

# Investigation of magnetic ordered states in the pyrochlore iridates (Nd,Ca)<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> probed by $\mu$ SR

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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 ( $SOC$ ) and electron correlation ( $U$ ), such as the Mott insulator, Weyl semimetal, and axion insulator.<sup>1)</sup> 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  $T_{\text{MI}}$ , which gradually decreases by increasing the ionic radius of the  $R^{3+}$  ion, and its boundary lies between  $R = \text{Nd}$  and  $\text{Pr}$ .<sup>2)</sup> Abundant emergent quantum states have been theoretically predicted to occur on the boundary of MIT.<sup>1)</sup> 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  $\mu$ SR measurements.

Pure  $\text{Nd}_2\text{Ir}_2\text{O}_7$  exhibits metallic behavior and undergoes MIT at  $T_{\text{MI}} = 33$  K.<sup>2)</sup> Our  $\mu$ SR study on  $\text{Nd}_2\text{Ir}_2\text{O}_7$  showed the appearance of a long-range magnetic order of  $\text{Ir}^{4+}$  moments below  $T_{\text{MI}}$  followed by an additional magnetic order of  $\text{Nd}^{3+}$  moments below 10 K.<sup>3)4)</sup> 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  $\lambda_r$ , and the second one expresses the muon-spin precession with initial asymmetry  $A_\omega$ , damping rate  $\lambda_\omega$  and phase of the precession  $\varphi$ . Here  $\gamma_\mu$  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- $\mu$ SR data are shown in Fig. 2. The dilute hole-doping gradually suppressed the onset of magnetic ordering and the internal field coming from the  $\text{Ir}^{4+}$  ordering, while the internal field coming from the  $\text{Nd}^{3+}$  ordered moments tended to increase below 5 K. The critical slowing down in the relaxation rate (Fig. 2b) indicates that  $\text{Nd}^{3+}$  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  $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$ .

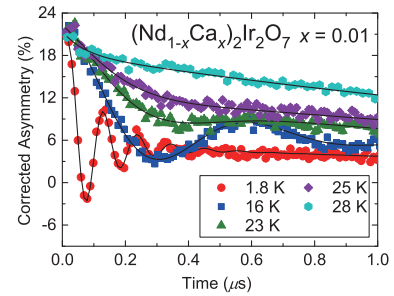


Fig. 1. Zero field time spectra of  $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$   $x = 0.01$  at the early time region. Solid lines show fits to the data described in the text.

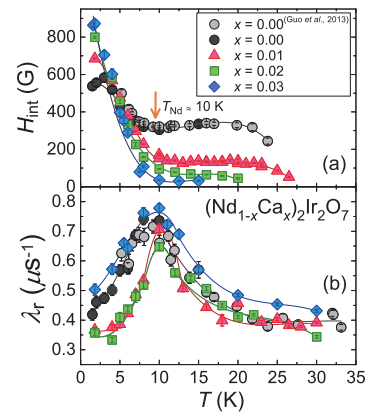


Fig. 2. Parameters derived from fitting Eq. 1 to the zero field  $\mu$ SR data of  $(\text{Nd}_{1-x}\text{Ca}_x)_2\text{Ir}_2\text{O}_7$ . (a) Internal field at muon sites  $H_{\text{int}}$  and (b) relaxation rate  $\lambda_r$ . Solid lines are guides for the eye.

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