Muon Knight shift in $d$-electron heavy fermion compound $Y_{0.95}Sc_{0.05}Mn_2$

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Abstract. We report on the muon Knight shift in a polycrystalline sample of $Y_{0.95}Sc_{0.05}Mn_2$ that is known as one of the $d$-electron heavy fermion compounds. Since the muon site is presumed to have a trigonal symmetry, it is expected that the Fourier-transformed (FT) spectrum line shape would exhibit a uni-axial powder pattern which has two edges determined by the parallel and perpendicular components of the Knight shift, $K_\parallel$ and $K_\perp$, where $K_\parallel$ ($K_\perp$) is proportional to the parallel (perpendicular) component of the spin susceptibility, $\chi_\parallel$ ($\chi_\perp$). The observed FT spectrum at 2 K largely disagrees with the calculated line shape in isotropic ($\chi_\parallel = \chi_\perp$) and anisotropic ($\chi_\parallel \neq \chi_\perp$) cases, suggesting that there must be field-induced staggered magnetization due to strong antiferromagnetic spin correlations.

1. Introduction

The origin of heavy fermion behavior in $d$-electron systems such as LiV$_2$O$_4$ [1] and $Y_{1-x}Sc_xMn_2$ [2, 3, 4] remains as an enigmatic issue in condensed matter physics. $Y_{1-x}Sc_xMn_2$ is a series of inter-metallic compound with the cubic C15 Laves phase structure (space group $Fd\bar{3}m$), where the Mn atoms form a three-dimensional (3D) network of corner-sharing tetrahedra called pyrochlore lattice. YMn$_2$ ($x = 0$) shows a first-order magnetic transition at $\sim 100$ K accompanied by a long-wavelength ($\sim 400$ Å) helical spin structure [5]. A large volume change ($\sim 5\%$) and small tetragonal distortion were also observed at the transition temperature [6]. These peculiar physical properties are presumed to be related with the quasi-one-dimensional antiferromagnetic (AF) correlation that is characteristic to the frustrated AF spin system on pyrochlore lattice as a manifestation of reduced dimensionality in the frustrated 3D systems [7, 8].

The magnetic ordering is suppressed by relatively low hydrostatic [9, 10] or chemical pressure due to partial substitution of Y by Sc [11]. Antiferromagnetic spin fluctuation [12, 13, 14] and large electronic contribution to the specific heat $\gamma$ ($\simeq 150$ mJ·mol$^{-1}$K$^{-2}$ [3, 15]) were found in the paramagnetic state in place of the magnetic ordering. As is widely observed in heavy fermion systems, the ratio $A/\gamma^2 \sim 1.0 \times 10^{-5}$ $\Omega$·cm (mJ/mol K)$^{-2}$ satisfies the Kadowaki-Woods relation, where $A$ is the coefficient of the $T^2$ term in the temperature ($T$) dependence of resistivity [4].

Previously, we have conducted a systematic study of $Y_{1-x}Sc_xMn_2$ (with a series of specimens with $x = 0.03, 0.05, 0.07$, and 0.08) by muon spin rotation/relaxation ($\mu$SR) under a longitudinal-field up to 5 T to elucidate the spin fluctuation and its relevance to the heavy fermion
behavior\cite{16}. The observed \( \mu \)SR time spectra exhibit a stretched exponential-damping, and the depolarization rate (\( \lambda \)) is mostly independent of the magnitude of the external field for \( x \gtrsim 0.05 \), while it shows a slight variation for \( x = 0.03 \) due to slowing down of spin fluctuation towards spin glass state at lower temperatures. Based on the conventional model of metals, \( \lambda \) is given by an equation,

\[
\lambda(\nu, H_0) = \frac{2(\gamma_\mu A_\mu)^2 \nu}{\nu^2 + (\gamma_\mu H_0)^2} k_B T \frac{\chi}{N A_H B^2},
\]

where \( \gamma_\mu \) (\( = 2\pi \times 135.539 \) MHz/T) is the muon gyromagnetic ratio, \( A_\mu \) is the hyperfine coupling constant, \( \nu \) is the fluctuation rate of \( A_\mu \), and \( H_0 \) is the external field. We have found that \( \nu \) deduced from \( \lambda \) using eq. (1) exhibits a power low, \( \nu \propto T^\alpha \), at lower temperatures (below 20\textdegree\textasciitilde40 K). More interestingly, \( \alpha \) approaches unity as the compound goes away from instability to spin glass state with increasing \( x \). Such \( T \)-linear dependence of the fluctuation rate is also reported in Li\textsubscript{2}O\textsubscript{4} \cite{17, 18} and discussed in terms of the hybridized one-dimensional (1D) Hubbard chain model\cite{19, 20}.

**Figure 1.** Fourier-transformed \( \mu \)SR spectra observed in \( Y_{0.95} Sc_{0.05} Mn_2 \) under a transverse-field of 6 T. The spectra are plotted against frequency shift from the reference, \( f - f_0 \).

