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NMR study in a half-metallic system, Sr_2FeMoO_6

Koji Ohno [a], Harukazu Kato [a,*], Takashi Nishioka [a], Masahiro Matsumura [a]

[a] *Dept. of Material Science, Fac. of Science, Kochi Univ., Kochi 780-8520, Japan*

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Abstract

Mo-NMR measurements have been carried out for a half-metallic compound, Sr_2FeMoO_6, which involves a highly-polarized conduction band consisting of Mo-4*d*, Fe-3*d*, and O-2*p* electrons. The spin lattice relaxation of the Mo nuclei was measured in zero magnetic field as well as an applied field. Two relaxation components come from the domain and domain-wall fluctuations are found in zero field. The former (in-domain) component is specified with a single spin lattice relaxation rate $1/T_1$ showing a Korringa-like temperature T dependence ($1/TT_1 = $ const.) at low temperatures, which reflects the itinerant character of the 4*d* electrons. A fraction of the latter component is strongly suppressed with an applied field, which is consistent with the easy domain-wall control by the magnetic field.

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1. Introduction

The double perovskite compound Sr_2FeMoO_6 (SFMO), showing a half-metallic ferromagnetic character with the relatively high Curie temperature of about $\sim$ 400K and a low-field magnetoresistance effect at room temperature[1], has stimulated both fundamental and applied research. The half-metallicity of SFMO is described as a model where the majority spin (up-spin) band is gapped and the corresponding $3d^5$ up-spin electrons localize in Fe, while a down-spin conduction band consisting of overlapping Fe-3*d*, O-2*p*, and Mo-4*d* bands is partially occupied[1]. It implies non-vanishing 4*d* electron densities with highly polarization, in other words, itinerant down-spin magnetic moments originating from 4*d*. Several experiments including nuclear magnetic resonance (NMR)[2–5] has proven the existence of the 4*d* magnetic moments at the Mo site, which is antiparallel to the Fe moments. In this paper, in order to elucidate the microscopic nature of the 4*d* electrons showing highly-polarized itinerancy, we report Mo-NMR studies in SFMO, which would directly probe the 4*d* electrons. Spin-lattice relaxation rate ($1/T_1$) measurements were newly done. As far as we know, this is the first report of the nuclear-relaxation measurements in the compound.

2. Experiments

A polycrystalline sample of SFMO was prepared by a solid state reaction in oxygen-getter-controlled low-O_2-pressure encapsulation[6]. With X-ray diffraction (XRD) experiments, the sample used was revealed as a cubic single phase of SFMO with the lattice parameter a of 7.896 Å, which involves a considerable Fe-Mo antisite disorder; we have evaluated the antisite concentration of, at most, $\sim$ 80 % from the intensity of the superlattice XRD peak. NMR measurements at zero field and in an applied magnetic field H were performed with a phase-coherent pulsed spectrometer. The relaxation was measured by monitoring the recovery of the spin echo intensity $M(t)$ following a saturating pulse sequence, where t is the separation time between the saturating pulse and the echo sequence.

3. Results and Discussion

The spectrum measured in this experiment is shown in the inset of Fig. 1. The spectrum shape, which consists of main asymmetric resonance line at about $\sim$ 67 MHz and a low-frequency tail, is quite similar to those reported by several authors[2–5]. This implies that the hyperfine fields are less affected by the antisite contained, as previously reported[2]. The main resonance line is assigned as superposition of ^{95}Mo and ^{97}Mo lines [2–4], where the two isotope

* Corresponding author.
Email address: katoharu@cc.kochi-u.ac.jp (Harukazu Kato).

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lines are indistinguishably close to each other due to the nearly same gyromagnetic constants. In fact, we have confirmed that the main line with H is shifted to higher in frequency at the rate corresponding to the gyromagnetic constants of ^{95}Mo and ^{97}Mo, implying a Mo hyperfine field parallel to H. This is consistent with the magnetic structure of SFMO, wherein the smaller magnetic moments at the Mo site (antiparallel to H) and the larger Fe moments (parallel to H) both give rise to respective on-site hyperfine fields with the negative coupling constants[4]. Although a ^{57}Fe line is expected to appear around the main line, it is too weak in intensity (by two orders smaller than the Mo line) to be resolved[3]: the Fe line is presumably hidden with the low-frequency tail of the Mo lines and it would be negligible in the experiments.

