

2 Geometrical Correlation Project

– Quest for the origin of heavy fermion behavior in *d*-electron systems –

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2-1 Quasi-One-Dimensional Spin Dynamics of Mn pyrochlore lattice in $\text{YMn}_2\text{Zn}_{20-x}\text{In}_x$

Geometrical frustration in the electronic degrees of freedom such as spin, charge, and orbit, which is often realized with the highly symmetric crystals, has been one of the major topics in the field of condensed matter physics. In particular, the heavy fermion (HF) behavior in $\text{Y}(\text{Sc})\text{Mn}_2$ [1] and LiV_2O_4 [2] has attracted broad interest, where such a local electronic correlation specific to highly symmetric pyrochlore structure may be of direct relevance to the formation of the heavy quasiparticle (QP) state. However, despite decades of studies, the microscopic mechanism of how such a *local*

correlation is transmuted into the heavy QP mass of *itinerant d*-electrons in those compounds remains controversial.

One of the difficulties for investigating the *d*-electron HF-like state is the limited number of actual compounds that exhibit such behavior. The recent development of a ternary intermetallic compound, $\text{YMn}_2\text{Zn}_{20}$, might ease the situation, as it exhibits a large Sommerfeld coefficient characteristic of the HF compounds [2]. It crystallizes in the cubic $\text{CeCr}_2\text{Al}_{20}$ structure with the space group $Fd\bar{3}m$ (see Fig. 1), where the Y and Mn atoms occupy the 8a and 16d sites forming diamond and pyrochlore lattices, respectively, common to the cubic Laves-phase Y Mn_2 . Meanwhile, Zn atoms at the 16c, 48f, and 96g sites are located between two other atoms, so that the pyrochlore lattice composed of Mn atoms is almost doubly expanded while keeping the tetrahedral symmetry in comparison with that in $\text{Y}(\text{Sc})\text{Mn}_2$. Although a compound with this ideal composition has not yet been obtained, partial substitution of In for Zn is known to be effective for stabilizing the structure.

We have investigated the spin dynamics of Mn *d*-electrons by means of muon spin relaxation under a zero/longitudinal field (experiment conducted at RIKEN-RAL) in a polycrystalline sample of $\text{YMn}_{2+\delta}\text{Zn}_{20-\delta-x}\text{In}_x$ with $x = 2.36$, in which the influence of excess Mn seems to be minimal [$\delta = 0.11(1)$]. The obtained μSR spectra [positron asymmetry, $A(t)$] were analyzed using a form $A(t) = A_0 G_z^{\text{KT}}(t) \exp(-\lambda t) + A_b$, where A_0 is the initial positron decay asymmetry, $G_z^{\text{KT}}(t)$ is the Kubo-Toyabe relaxation function, λ is the depolarization rate, and A_b is the time-independent background mainly coming from muons stopped in the sample holder (made of silver). The signal to noise ratio (A_0/A_b) was ~ 3 with a time window for measurement

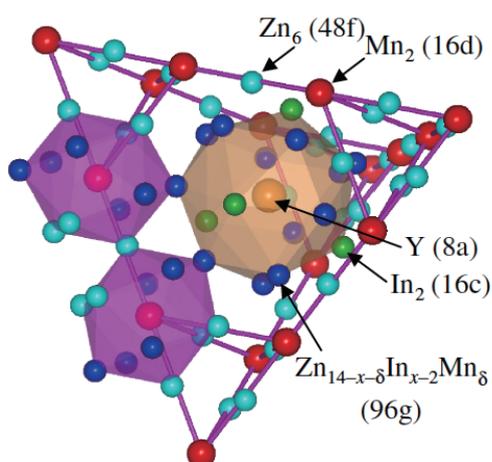


Fig. 1: Crystal structure of $\text{YMn}_{2+\delta}\text{Zn}_{20-\delta-x}\text{In}_x$, where Y, Mn, and Zn atoms are indicated by yellow, red, and blue spheres, respectively. A part of Zn at the 96g site is substituted by In to stabilize the $\text{CeCr}_2\text{Al}_{20}$ structure. The actual compound exhibits a small amount (δ) of excess Mn that occupies the 96g site to substitute Zn. The Mn-Zn tetrahedra form a network similar to the pyrochlore lattice, where the local interaction between Mn atoms is relatively weaker than that in $\text{Y}(\text{Sc})\text{Mn}_2$ due to Zn atoms situated between Mn atoms.

