Magnetic ground state of $J_{\rm eff} = 0$ Mott insulator $Ag_3LiRu_2O_6$ under pressure

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The interplay of strong spin-orbit coupling (SOC) and electron correlation in heavy transition-metal oxides has been found to give rise to unprecedented electronic phases. Among such oxides, Mott insulators with a d^4 (t_{2g}^4) configuration are expected to exhibit exotic magnetism. In the d^4 systems, Hund's coupling produces an $S = 1$ and $L_{\rm eff} = 1$ state. SOC thus yields a spin-orbit-entangled $J_{\rm eff} = 0$ ground state. Although the $J_{\rm eff} = 0$ state is non-magnetic, it may interact magnetically with each other through the excited $J_{\rm eff} = 1$ levels separated by SOC. It was proposed that the system undergoes magnetic ordering when the energy gain by magnetic interactions overcomes the energy cost of the $J_{\rm eff} = 0$ to 1 excitation, which is dubbed excitonic magnetism.[1] Particularly, when the $J_{\rm eff} = 0$ state is arranged on a honeycomb lattice, exotic magnetic ground states such as a spin-nematic phase or magnetic triplon liquid, are expected to emerge.

The $J_{\rm eff} = 0$ honeycomb lattice may be realized in Ru^{4+} ($4d^4$) oxides. However, a honeycomb ruthenate Li_2RuO_3 is known to display spin-singlet dimerization accompanying orbital ordering.[2] In the dimerized state, the spin and orbital degrees of freedom are quenched, and spin-orbit coupling is inactive. In order to suppress the dimerization and realize the $J_{\rm eff} = 0$ state, we employed an ion-exchange reaction and obtained the silver-intercalated compound $Ag_3LiRu_2O_6$.[3] The crystal structure at room temperature shows no appreciable distortion in the Ru honeycomb lattice, indicating the disappearance of dimerization.

The magnetic susceptibility $\chi(T)$ of $Ag_3LiRu_2O_6$ increases on cooling from room temperature and shows saturation below 100 K. Interestingly, no magnetic order is observed down to 0.3 K. From inelastic neutron scattering and ^{7}Li-NMR measurements, $Ag_3LiRu_2O_6$ was found to have a gapped magnetic excitation of ∼35 meV, as expected for the $J_{\rm eff} = 0$-type singlet ground state.[4] The singlet ground state suggests that the magnetic exchange interactions are not strong enough to drive excitonic magnetism. We thus investigated the magnetic properties under high pressures as an attempt to enhance magnetic interactions. Indeed, under a pressure above 0.4 GPa, $\chi(T)$ shows a broad peak below room temperature, indicating the appearance of a pressure-induced electronic phase transition. However, the nature of the magnetic ground state under pressure remains unclear.[4]

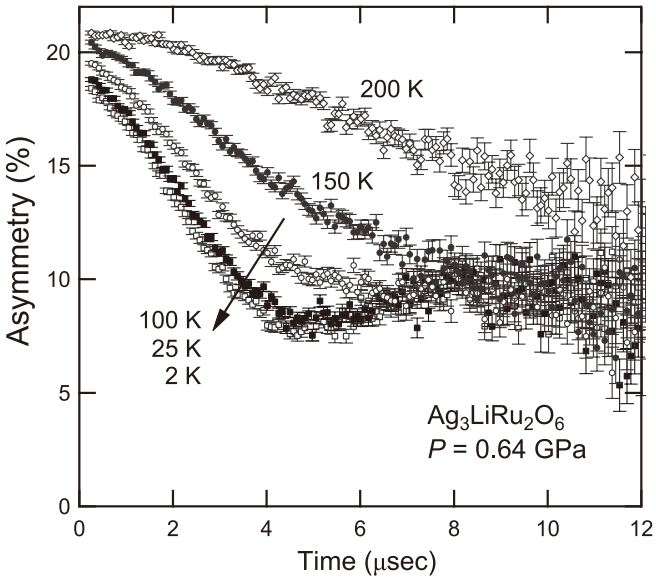

Fig. 1. Zero-field time spectra of muon asymmetry for $Ag_3LiRu_2O_6$ under a pressure of 0.64 GPa.

In order to understand the magnetic ground state under pressure, we performed a muon spin rotation measurement on $Ag_3LiRu_2O_6$ under pressure by using ARGUS, ISIS. At ambient pressure, the time dependence of muon asymmetry at low temperatures only shows a monotonic decrease, which is consistent with the nonmagnetic singlet ground state. As shown in Fig. 1, the time dependence of muon asymmetry did not show any pronounced change down to 2 K even at a pressure of 0.64 GPa, while $\chi(T)$ showed a broad peak at approximately 150 K at that pressure. This suggests that the broad peak observed in $\chi(T)$ under pressure is not associated with a magnetic ordering but represents another electronic phase transition while retaining the nonmagnetic ground state.

The nature of the pressure-induced nonmagnetic phase, revealed by the μSR measurement, remains elusive. The x-ray diffraction under pressure excluded the restored dimerization in this pressure range. The candidates of possible ground states include a spin-orbit triplon liquid, molecular orbital formation, or fluctuating dimers. We are currently attempting to determine the nature of this phase in combination with other spectroscopic probes.

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