ionic radius of the $R^{3+}$ ion, and its boundary lies between $R$= Nd and Pr$^2)$. Abundant emergent quantum states have been theoretically predicted to occur on the boundary of MIT$^,$. In order to unravel those states, it is necessary to finely tune $U$ in this MIT-critical region. One way to do this is to substitute a nonmagnetic ion such as Ca for Nd, (Nd$_{1-x}$Ca$_x$)$_2$Ir$_2$O$_7$, which leads to the doping of holes in the Ir 5d band, and hence drives the transition from insulator to metal at the ground state and simultaneously suppresses magnetic orders. In this study, we systematically investigated changes in magnetic ordered states of Nd$_2$Ir$_2$O$_7$ due to hole doping by means of μSR measurements.

Pure Nd$_2$Ir$_2$O$_7$ exhibits metallic behavior and undergoes MIT at $T_{\text{MIT}}= 33$ K$^,$. Our μSR study on Nd$_2$Ir$_2$O$_7$ showed the appearance of a long-range magnetic order of Ir$^{4+}$ moments below $T_{\text{MIT}}$ followed by an additional magnetic order of Nd$^{3+}$ moments below 10 K$^,$. In the dilute hole-doped system $x = 0.01$, this Ir ordering appears at a lower temperature of around 26 K, as displayed in Fig. 1, indicating the suppression of the onset of the magnetic ordering. The zero-field (ZF) time spectra showed spontaneous muon-spin precession below 26 K, which was then well analyzed by the following function.

$$A(t) = A_r e^{-\lambda_r t} + A_c \cos(\gamma H_{\text{int}} t + \varphi)e^{-\lambda_c t}$$

(1)

The first component expresses the relaxing behavior with initial asymmetry $A_r$ and relaxation rate $\lambda_r$, and the second one expresses the muon-spin precession with initial asymmetry $A_c$, damping rate $\lambda_c$, and phase of the precession $\varphi$. Here $\gamma$ and $H_{\text{int}}$ are the gyromagnetic ratio of the muon spin ($2\pi \times 13.55$ kHz/G) and the internal field at the muon site, respectively.

The temperature dependences of the parameters obtained from the analysis of the ZF-μSR data are shown in Fig. 2. The dilute hole-doping gradually suppressed the onset of magnetic ordering and the internal field coming from the Ir$^{4+}$ ordering, while the internal field coming from the Nd$^{3+}$ ordered moments tended to increase below 5 K. The critical slowing down in the relaxation rate (Fig. 2b) indicates that Nd$^{3+}$ moments form a static ordering below about 10 K that does not rely on Ca concentration. Further measurements will be conducted on the intermediate and heavy Ca-doped systems to complete the magnetic phase diagram of (Nd$_{1-x}$Ca$_x$)$_2$Ir$_2$O$_7$.

Fig. 1. Zero field time spectra of (Nd$_{1-x}$Ca$_x$)$_2$Ir$_2$O$_7$ $x = 0.01$ at the early time region. Solid lines show fits to the data described in the text.

Fig. 2. Parameters derived from fitting Eq. 1 to the zero field μSR data of (Nd$_{1-x}$Ca$_x$)$_2$Ir$_2$O$_7$. (a) Internal field at muon sites $H_{\text{int}}$ and (b) relaxation rate $\lambda_r$. Solid lines are guides for the eye.

References