Magnetic Ground State of 4d Pyrochlore Oxides with Modified t_{2g} Band Filling

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The ruthenium pyrochlore oxides, $Hg_2Ru_2O_7$, $Tl_2Ru_2O_7$ and $Tl_2Rh_2O_7$, serve as model systems to understand the effect of orbital degeneracy controlled by population of t_{2g} band ($4d^3$, $4d^4$, and $4d^5$ in their ionic limit, respectively). It is inferred from our muon spin rotation/relaxation (μSR) measurements that these compounds exhibit completely different electronic ground states with each other, while they show a common bulk property of reduced magnetic susceptibility (χ_0) associated with metal-insulator transition at low temperatures.

$Hg_2Ru_2O_7$, having no orbital freedom (half-filled t_{2g}), exhibits antiferromagnetic order as inferred from zero-field (ZF) μSR spectra shown in Fig.1(a) [1]. Meanwhile, $Tl_2Ru_2O_7$ ($S = 1$) and $Tl_2Rh_2O_7$ ($S = 1/2$) exhibit no signs of magnetic order in ZF-μSR spectra [Figs.1(b) and 1(c)]. Moreover, they show reduction of muon Knight shifts associated with the reduction of χ_0. These observations support the formation of Haldane chains in $Tl_2Ru_2O_7$[1-3], and further suggest the occurrence of a spin-singlet state (e.g., associated with spin-Peierls transition for $S = 1/2$ chains) in $Tl_2Rh_2O_7$.

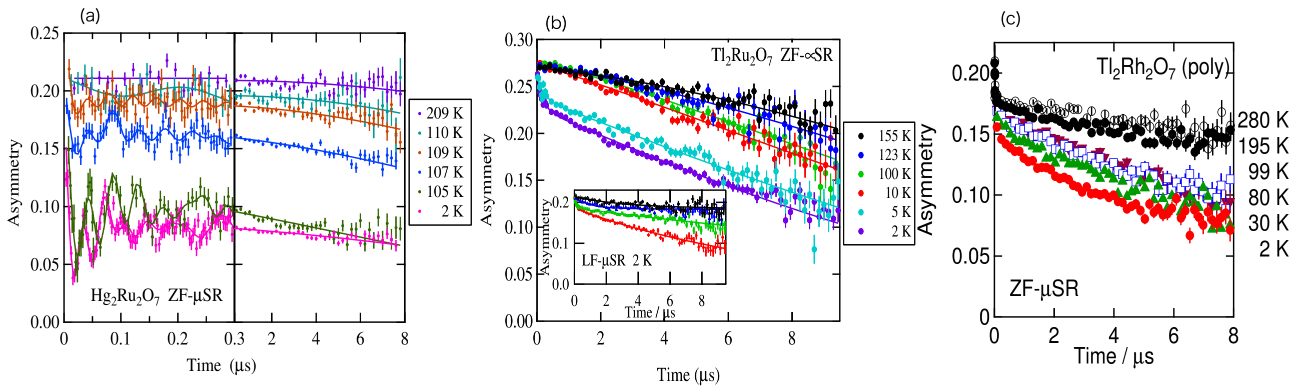

Figure 1: ZF-μSR time spectra observed in (a) $Hg_2Ru_2O_7$ (with the metal-inslator transition temperature $T_{MI} = 107$ K), (b) $Tl_2Ru_2O_7$ ($T_{MI} = 125$ K, inset: μSR time spectra under a longitudinal field), and $Tl_2Rh_2O_7$ ($T_{MI} = 95$ K).

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[3] S. Lee *et. al.* Nature Mater. **5** (2006) 471.