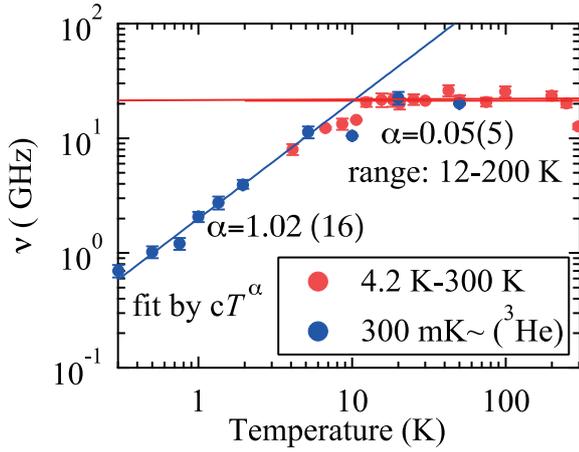


Fig. 2: Spin fluctuation rate (ν) as a function of temperature in $\text{YMn}_{2+\delta}\text{Zn}_{20-\delta-x}\text{In}_x$ with $x = 2.36$ and $\delta = 0.11$. Solid lines are the results of curve fitting using a power-law ($\nu \propto T^\alpha$) for the respective range of temperature.

of up to 20 μs , allowing reliable deduction of λ at small values ($\sim 10^{-2} \mu\text{s}^{-1}$).

The spin fluctuation rate ν at a given temperature T is deduced using the following relationship between λ and ν in metals,

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

where $\chi(T)$ is the magnetic susceptibility, N_A is the Avogadro number, δ_μ is the muon hyperfine parameter, γ_μ is the muon gyromagnetic ratio, and H_0 is the external field. Assuming that i) $\delta_\mu = \gamma_\mu \times 195.9 \text{ G}/\mu_B$ for a presumed muon site at the center of Mn tetrahedra, and that the effect of excess Mn can be subtracted from $\chi(T)$ by curve fitting (sum of two Curie terms), we obtain ν vs T as shown in Fig. 2.

Interestingly, ν is linearly dependent on temperature ($\nu \propto T$) below a crossover temperature (T^*) with frequencies ranging from 10^9 to 10^{10} s^{-1} . The presence of such linear T dependence of spin fluctuation strikingly resembles that in the cases of $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ [4] and LiV_2O_4 [5], which is understood as a characteristic property of spin-spin correlations for the intersecting Hubbard chains as a model of the pyrochlore lattice [6]. 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 [7].

Meanwhile, in contrast to $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$, ν is mostly independent of T above $T^* \sim 10 \text{ K}$. This is in line with the fact that the HF behavior seems to set in only

below T^* as suggested from the temperature dependence of the Sommerfeld coefficient [3], and it may be understood to be a consequence of weaker Mn-Mn interaction in comparison with that in $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$.

2-2 Strongly anisotropic muon-Mn hyperfine interaction in $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ ($x = 0.05$)

As mentioned in the previous section, the origin of HF behavior in d -electron systems such as $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ and LiV_2O_4 remains an enigmatic problem in condensed matter physics, and is the primary subject of the Geometrical Correlation Project. We have been focusing on $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ for the past few years to clarify the relation between spin dynamics and the HF behavior.

