2 Geometrical Correlation Project

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2-1 Quasi-one-dimensional spin dynamics in *d*-electron heavy-fermion metal $Y_{1-x}Sc_xMn_2$

Yttrium manganite (YMn_2) is an intermetallic Laves phase (C15-type) compound and the first example of transition-metal systems in which a heavy quasiparticle (QP) state has been observed. It comprises a three-dimensional network of corner-shared tetrahedra with Mn ions at their corners, providing a stage equivalent to the cubic pyrochlore lattice. Although YMn_2 exhibits magnetic order with complex helical modulation and a large volume expansion below T_N ~100 K, it remains in a paramagnetic state under hydrostatic pressure (• 0.4 GPa) or upon the substitution of Y by Sc ($Y_{1-x}Sc_xMn_2$, with x • 0.03) which is also accompanied by a large increase in QP mass (m^* ~15 times the band mass) as inferred from the electronic specific heat [1,2]. Despite studies focused on identifying the origin of the heavy-QP state, the issue remains a major challenge that has increased in importance with the succeeding discovery of LiV_2O_4 [3].

The heavy (QP) mass (m^*) is phenomenologically understood to come from the sharp increase in the electronic density of states near the Fermi surface and associated flattening of band dispersion [$dN(E_F)/dE \propto m^* \rightarrow \infty$]. In rare-earth compounds, such enhancement is induced by the conversion of local *f*-electron degrees of freedom into $N(E)$ by the Kondo effect, which is observed as a peak structure of $N(E)$ near E_F. While such a structure in $N(E)$ has been suggested in LiV_2O_4 by photoemission spectroscopy [4], the dynamical fluctuation of seemingly "local" vanadium magnetic moments associated with the heavy-QP state has been inferred from muon spin rotation and relaxation (μSR) measurements [5,6]. The latter indicates that the conventional Kondo coupling (which virtually eliminates "local" spins over a time scale longer than $\nu_{ex}^{-1} = h/J_{cf} \sim 10^{-13}$–$10^{-14}$ s, where J_{cf} is the exchange energy between conduction electrons and *f* electrons) is not in effect, thereby suggesting that the heavy QP state has a different origin.

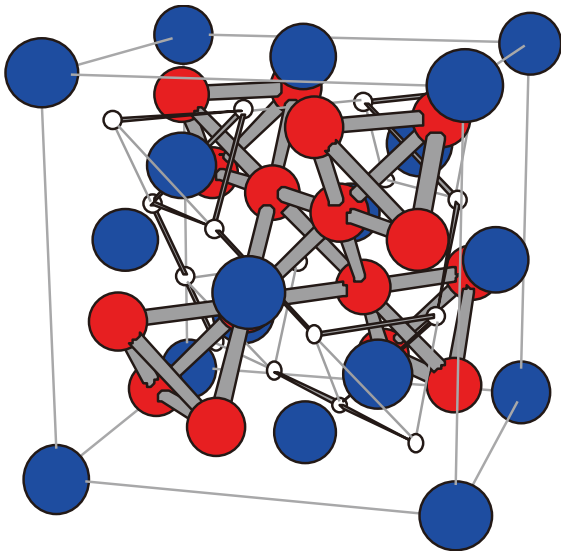

Fig. 1 Crystal structure of YMn_2, where Y and Mn atoms are indicated by blue and red circles, respectively. Small open circles show 16c sites where hydrogen atoms are located in hydrates, which are presumed to be occupied by implanted muons. These Mn atoms form a network of corner-shared tetrahedra known as a "pyrochlore" lattice.

