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

Ichihiro Yamauchi[1], Masanori Miyazaki[1], Masatoshi Hiraishi[1], Akihiro Koda[1], Kenji M Kojima[1], Ryosuke Kadono[1] and Hiroyuki Nakamura[2]

[1]Muon Science Laboratory and Condensed Matter Research Center, Institute for Materials Structure Science, High Energy Accelerator Research Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan

[2]Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501, Japan

E-mail: ichihiro@post.kek.jp

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_2O_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 ∼100 K accompanied by a long-wavelength (∼400 Å) helical spin structure [5]. A large volume change (∼ 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 γ ($\simeq$ 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 $A/\gamma^2 \sim 1.0\times10^{-5}$ Ω·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 (μSR) under a longitudinal-

field up to 5 T to elucidate the spin fluctuation and its relevance to the heavy fermion behavior[16]. The observed μSR time spectra exhibit a stretched exponential-damping, and the depolarization rate (λ) is mostly independent of the magnitude of the external field for $x \geq 0.05$, 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, λ is given by an equation,

$$\lambda(\nu, H_0) = \frac{2(\gamma_\mu A_\mu)^2 \nu}{\nu^2 + (\gamma_\mu H_0)^2} k_{\rm B} T \frac{\chi}{N_{\rm A} \mu_{\rm B}^2}, \tag{1}$$

where γ_μ ($= 2\pi \times 135.539$ MHz/T) is the muon gyromagnetic ratio, A_μ is the hyperfine coupling constant, ν is the fluctuation rate of A_μ, and H_0 is the external field. We have found that ν deduced from λ using eq. (1) exhibits a power low, $\nu \propto T^\alpha$, at lower temperatures (below 20$\sim$40 K). More interestingly, α 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 LiV_2O_4 [17, 18] and discussed in terms of the hybridized one-dimensional (1D) Hubbard chain model[19, 20].

However, there remains an issue in the above mentioned analysis that we rely on the bulk magnetic susceptibility for evaluating χ 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°C. μSR 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

$$K = \frac{f - f_0}{f_0}, \tag{2}$$

where a care was taken to keep the muon stopping condition unchanged in replacing the sample to the silver plate.

3. Results and discussion

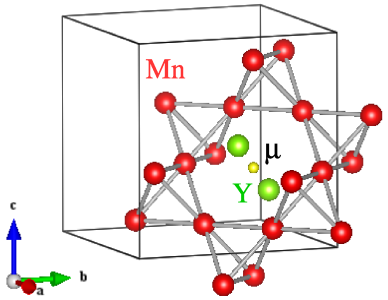

Figure 1. Local atomic structure of YMn_2 with muon shown at the 16c site.

Typical examples of Fourier-transformed (FT) μSR spectra at various temperatures are shown in Fig. 2, where the spectra are plotted against the frequency shift from the reference, $f - f_0$.

We observed a sharp and symmetric single line above ∼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 $Y_{1-x}Sc_xMn_2$ as a hydrogen isotope (see Fig.1). 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 μSR spectrum measured in polycrystalline samples. However, as shown in Fig. 2, no such fine structure is observed in the spectra above 125 K. This indicates that the anisotropy of A_μ is small and that the detailed structure of the spectra is masked by the natural line width determined by the muon decay lifetime.

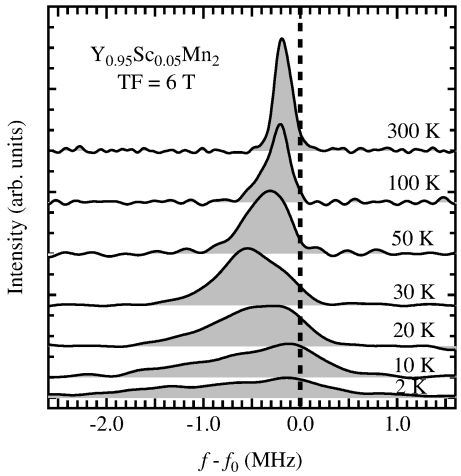

Figure 2. Fourier-transformed μSR spectra observed in $Y_{0.95}Sc_{0.05}Mn_2$ under a transverse-field of 6 T. The spectra are plotted against frequency shift from the reference, $f - f_0$.