$\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ is an intermetallic compound with the cubic C15 Laves phase structure [space group $Fd\bar{3}m$, as shown in Fig. 3(a)], where the Mn atoms

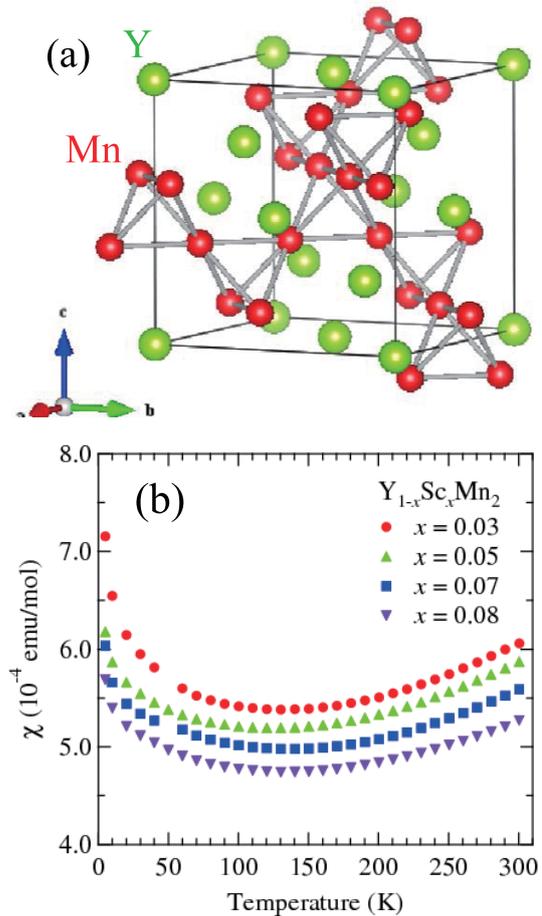


Fig. 3: a) Crystal structure of YMn_2 , where Y and Mn atoms are indicated by green and red spheres, respectively. The network of corner-shared Mn tetrahedra comprises the pyrochlore lattice. Spin susceptibility vs. temperature in $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ with $x = 0.03, 0.05, 0.07$ and 0.08 . (after Ref. [4])

form a three-dimensional (3D) network of corner-sharing tetrahedra called pyrochlore lattice. While it falls into an antiferromagnetically ordered state with a helical spin structure below $T_N = 100$ K, substitution of Y by Sc leads to the HF behavior upon suppression of the magnetic order for $x > 0.03$.

In our previous study, we conducted μ SR measurements in $Y_{1-x}Sc_xMn_2$ (with a series of specimens with $x = 0.03, 0.05, 0.07,$ and 0.08) under a longitudinal field of up to 5 T to elucidate the spin fluctuation and its relevance to the HF behavior [4]. We found that μ SR time spectra exhibit a stretched exponential damping, and the depolarization rate (λ) is mostly independent of the magnitude of the external field for $x > 0.03$, while it shows a slight variation for $x = 0.03$. Based on the conventional model of longitudinal depolarization under spin fluctuation in metals that is expressed by the previous Eq. (2), we deduced the fluctuation rate ν from λ to find that the temperature dependence of ν exhibits a power law as in the case of YMn_2Zn_{20} with a clear tendency of the power to converge to unity with increasing x . Such a behavior has been discussed in terms of the hybridized one-dimensional (1D) Hubbard chain [6] which comprises the essential basis for a microscopic model to reproduce the heavy fermion behavior in LiV_2O_4 where the heavy quasiparticles are induced by enhanced inter-chain correlation between the 1D Hubbard chains [7].

However, there is a remaining issue in the analysis mentioned above. Previously, we used the bulk magnetic susceptibility to substitute for χ in Eq. (2) in deducing the spin fluctuation rate. Since the bulk susceptibility is the sum of many different contributions averaged over the sample, the possibility of extrinsic contributions such as magnetic impurities and defects within the relevant specimen cannot be ruled out. It is difficult to separate all these different contributions in the bulk property measurements.

Muon Knight shift (K) provides useful information on intrinsic electronic properties independent of those extrinsic contributions because K monitors local magnetic susceptibility at the atomic scale. Moreover, μ SR spectra under a high transverse field (TF) will provide further information on the hyperfine interaction between muons and d -electrons.