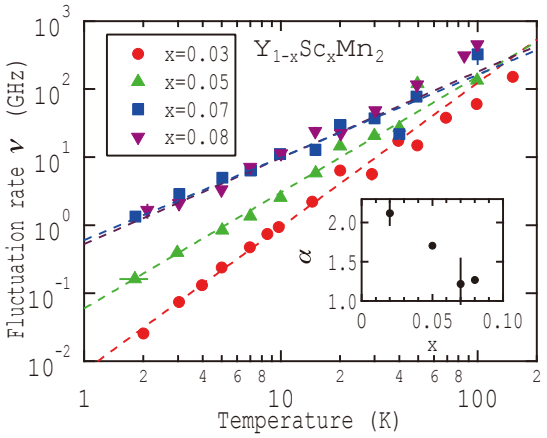

Fig. 2 Spin fluctuation rate (ν) as a function of temperature and Sc content (x) in $Y_{1-x}Sc_xMn_2$. Solid lines are the results of curve fitting using a power-law ($\nu \propto T^{\alpha}$). Inset: α obtained by curve fits vs x.

This situation naturally turns our interest to the role of geometrical frustration in electronic degrees of freedom such as spin, charge, and orbit, which is relevant to atoms interacting on the cubic pyrochlore lattice. The pyrochlore structure is shared by LiV_2O_4 and $Y_{1-x}Sc_xMn_2$ (see Fig. 1), where the t_{2g} orbits associated with vanadium or manganese ions form a network of intersecting chains. Since these compounds remain mostly paramagnetic, spin fluctuation would be crucially important information for understanding their electronic state. In FY2010–2011, we performed a detailed study on the spin dynamics of $Y_{1-x}Sc_xMn_2$ by the muon spin relaxation (μSR) technique at various Sc contents x. As shown in Fig. 2, we found that Mn spin fluctuation persists over a low frequency range ($\nu < 10^{12}$ s^{-1}), which is characterized by a power-law temperature dependence, $\nu \simeq c \cdot T^{\alpha}$. The power α asymptotically approaches unity with increasing x (∝ chemical pressure), while $\alpha \sim 2$ as ν shows a tendency of rapid slowing down toward a quasistatic spin-glass state

near x = 0.03 [7]. The presence of such linear T dependence of spin fluctuation strikingly resembles that in the case of LiV_2O_4, which is understood as a characteristic property of spin-spin correlations for the intersecting Hubbard chains as a model of the pyrochlore lattice [8]. This implies the crucial role of t_{2g} orbitals as one-dimensional chains that are under a strong geometrical constraint of pyrochlore lattice structures, and further suggests that the dimensional crossover due to coupling between these chains is one of the origins of the heavy-fermion state [9].

2-2 Quasi-one-dimensional spin dynamics in LiV_2O_4: one-to-three-dimensional crossover as a possible origin of heavy fermion state

As mentioned in the previous section, the heavy fermion behavior observed in a cubic vanadium spinel, LiV_2O_4, has drawn much interest, since it is one of the remarkable examples in which only *d*-orbital electrons are relevant to the electronic property. The formation of a heavy QP state below a characteristic temperature, i.e., T^*~20 K, is suggested by a large Sommerfeld coefficient (~420 mJ/mol K^2) and other bulk properties that are strikingly similar to typical *f*-electron heavy fermion compounds [3]. Moreover, it has been inferred from recent photoemission spectroscopy that the peak of the density of states (DOS) just above E_F develops for $T<T^*$, which may correspond to the QP peak typically found for *f*-electron systems [4].

In our previous μSR study, we showed in a powder specimen of LiV_2O_4 that the observed μSR signal consists of two components characterized by different responses of depolarization rate (λ) to external magnetic field (H_0) [5]. In particular, the signal with λ showing the weakest dependence on H_0 (λ_D) with a fractional yield (f~0.4) is mostly independent of temperature below ~10^2 K, from which we suggested that the corresponding fluctuation rate derived from the general relation between λ and ν, i.e.,

$$\lambda \approx \frac{2\delta_\mu^2 \nu}{\nu^2 + \gamma_\mu^2 H_0^2} \qquad (1)$$

is also independent of temperature ($\nu_D > 10^9$ s^{-1}). In contrast, λ associated with another signal (λ_S, with 1-f ~ 0.6) is readily suppressed by H_0, which has been ascribed to slowly fluctuating local magnetic moments (ν_S ~ 10^6–10^7 s^{-1}). Although the occurrence of such phase separation has been confirmed by a subsequent μSR study of high-quality single-crystalline samples, the increased yield f (~0.8) strongly suggests that it is essential to clarify the origin of ν_D in order to understand the electronic state in LiV_2O_4 [6].