Below ∼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

$$K_\parallel = \frac{A_\mu^\parallel}{N_{\rm A}\mu_{\rm B}}\chi_\parallel, \quad K_\perp = \frac{A_\mu^\perp}{N_{\rm A}\mu_{\rm B}}\chi_\perp, \tag{3}$$

where $A_\mu^{\parallel,\perp}$ and $\chi_{\parallel,\perp}$ 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 χ. 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 $1\mu_{\rm B}$ Mn moments at the nearest-neighboring six Mn atoms. Generally, FT μSR 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 ∼0.2 MHz from the FT spectra at 300 K. Thus the observed spectra at 2 K is significantly broader than the calculated ones.

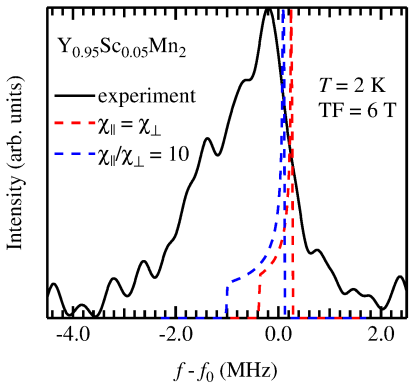

Figure 3. Fourier-transformed μSR spectrum at 2 K under a transverse-field of 6 T. The red and blue broken lines represent calculated spectra for the expected muon site (16c) assuming magnetically isotropic ($\chi_{\parallel} = \chi_{\perp}$) and anisotropic ($\chi_{\parallel} \neq \chi_{\perp}$) cases.

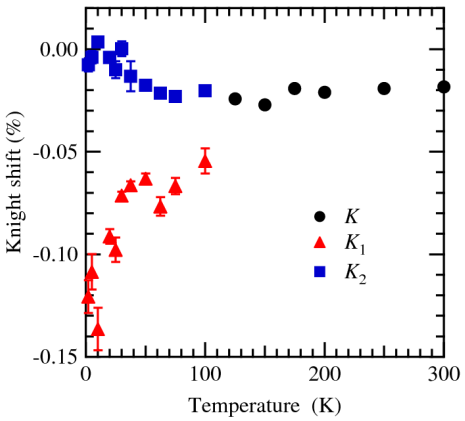

Figure 4. Temperature dependence of the muon Knight shift for $Y_{0.95}Sc_{0.05}Mn_2$.

One possibility is that the broadening comes from local magnetic anisotropy. In general, bulk average of spin susceptibility ($\chi_{\rm spin}$) is given as the average of the three principal values of local magnetic susceptibility, i.e., $\chi_{\rm spin} = \frac{1}{3}\chi_{\parallel} + \frac{2}{3}\chi_{\perp}$ 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 $\chi_{\rm spin}$ 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,

$$A_0 G(t) = A_0 \mathrm{e}^{-i\phi_0 t} \sum_{i=1}^{2} a_i \exp\left(2\pi i f_i t - \lambda_i t\right), \tag{4}$$

where A_0 is the initial muon-positron decay asymmetry, ϕ_0 is the initial phase, a_i is the fractional yield, f_i is the precession frequency, and λ_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 $Y_{1-x}Sc_xMn_2$[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 μSR experiments so that the line shape might be described as a sum of the two types of powder patters. 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,

$$K_{\parallel} = \frac{2A_{\mu}^{\parallel}m_{\uparrow} + A_{\mu}^{\parallel}m_{\downarrow}}{3H_0}, \;\; K_{\perp} = -\frac{7A_{\mu}^{\parallel}m_{\uparrow} - A_{\mu}^{\parallel}m_{\downarrow}}{12H_0}, \tag{5}$$

for the 4U-2D site, and

$$K_{\parallel} = \frac{A_{\mu}^{\parallel}m_{\uparrow} + 2A_{\mu}^{\parallel}m_{\downarrow}}{3H_0}, \;\; K_{\perp} = \frac{A_{\mu}^{\parallel}m_{\uparrow} - 7A_{\mu}^{\parallel}m_{\downarrow}}{12H_0}, \tag{6}$$

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 $m_{\uparrow} = 0.02 \sim 0.03\mu_{\rm B}$ and $m_{\downarrow} = -0.005 \sim -0.006\mu_{\rm B}$. It is confirmed that the uniform magnetization per Mn atom, which is deduced from the present results as $(m_{\uparrow} + m_{\downarrow})/2 = 0.007 \sim 0.009\mu_{\rm B}$, is consistent with the bulk magnetization.