All relaxation measurements for the Mo line have been carried out at the main peak position (denoted by an arrow in the inset of Fig. 1). Figure 1 shows the representative recovery curves, semilog plots of $1 - M(t)/M(\infty)$ vs. t. The recovery is quite different according to the presense/absense of H: the recovery data with H of 0.5 T lie on a straight line with a well-defined $1/T_1$ determined from a fit of the date by

$$1 - M(t)/M(\infty) = A\exp(-t/T_1) \quad (1)$$

where A is the prefactor, while that in zero field does not show a single-exponential-like behavior. For the fit to the data in zero field ($H = 0$), we use a multiexponential function of,

$$1 - M(t)/M(\infty) = A'_f\exp(-t/T_{1f}) + A'_s\exp(-t/T_{1s}) \quad (2)$$

with the two $1/T_1$ parameters of $1/T_{1f}$ and $1/T_{1s}$ and the corresponding prefactors A_f and A'_s, wherein "f" and "s" in the subscripts mean the fast and slow components, respectively. The resulting fit with $1/T_{1f}$, $1/T_{1s}$, A'_f and A'_s as free parameters is well-done as shown by the line in Fig. 1. Here, A'_f and A'_s gave similar values at 4.2 K.

With changing temperature T, we have evaluated $1/T_1$ from the data with $H = 0.5$ T and $1/T_{1f}$ and $1/T_{1s}$ from the data with $H = 0$. In estimating T_{1f} and T_{1s}, we imposed a constraint that $A'_f = A'_s$ according to the 4.2 K result. The T evolutions of $1/T_1$, $1/T_{1f}$ and $1/T_{1s}$ divided by T are shown in Fig. 2. While $1/TT_{1f}$ shows a strong T dependence, $1/TT_{1s}$ and $1/TT_1$ are almost constant, at least below 150 K, and their values are of the same order ($1/TT_{1s} \sim 1.9$ (s K)$^{-1}$ and $1/TT_1 \sim 1.0$ (s K)$^{-1}$, respectively). That is, it is found that a fraction of the fast relaxation component observed in the zero field case is rapidly suppressed with H, while the slow component survives with the relaxation rate rather modified. Therefore we infer that the fast component (specified by $1/T_{1f}$) comes from the nucleus in the domain walls and the slow ($1/T_{1s}$) in the domains of SFMO. This involves a picture of domain-wall control achieved even by a low-field, which is suggested as an origin of the low-field magnetoresistance effect founds in SFMO[1]. The constant behavior of $1/TT_1$ and $1/TT_{1s}$ found at low temperatures,

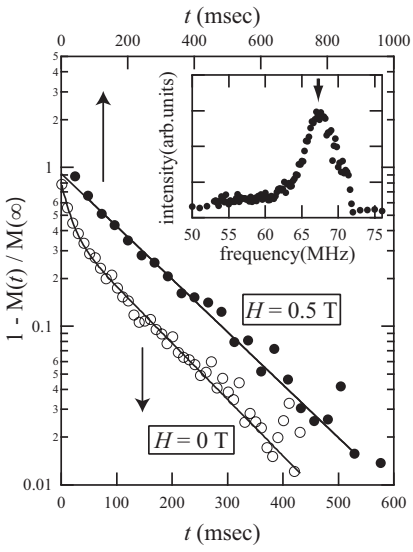

Fig. 1. Representative relaxation curves with $H = 0$ T and 0.5 T. Both the data were taken at $T = 4.2$ K. The inset shows the Mo NMR spectrum.

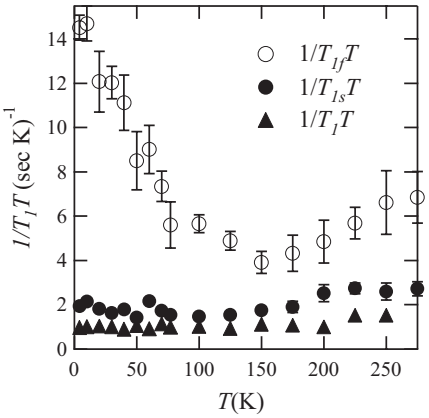

Fig. 2. Temperature T evolution of the nuclear spin lattice relaxation divided by T. The open and solid circles represent the $1/T_{1f}T$ and $1/T_{1s}T$, respectively, both of which was estimated by the data in zero field ($H = 0$). The solid triangle shows the $1/T_1T$, which is obtained with $H = 0.5$ T.

which is commonly known as the Korringa law, reflects magnetic fluctuations on the Fermi surface dominating the dynamical nature of the 4d electron. The quantitative analyses will be described elsewhere.

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