Investigations of the quantum critical behavior of the $S = 1/2$ J_1-J_2 square lattice antiferromagnets $Sr_2Cu(Te_{1-x}W_x)O_6$

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The J_1-J_2 square-lattice Heisenberg model offers a rich reservoir of exotic quantum phases. Depending on the ratio J_2/J_1 (the ratio of exchange interactions between a diagonal and a side of the square), the classical ground states encompass Néel, columnar antiferromagnetic, and disordered states. In particular, the B-site ordered double perovskites $Sr_2Cu(Te_{1-x}W_x)O_6$ ($B = Cu^{2+}$, $B'' = Te^{6+}$ or W^{6+}) with the tetragonal $I4/m$ structure are regarded as near-optimal realizations of the $S = 1/2$ Heisenberg model on a square lattice.[1–4] Notably, the magnetic ground state of $Sr_2Cu(Te_{1-x}W_x)O_6$ can be tuned from a Néel order ($x = 0$; $J_2/J_1 = 0.03$) through a quantum spin liquid to a columnar antiferromagnetic order ($x = 1$; $J_2/J_1 = 7.92$) by varying the B'' cation between Te^{6+} and W^{6+} ions.[3] Owing to the similar sizes of the Te^{6+} and W^{6+} ions, $Sr_2Cu(Te_{1-x}W_x)O_6$ retains the isostructural crystal lattice.

Unlike the crystal structure, the magnetism is susceptible to a composition of the B'' cations that are involved in the magnetic exchange paths mediating the Cu^{2+} ions. W^{6+} ($5d^0$) hybridizes strongly with O $2p$, thereby enabling the magnetic exchange path Cu-O-W-O-Cu (J_2). This is in sharp contrast to the Te^{6+} ($4d^{10}$) ion, which is in favor of J_1. The distinct role of the Te^{6+}/W^{6+} ions in relation to the exchange interaction poses the question as to whether the theoretically predicted spin liquid phase can be achieved by controlling the composition of the B'' cations. Strikingly, there is tantalizing experimental evidence for a quantum disordered state in a wide composition range of $x \approx 0.1 - 0.7$.[3] However, the observed wide range of the spin-liquid phase is not compatible with the theoretically predicted narrow composition range of $x \approx 0.23 \sim 0.33$. Recent theoretical calculations have proposed quenched disorders as a key ingredient to this discrepancy.[3,4] To clarify the precise nature of the ground state, we focus on the compositions in the range of $x = 0.05 - 0.1$, which is at the boundary between the Neel antiferromagnetic order and the quantum disordered state.

Muon spin relaxation measurements were performed on ARGUS and CHRONUS spectrometers at RIKEN-RAL towards understanding the precise nature of the ground state and associated spin dynamics of the $x = 0.05$, 0.08, and 0.1 compounds. Representative spectra are shown in Fig. 1. No spontaneous muon spin oscillation is observed, indicating the absence of long-range magnetic order. Moreover, all the observed μSR spectra are described by a stretched exponential function $\exp[-(\lambda t)^{\beta}]$, where λ is the muon relaxation rate and β is the exponent.

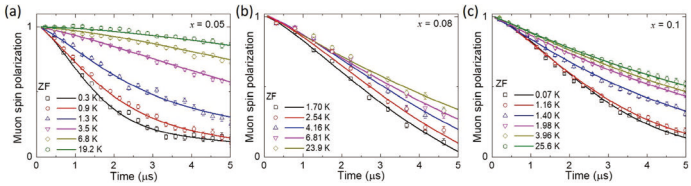

Fig. 1. Muon spin polarization data observed at zero field together with fitting curves (see the text for details) for $x = 0.05$, 0.08, and 0.1.

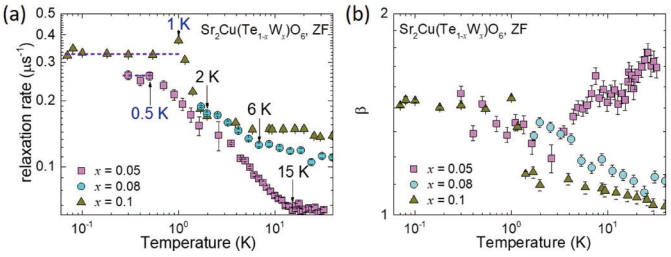

Fig. 2. Temperature dependences of the relaxation rate (a) and its stretch exponent (b) for $x = 0.05$, 0.08, and 0.1.

Temperature dependences of the muon spin relaxation rate, $\lambda(T)$, and its stretch exponent, $\beta(T)$, for $x = 0.05$, 0.08, and 0.1 are plotted in Fig. 2. $\lambda(T)$ of $x = 0.05$ starts to increase on cooling below 15 K and subsequently becomes constant for temperatures below 0.5 K. In the $x = 0.08$ and 0.1 compounds, the crossover regime shrinks, and onset temperatures shift to 6 K and 2 K for $x = 0.08$ and 0.1, respectively; on the other hand, a plateau appears at 1 K for $x = 0.1$. We comment that the $x = 0.05$ and 0.1 compounds reveal the persistent spin dynamics below $T = 0.5 - 1$ K, which is often observed in frustrated magnets.

In addition, the T-dependence of the stretching exponent, $\beta(T)$, of $x = 0.08$ and 0.1 resemble $\lambda(T)$ while increasing from 1 to 1.6 with decreasing temperature. In sharp contrast, $\beta(T)$ of $x = 0.05$ decreases from 1.8 to 1.3 on cooling to 25 K. Noticeably, the low-T $\beta(T)$ of the compounds is between those of Gaussian and Lorentzian relaxation functions. When we consider that a Gaussian-like distribution is expected for dense disordered systems or migrating unpaired spins in the matrix of singlet spins and that a Lorentzian-like distribution is expected for dilute systems, the ground state for $x = 0.05$, 0.08, and 0.1 is largely compatible with the random singlet picture comprising resonating singlets and propagating orphan-like spins.

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