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論文題目: <u>Muon-spin</u> relaxation studies of the pyrochlore iridates $Sm_2Ir_2O_7$, $Nd_2Ir_2O_7$ and $(Nd_{1-x}Ca_x)_2Ir_2O_7$

(パイロクロアイリジウム酸化物 Sm₂Ir₂O₇, Nd₂Ir₂O₇, (Nd_{1-x}Ca_x)₂Ir₂O₇ のミュ オンスピン緩和法による研究)

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概要: Pyrochlore iridates, R_2 Ir₂O₇ (R = rare-earth elements), provide an ideal platform to investigate novel topological phases built upon the network of the corner-sharing tetrahedra and the large spin-orbit interaction drawn from Ir 5*d* electrons. These compounds exhibit the metal-insulator transition (MIT) across the rare-earth series at room temperature. With increasing the ionic radius of R^{3+} , the temperature of MIT, $T_{\rm MI}$, gradually decreases and disappears between R = Nd and Pr.

This thesis describes μ SR studies on the pyrochlore iridates Sm₂Ir₂O₇, Nd₂Ir₂O₇ and (Nd_{1-x}Ca_x)₂Ir₂O₇. Sm₂Ir₂O₇ and Nd₂Ir₂O₇ are particularly attractive because they lie in the boundary of MIT and undergo the magnetic transition with MIT concomitantly at $T_{\rm MI} \approx 117$ and 33 K, respectively. By means of μ SR experiments, we confirmed the appearance of magnetic long-range ordering (LRO) of Ir moments below $T_{\rm MI}$ followed by additional LRO of Nd/Sm moments below about 10 K. The all-in all-out (AIAO) spin-structure was confirmed to be the most convinced model to explain our μ SR results. We compared observed internal fields at the muon site to those estimated from dipolar field and density functional theory calculations, and confirmed that lower limits of magnetic ordered moments were 0.12 μ B/Ir⁴⁺ and 0.2 μ B/Nd³⁺ in Nd₂Ir₂O₇ and 0.3 μ B/Ir⁴⁺ and 0.1 μ B/Sm³⁺ in Sm₂Ir₂O₇, respectively. By further analysis, it is concluded that the spin coupling between *R* and Ir moments should be ferromagnetic for Nd₂Ir₂O₇ and antiferromagnetic for Sm₂Ir₂O₇, respectively

The hole doping in Nd₂Ir₂O₇ via the Ca²⁺-substitution for Nd³⁺ was found to suppress a LRO of Ir magnetic moments. It was also confirmed that Nd moments keep undergoing the LRO below 10 K in $(Nd_{1-x}Ca_x)_2Ir_2O_7$ for $x \le 0.03$. No clear indication of the LRO was observed for x = 0.07 and 0.10 down to 0.3 K, except for the appearance of the slowing-down behaviour in the spin fluctuations below 2 K, signifying a possible occurrence of a magnetically short-range ordering (SRO). The magnetic phase diagram of $(Nd_{1-x}Ca_x)_2Ir_2O_7$ was proposed on the basis of the present studies.