Abstract of Doctoral Dissertation

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Title of Doctoral Dissertation

Muon-Spin Relaxation Study of Ir-Spin Fluctuations in Hole-Doped Pyrochlore Iridates $(Y_{1-x-y}Cu_xCa_y)_2Ir_2O_7$

(正孔ドープしたパイロクロア型イリジウム酸化物 $(Y_{1-x-y}Cu_xCa_y)_2Ir_2O_7$ における Ir スピン揺らぎのミュオンスピン緩和法による研究)

Recently, how the magnetic ordering of pyrochlore iridates $R_2Ir_2O_7$ changes in the boundary of metal-insulator transition (MIT) and eventually to be a possible spin liquid in $Pr_2Ir_2O_7$ remains unknown although those have been studied intensively over the past years. Magnetic properties of Ir moment are expected to play an essential role in various exotic states in $R_2Ir_2O_7$ such as spin liquid, Weyl semimetal, axion insulator, Mott insulator and so on. Of the pyrochlore iridates, Mott insulator $Y_2Ir_2O_7$ (Y^{3+}: non-magnetic; Ir^{4+}: $5d^5$) serves as an ideal system to investigate the magnetic properties of Ir moment to clarify its origin. This is because the Y atom does not possess a local magnetic moment and no *f* moment/no *d-f* interaction (larger spin-orbit interactions). $Y_2Ir_2O_7$ is reported to involve antiferromagnet ordering of Ir moment with all-in-all-out (AIAO) arrangement below the metal-insulator transition temperature, T_{MI} of around 170 K. Further, the mechanism of MIT also should be studied by doping holes to the system. Since a critical issue on Mott insulator is the hole-doping effect, we, therefore, investigated the changes in the magnetic properties of $(Y_{1-x-y}Cu_xCa_y)_2Ir_2O_7$ (x=0.05) in which the hole concentration can be controlled by substituting Ca for Y. The existence of a so-called quantum critical point (QCP) where the electronic ground state is changed in the vicinity of the zero temperature and near the disappearance of antiferromagnet transition is also theoretically predicted by carrier doping.

In this study, we have initially studied the magnetic properties of $Y_2Ir_2O_7$ and its hole-doped, $(Y_{1-x-y}Cu_xCa_y)_2Ir_2O_7$ with x = 0.05 and y = 0, 0.05, 0.08, 0.10, 0.20, and 0.25 by muon-spin relaxation (μSR) measurement. The synthesis of these compounds is reported in the thesis, as well. We also performed electrical resistivity, heat capacity, and magnetic susceptibility measurements. Finally, the study has indicated that the substitution of Ca^{2+} for Y^{3+} in hole-doped $Y_2Ir_2O_7$ suppresses both the magnetic transition temperature, T_N and T_{MI}. Moreover, T_N and T_{MI} may occur at the same temperature and decreases with increasing value of y. Both temperatures seem to vanish at around y = 0.15 with x = 0.05, indicating the existence of QCP. Our μSR results confirm the presence of an ordered magnetic state at below T_N. From the density functional theory calculation, the AIAO spin-structure is confirmed to be the convinced model to explain our μSR results. As far as we know, we have revealed the phase diagram of hole-doped $Y_2Ir_2O_7$ for the first time based on electrical transport and μSR measurements.