However, there remains an issue in the above mentioned analysis that we rely on the bulk magnetic susceptibility for evaluating \( \chi \) in eq. (1). Since the bulk susceptibility is a sum of many different contributions averaged over the specimen, possibility of extrinsic contributions such as magnetic impurities and defects cannot be ruled out. It is often difficult to separate all these different contributions in the bulk property measurements. The muon Knight shift (\( K \)) provides useful information to extract intrinsic electronic property out of those extrinsic contributions because \( K \) monitors local magnetic susceptibility in the atomic scale.

2. **Experiments**

Polycrystalline sample of \( Y_{0.95} Sc_{0.05} Mn_2 \) was prepared by arc melting in an argon atmosphere. The prepared compound was annealed for a week in a evacuated quartz tube at 800\textdegree C. \( \mu \)SR measurements under a transverse-field (TF) of 6 T were conducted at M15 beamline of TRIUMF, Canada. The shift was determined by the spin rotation frequency observed in the sample (\( f \)) and that in a reference silver plate (\( f_0 \)) as

\[
K = \frac{f - f_0}{f_0},
\]
where a care was taken to keep the muon stopping condition unchanged in replacing the sample to the silver plate.

3. Results and discussion

Typical examples of Fourier-transformed (FT) $\mu$SR spectra at various temperatures are shown in Fig. 1, where the spectra are plotted against the frequency shift from the reference, $f - f_0$, observed a sharp and symmetric single line above $\sim 125$ K. YMn$_2$ is known as a hydrogen-absorbing alloy, and the hydrogen site is identified to be 16$c$ site by $^1$H NMR experiments [21]. It is then naturally expected that muon also occupies the 16$c$ site in Y$_{1-x}$Sc$_x$Mn$_2$ as a hydrogen isotope (see Fig.2). Since the muon site has a trigonal $\bar{3}m$ point symmetry for cubic $Fd\bar{3}m$ structure, it is predicted that a uni-axial powder pattern will be observed in the FT $\mu$SR spectrum measured in polycrystalline samples. However, as shown in Fig. 1, no such fine structure is observed in the spectra above 125 K. This indicates that the anisotropy of $A_\mu$ is small and that the detailed structure of the spectra is masked by the natural line width determined by the muon decay lifetime.

Below $\sim 100$ K, the observed FT spectra gradually become broad and asymmetric with decreasing $T$. The uni-axial powder pattern has two edges determined by the parallel and perpendicular components of the Knight shift, $K_\parallel$ and $K_\perp$, where $K_\parallel$ ($K_\perp$) is defined as the Knight shift with an external field parallel (perpendicular) to the principal axis. They are generally related with the local magnetic susceptibility by the equations

$$K_\parallel = \frac{A_\mu^\|}{N_A\mu_B} \chi_\|, \quad K_\perp = \frac{A_\mu^\perp}{N_A\mu_B} \chi_\perp,$$

(3)

where $A_\mu^\|$, $A_\mu^\perp$, and $\chi_\|$, $\chi_\perp$ are the corresponding hyperfine parameters and the local magnetic susceptibility per 1 mol Mn atoms, respectively. Since the sign of $A_\perp$ is opposite to $A_\parallel$, $A_\perp = -\frac{1}{2}A_\parallel$, the line width increases with an enhancement of $\chi$. The FT spectrum at 2 K and calculated powder patterns are displayed in Fig. 3, where the red broken line represents an ideal uni-axial powder pattern at 2 K without magnetic anisotropy (i.e., $\chi_\parallel = \chi_\perp$). The hyperfine fields are obtained by taking a summation of the classical dipole fields from 1$\mu_B$ Mn moments at the nearest-neighbor six Mn atoms. Generally, FT $\mu$SR spectrum has unavoidable line broadening due to the limitation of time window and finite muon lifetime. The line broadening is estimated to be less than $\sim 0.2$ MHz from the FT spectra at 300 K. Thus the observed spectra at 2 K is significantly broader than the calculated ones.

One possibility is that the broadening comes from local magnetic anisotropy. In general, bulk average of spin susceptibility ($\chi_{\text{spin}}$) is given as the average of the three principal values of

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Figure 2. Local atomic structure of YMn$_2$ with muon shown at the 16$c$ site.
local magnetic susceptibility, i.e., $\chi_{\text{spin}} = \frac{1}{3}\chi_\parallel + \frac{2}{3}\chi_\perp$ in the case of uni-axial anisotropy. Then, we can simulate the influence of anisotropy by changing $\chi_\parallel$ and $\chi_\perp$ with the above mentioned constraints. For instance, the blue broken line in Fig. 3 shows a calculated spectrum assuming $\chi_\parallel/\chi_\perp = 10$. It is clear that the observed FT spectrum is not reproduced by the calculated FT spectrum. Although we can calculate more anisotropic FT spectra with $\chi_\parallel/\chi_\perp > 10$, the rotation frequency for lower edge of the spectrum is limited to $f - f_0 \sim -1$ MHz because the $\chi_\parallel$ can not exceed the $\chi_{\text{spin}}$ value. Then, the observed spectra is not explained only by taking into the magnetic anisotropy.