Finally, we stress that a similar broadening of TF-μSR line shape has been observed in our previous study on LiV_2O_4 [22]. These features led us to speculate that the same microscopic magnetism may be present in LiV_2O_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.

References

[1] Kondo S, Johnston D C, Swenson C A, Borsa F, Mahajan A V, Miller L L, Gu T, Goldman A I, Maple M B, Gajewski D A, Freeman E J, Dilley N R, Dickey R P, Merrin J, Kojima K, Luke G M, Uemura Y J, Chmaissem O and Jorgensen J D 1997 *Phys. Rev. Lett.* **78** 3729

[2] Shiga M 1988 *Physica B* **149** 293

[3] Shiga M, Fujiwara K and Wada H 1993 *J. Phys. Soc. Jpn.* **62** 1329

[4] Hauser R, Bauer E, Gratz E, Dubenko I S and Markosyan A S 1994 *Physica B* **199&200** 662

[5] Cywinski R, Kilcoyne S H and Scott C A 1991 *J. Phys. : Condens. Matter.* **3** 6473

[6] Shiga M, Wada H and Nakamura Y 1983 *J. Mag. Mag. Matt.* **31-34** 119

[7] Nakamura H, Metoki N, Suzuki S, Takayanagi F and Shiga M 2001 *J. Phys. : Condens. Matter* **13** 475

[8] Nakamura H and Shiga M 2001 *J. Alloys Comp.* **326** 157

[9] Fisher R A, Ballou R, Emerson J P, Leievre-Berna E and Phillips N E 1993 *Int. J. Mod. Phys. B* **7** 830

[10] Zheng G-q, Nishikido K, Ohnishi K, Kitaoka Y and Asayama K 1999 *Phys. Rev. B* **59** 13973

[11] Nakamura H, Wada H, Yoshimura K, Shiga M, Nakamura Y, Sakurai J and Komura Y 1988 *J. Phys. F : Met. Phys.* **18** 981

[12] Shiga M, Wada H, Nakamura Y, Deportes J, Ouladdiaf B and Ziebeck K R A, 1988 *J. Phys. Soc. Jpn.* **57** 3141

[13] Ballou R, Leliévre-Berm E and Fåk B 1996 *Phys. Rev. Lett.* **76** 2125
[14] Nakamura H, Takayanagi F, Shiga M, Nishi M and Kakurai K 1996 *J. Phys. Soc. Jpn.* **65** 2779
[15] Wada H, Shiga M and Nakamura Y 1989 *Physica B* **161** 197
[16] Miyazaki M, Kadono R, Hiraishi M, Masuda T, Koda A, Kojima K M, Yamazaki T, Tabata Y and Nakamura H 2011 *J. Phys. Soc. Jpn.* **80** 063707
[17] Lee S H, Qiu Y, Broholm C, Ueda Y and Rush J J 2001 *Phys. Rev. Lett.* **86** 5554
[18] Kadono R, Koda A, Higemoto W, Ohishi K, Ueda H, Urano C, Kondo S, Nohara M and Takagi H 2012 *J. Phys. Soc. Jpn.* **81** 014709
[19] Fujimoto S 2002 *Phys. Rev. B* **65** 155108
[20] Lee J D 2003 *Phys. Rev. B* **67** 153108
[21] Fujiwara K, Ichinose K, Nagai H and Tsujimura A 1987 *J. Mag. Mag. Matter.* **70** 184
[22] Koda A, Kadono R, Ohishi K, Saha S R, Higemoto W, Matsushita Y and Ueda Y 2005 *J. Phys. : Condens. Matter.* **17** L257