Despite the fact that LiV_2O_4 is metallic, the use of eq. (1), which is valid in the limit of localized spins with ν determined by the local exchange interaction ($J \sim h\nu$), is presumed to be justified by the presence of staggered vanadium moments suggested by a broad μSR linewidth at low temperatures (which is also in line with presumption of the Kondo scenario for $T>T^*$) [5]. Moreover, eq. (1) may be regarded as a good approximation at high temperatures where the electronic state is subject to strong damping by phonons. However, as shown in the previous section, our recent μSR study on $Y_{1-x}Sc_xMn_2$ demonstrated that the interpretation of depolarization at low temperatures requires greater care.

Given this situation, we re-examined the spin dynamics of LiV_2O_4 on an alternative basis that staggered magnetic moments are carried by itinerant *d* electrons. The corresponding muon spin depolarization is described by a modified version of eq. (1), i.e.,

$$\lambda \approx \frac{2\delta_\mu^2 \nu}{\nu^2 + \gamma_\mu^2 H_0^2} \cdot \frac{\chi k_B T}{N_A \mu_B^2} \qquad (2)$$

where factors additional to those in eq. (1) stem from the electronic density of state at the Fermi level, with χ being the magnetic susceptibility, N_A the Avogadro number, and μ_B the Bohr magneton. As a consequence, our reanalysis indicates that ν_D is linearly dependent on temperature ($\nu \propto T$) with frequencies ranging from 10^8 to 10^{12} s^{-1}, which coincidentally serves to cancel the T-dependence of λ. As shown in Fig. 3, the distilled behavior of ν_D turns out to be in excellent agreement with the previous result of inelastic neutron scattering [10], strongly suggesting that both μSR and INS have

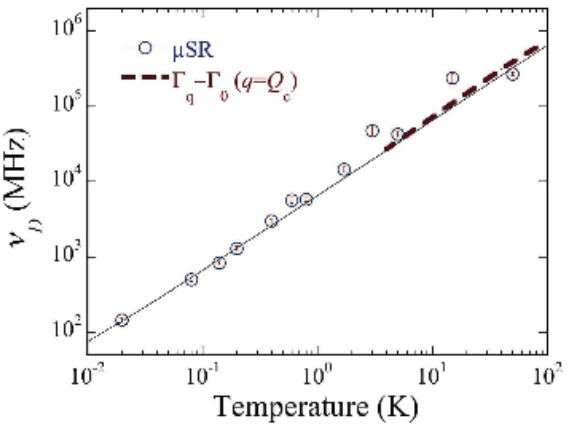

Fig. 3 Spin fluctuation rate (ν) as a function of temperature in LiV_2O_4. The thin solid line shows a linear T dependence ($\nu \propto T$). Inelastic neutron scattering (INS) data are also shown for comparison, where the dashed curve shows the linewidth ($_q$ − $_0$)/h at Q_c = 0.64 A^{-1} with $_0$~1.5 meV.

observed a common phenomenon [11]. Moreover, the linear T dependence of spin fluctuation, which is commonly observed in $Y_{1-x}Sc_xMn_2$, is understood as a property characteristic of the spin-spin correlation of intersecting Hubbard chains that simulate the pyrochlore lattice [8]. This again implies the important role of strong geometrical constraint to t_{2g} orbitals exerted by a pyrochlore lattice structure, and the origin of the heavy-fermion state common to that of $Y_{1-x}Sc_xMn_2$.

Acknowledgement

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