It is expected from the known interstitial hydrogen site that the implanted muon is located at

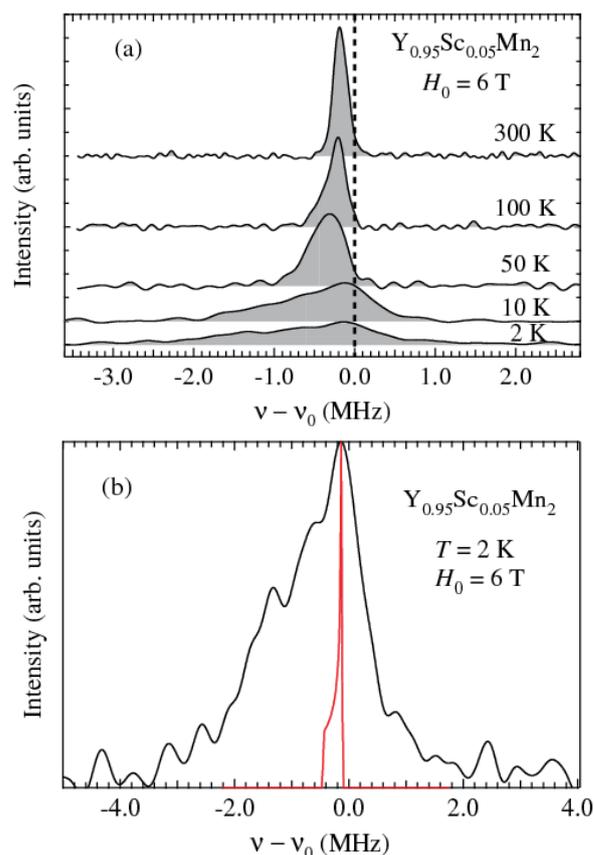


Fig. 4: a) Temperature variation of Fourier-transformed (FT) μ SR spectra observed in $Y_{0.95}Sc_{0.05}Mn_2$. (b) FT- μ SR spectrum at 2 K displayed in (a). The red line is a calculated FT spectrum for the $16c \mu^+$ site.

the $16c$ site of $Y_{1-x}Sc_xMn_2$ which has a trigonal $\bar{3}m$ point symmetry for the cubic $Fd\bar{3}m$ structure. Therefore we expect that the uni-axial powder pattern will be observed in the Fourier-transformed (FT) μ SR spectrum for the powdered sample. The powder pattern has two edges determined by the parallel and perpendicular components of the muon Knight shift, K_{\parallel} and K_{\perp} , where K_{\parallel} (K_{\perp}) is defined as the shift with an external field parallel (perpendicular) to the principal axis. In general, K_i ($i = \parallel$ and \perp) is given by $K_i = (A_i\chi_i)/N_A\mu_B$, where A_i and χ_i are parallel and perpendicular components of the hyperfine coupling tensor and local magnetic susceptibility.

In FY2013, we performed muon Knight shift measurements on a powder sample of $Y_{1-x}Sc_xMn_2$ with $x = 0.05$, using the HiTime μ SR spectrometer on the M15 beamline of TRIUMF. The result is summarized in Fig. 4(a), where the Fourier transform (FT) of μ SR spectra measured under TF = 6 T are shown for some representative points of temperature. The FT- μ SR spectrum becomes broad and asymmetric with decreasing temperature,

which is in marked contrast to a sharp single line at 300 K. In Fig. 4(b), the spectrum at 2 K is compared with a calculated powder pattern assuming no magnetic anisotropy (i.e., $\chi_{\parallel} = \chi_{\perp}$). It is clear that the observed FT spectrum is not reproduced by the calculated FT spectrum, suggesting that there might be a strong anisotropy in the local spin susceptibility due to the crystal field and/or spin-orbit coupling even in this kind of metallic compound. We also note that similar FT spectra were found in our previous μ SR study on LiV_2O_4 [8]. These features lead us to speculate that the hybridized 1D-Hubbard chain model, which is based on 1D-like bands formed by the t_{2g} orbitals [7], may be applicable to $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$. Given this situation, we are now planning detailed measurements of the muon Knight shift on a series of $\text{Y}_{1-x}\text{Sc}_x\text{Mn}_2$ to clarify the microscopic origin of anisotropic hyperfine structure.

Acknowledgements

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