These facts suggest the appearance of staggered moments due to strong AF correlation that does not contribute to the uniform magnetic susceptibility even though the local magnetizations are enhanced. We made curve-fit analysis of time dependent asymmetry assuming two components,

$$A(t) = A_0 \exp(-i\phi_0 t) \sum_{i=1}^{2} a_i \exp(2\pi f_i t - \lambda_i t), \quad (4)$$

where $A_0$ is the initial muon-positron decay asymmetry, $\phi_0$ is the initial phase, $a_i$ is the fractional yield, $f_i$ is the precession frequency, and $\lambda_i$ is the relaxation rate. Fig. 4 shows the temperature dependence of the observed Knight shift $K_i$ ($i = 1, 2$) determined from $f_i$ as $K_i = (f_i - f_0)/f_0$. Below $\sim 100$ K, $K_1$ decreases with $T$ whereas $K_2$ shows increasing behavior, indicating that the sign of the internal field for $K_1$ and $K_2$ are mutually opposite (i.e., negative for $K_1$ and positive for $K_2$).

Here, we briefly discuss the origin of unusually broad line shape shown in Fig. 3. It is expected that a characteristic quasi-one-dimensional AF correlation would develop in $Y_{1-x}\text{Sc}_x\text{Mn}_2$\cite{7,8}. In this situation, two types of internal field associated with the staggered moments may be generated at the presumed muon site; it accompanies six nearest-neighboring Mn atoms, where a half of the implanted muons are surrounded by four-up and two-down spins (4U-2D) while those for another half are next to two-up and four-down spins (2U-4D). It is naturally expected that the sign of the internal fields is opposite between these two sites. While no quasistatic field is expected from those moments, the spin fluctuation may become slow enough at lower temperatures in comparison with the time window of the present $\mu$SR experiments so that the
line shape might be described as a sum of the two types of powder patterns. This assumption is also supported by the shoulder-like structure of the FT spectra in the $T$ range between 50 K and 20 K. In such a situation, the $K_{\parallel}$ and $K_{\perp}$ are given as,

$$K_{\parallel} = \frac{2A_{\parallel}m_{\uparrow} + A_{\parallel}m_{\downarrow}}{3H_0}, \quad K_{\perp} = -\frac{7A_{\parallel}m_{\uparrow} - A_{\parallel}m_{\downarrow}}{12H_0}, \quad (5)$$

for the 4U-2D site, and

$$K_{\parallel} = \frac{A_{\parallel}m_{\uparrow} + 2A_{\parallel}m_{\downarrow}}{3H_0}, \quad K_{\perp} = \frac{A_{\parallel}m_{\uparrow} - 7A_{\parallel}m_{\downarrow}}{12H_0}, \quad (6)$$

for the 2U-4D site, where $m_{\uparrow}$ and $m_{\downarrow}$ are the magnitude of the Mn moments with spin up and down, respectively. Then, we can simulate the line shape by varying $m_{\uparrow}$ and $m_{\downarrow}$ as parameters. We note that the model exhibits reasonable agreement with the FT spectrum in Fig. 3 for $m_{\uparrow} = 0.02 \sim 0.03\mu_B$ and $m_{\downarrow} = -0.005 \sim -0.006\mu_B$. It is confirmed that the uniform magnetization per Mn atom, which is deduced from the present results as $(m_{\uparrow} + m_{\downarrow})/2 = 0.007 \sim 0.009\mu_B$, is consistent with the bulk magnetization.

**Figure 4.** Temperature dependence of the muon Knight shift for $Y_{0.95}Sc_{0.05}Mn_2$.

Finally, we stress that a similar broadening of TF-$\mu$SR line shape has been observed in our previous study on LiV$_2$O$_4$ [22]. These features led us to speculate that the same microscopic magnetism may be present in LiV$_2$O$_4$. Further experimental investigation such as $H_0$ and $x$ dependence of the line broadening would be useful to clarify the local magnetism of these compounds.

4. Summary
We measured the muon Knight shift in the $d$-electron heavy fermion system $Y_{0.95}Sc_{0.05}Mn_2$ under a TF of 6 T. The FT spectra gradually became broad and asymmetric with decreasing temperature, where those at lower temperatures were not reproduced by the powder pattern even for an extreme case of magnetic anisotropy as large as $\chi_{\parallel}/\chi_{\perp} = 10$. This result suggests occurrence of short-range ordered staggered moments due to a strong antiferromagnetic correlation.
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