Non-magnetic ground state of $In_2Ru_2O_7$ probed by muon spin rotation

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The interplay of charge, spin, and orbital degrees of freedom in correlated systems gives rise to a variety of exotic electronic states. Among these materials, ruthenium oxides containing Ru^{4+} cations (d^4 electron count), which host orbital degeneracy, display a variety of spin-orbital ordering phenomena such as spin singlet dimer formation in Li_2RuO_3[1)] and La_2RuO_5.[2)] A particularly interesting situation may be found when Ru^{4+} cations are placed on a frustrated lattice, for example, in pyrochlore ruthenate $Tl_2Ru_2O_7$, which displays a metal-to-insulator transition with a spin-singlet ground state.[3)] The spin-singlet state is discussed to originate from the formation of 1D zig-zag Haldane chains and is potentially the first example of such a system emerging in a 3D lattice. While there is still discussion on the plausibility of the Haldane-chain scenario, the origin of this unique state and behavior distinct from those of other pyrochlore ruthenates remains unclear. Pyrochore ruthenates appear to be an interesting playground for novel electronic states.

We discovered a new pyrochlore ruthenate $In_2Ru_2O_7$ which adopts a weakly distorted pyrochlore structure at 300 K. $In_2Ru_2O_7$, is an insulator and shows Curie-Weiss behavior at high temperatures. At $T_m \sim 220$ K, a sharp drop in magnetic susceptibility $\chi(T)$, similar to those of other spin-singlet ruthenate compounds,[1–3)] is observed. To elucidate the nature of the ground state of $In_2Ru_2O_7$ and the absence of a long-range magnetic ordered state, the temperature dependence of zero-field muon spin rotation (ZF-μSR) is necessary. In this report, we present the result of a ZF-μSR experiment performed on CHRONUS at ISIS, UK. We recorded the time evolution of ZF-μSR spectra of $In_2Ru_2O_7$ from 300 K down to 10 K (Fig. 1(a)). No oscillation down to the lowest temperature was observed, excluding the presence of long-range antiferromagnetic order and pointing to a non-magnetic spin singlet state. The data over the whole studied temperature range were fitted to a "stretched" exponential:

$$P_z(t) = e^{-(\lambda t)^\beta}, \tag{1}$$

where $P_z(t)$ is the asymmetry, t is the time, λ is the relaxation rate, and β is the shape parameter. We observed a cross-over from a Gaussian-like to a Lorentzian-like damping with decreasing temperature (Figs. 1(a) and (b)). The high-temperature and low-temperature response can be explained by the slowing behaviour of the fluctuations of dilute Ru spins, which originate from the defects present in the system, or by the appearance of slowly fluctuating quasi-particle spins excited from the spin-singlet state induced by injected muons. Such a response was reported in other materials hosting a spin-singlet ground state.[5,6)] In addition, a sharp change in λ and β is observed at T_m (Fig. 1(b)). The enhancement of λ below T_m would point to the opening of a gap.

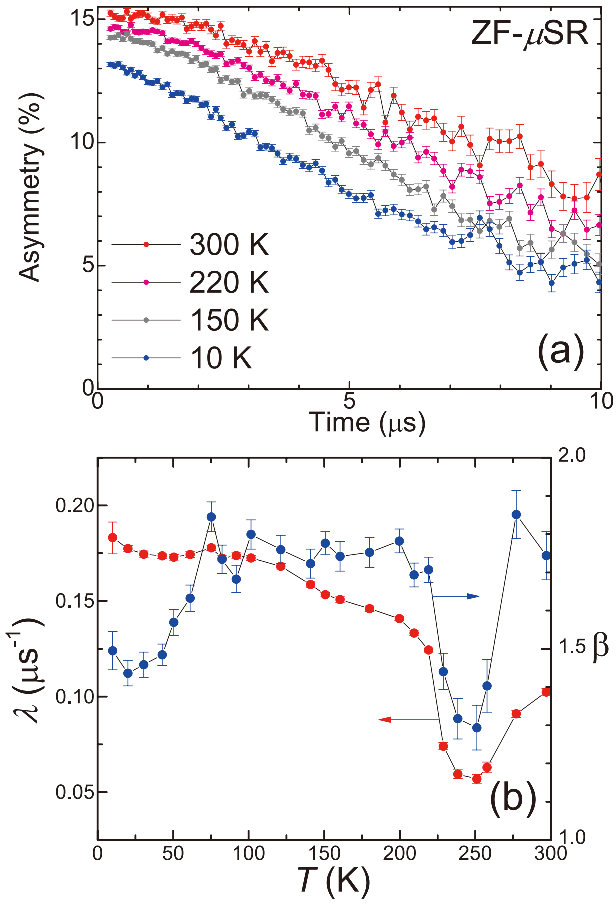

Fig. 1. (a) Time evolution of ZF-μSR spectrum of $In_2Ru_2O_7$ powder sample at various temperatures. (b) Temperature dependence of λ and β fitted using equation (1).

In this report, the ground state of $In_2Ru_2O_7$ was investigated with ZF-μSR. No evidence of long-range magnetic order was found; hence, the non-magnetic ground state was confirmed. The temperature dependence of the relaxation rate implies the opening of a gap. Other techniques such as resonant inelastic x-ray scattering could aid the determination of the gap size.

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References

1) Y. Miura *et al.*, J. Phys. Soc. Jpn. **76**, 033705 (2007).
2) H. Wu *et al.*, Phys. Rev. Lett. **96**, 256402 (2006).
3) S. Lee *et al.*, Nat. Mat. **5** (2006).
4) K. Kojima *et al.*, Phys. Rev. Lett. **74**, 3471 (1995).
5) S. J. Blundell *et al.*, Phys. Rev. B **77**, 094424 (2008).
6) M. Miyazaki *et al.*, Phys. Rev. B **75**, 09440 (2007).