

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

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Pyrochlore iridates $R_2\text{Ir}_2\text{O}_7$ ($R227$, 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 ($SO\text{C}$) and electron correlation (U), such as the Mott insulator, Weyl semimetal, and axion insulator.¹⁾ Interestingly, the electron correlation (U) in these compounds can be systematically tuned by changing the ionic radius of the R -ion (r). $R227$ shows systematic metal-insulator transition (MIT) at T_{MI} , which gradually decreases by increasing the ionic radius of the R^{3+} ion, and its boundary lies between $R = \text{Nd}$ and Pr .²⁾ Abundant emergent quantum states have been theoretically predicted to occur on the boundary of MIT.¹⁾ 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, $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$, 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 $\text{Nd}_2\text{Ir}_2\text{O}_7$ due to hole doping by means of μSR measurements.

Pure $\text{Nd}_2\text{Ir}_2\text{O}_7$ exhibits metallic behavior and undergoes MIT at $T_{\text{MI}} = 33$ K.²⁾ Our μSR study on $\text{Nd}_2\text{Ir}_2\text{O}_7$ showed the appearance of a long-range magnetic order of Ir^{4+} moments below T_{MI} followed by an additional magnetic order of Nd^{3+} moments below 10 K.³⁾⁴⁾ 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.

$$A(t) = A_r e^{-\lambda_r t} + A_\omega \cos(\gamma_\mu H_{\text{int}} t + \varphi) e^{-\lambda_\omega t} \quad (1)$$

The first component expresses the relaxing behavior with initial asymmetry A_r and relaxation rate λ_r , and the second one expresses the muon-spin precession with initial asymmetry A_ω , damping rate λ_ω and phase of the precession φ . Here γ_μ and H_{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 $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$.

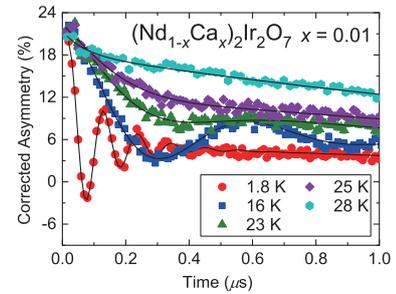


Fig. 1. Zero field time spectra of $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$ $x = 0.01$ at the early time region. Solid lines show fits to the data described in the text.

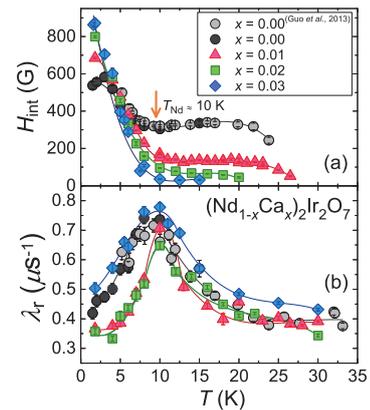


Fig. 2. Parameters derived from fitting Eq. 1 to the zero field μSR data of $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$. (a) Internal field at muon sites H_{int} and (b) relaxation rate λ_r . Solid lines are guides for the eye.

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