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^{73}Ge-NMR study on magnetic fluctuations of ferromagnetic superconductor UGe_2

Y. Noma[a,*], H. Kotegawa[a], T. Kubo[a], H. Tou[a], H. Harima[a], Y. Haga[b], E. Yamamoto[b], Y. Ōnuki[c], K.M. Itoh[d], E.E. Haller[e], A. Nakamura[f], Y. Homma[f], F. Honda[f], D. Aoki[f]

[a] Department of Physics, Kobe University, Kobe 657-8501, Japan
[b] Advanced Science Research Center, Japan Atomic Energy Agency, Ibaragi 319-1195, Japan
[c] Faculty of Science, University of the Ryukyus, Nishihara, Okinawa 903-0213, Japan
[d] School of Fundamental Science and Technology, Keio University, Kanazawa, Yokohama 223-8522, Japan
[e] Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720, USA
[f] Institute for Materials Research, Tohoku University, Oarai 311-1313, Ibaraki, Japan

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ABSTRACT

We report ^{73}Ge-NMR measurement on the ferromagnetic superconductor UGe_2 at ambient pressure. The observed NMR spectrum supports that the electric field gradient at three inequivalent Ge sites is correctly deduced by a LDA calculation. The temperature dependences of the nuclear spin lattice relaxation rate $1/T_1$ for $H_0 \perp a$ (easy axis) and $H_0 \| a$ were obtained for the oriented sample. The contrasting behavior in $1/T_1$ for $H_0 \perp a$ and $H_0 \| a$ reveals that the magnetic fluctuation of UGe_2 is highly anisotropic.

1. Introduction

The coexistence of ferromagnetic (FM) and superconducting (SC) states in U-based compounds such as UGe_2, URhGe, UCoGe and UIr has given a large impact. [1–4] In the first three of them, the microscopic coexistence was observed by nuclear magnetic resonance (NMR), μSR and neutron scattering. [5–8] The detailed phase diagram varies depending on the materials. In UCoGe, the SC transition temperature (T_{SC}), which is about 0.7 K at ambient pressure, goes through a maximum at the critical pressure P_c, where the transition temperature between FM and paramagnetic (PM) approaches zero due to pressure. [9] The superconductivity remains even in the PM state above P_c. In URhGe, the superconductivity also appears at ambient pressure, while T_{SC} decreases gradually with applying pressure and approaches zero inside the FM state. [10] As for the relationship between the magnetic fluctuations and superconductivity, ^{59}Co-NMR measurements for UCoGe suggest that the Ising type magnetic fluctuations strongly correlate with the superconductivity. [11,12] ^{73}Ge-NMR measurements for URhGe also show the Ising character of the magnetic fluctuations. [13] NMR is a key experiment to elucidate the relationship between the anisotropic fluctuations and superconductivity in the FM superconductors.

On the other hand, the phase diagram of UGe_2 is a unique among the FM superconductors. UGe_2 shows two types of FM phases with different size of the ordered moments: FM1 and FM2 phases. The transition between the FM1 and FM2 phases is a crossover at low pressure and changes into first order at a critical point and finally terminates at $P_x \sim 1.2$ GPa. Interestingly, the superconductivity emerges only in the FM state and T_{SC} becomes a maximum at P_x. In the previous NMR measurement for UGe_2, which was performed at ambient pressure, [14] however, the anisotropy of the magnetic fluctuations was not clarified from the nuclear spin lattice relaxation rate $1/T_1$. Therefore, we carried out ^{73}Ge-NMR measurements to investigate the detail of the magnetic fluctuations in UGe_2.

2. Experiment

We have performed ^{73}Ge NMR measurements using a polycrystalline sample with enriched ^{73}Ge, which was crushed into powder and annealed at a high temperature of ~ 900 °C to reduce a deformation. In order to investigate the magnetic field direction dependence of $1/T_1$, the sample was oriented along a axis and fixed in paraffin. The nuclear spin and the gyromagnetic ratio of ^{73}Ge are $I = 9/2$ and $\gamma_n/2\pi = 1.4852$ MHz/T, respectively.

3. Results

Fig. 1(a) shows the crystal structure of UGe_2, which belongs to the

* Corresponding author.
E-mail address: y_noma@stu.kobe-u.ac.jp (Y. Noma).

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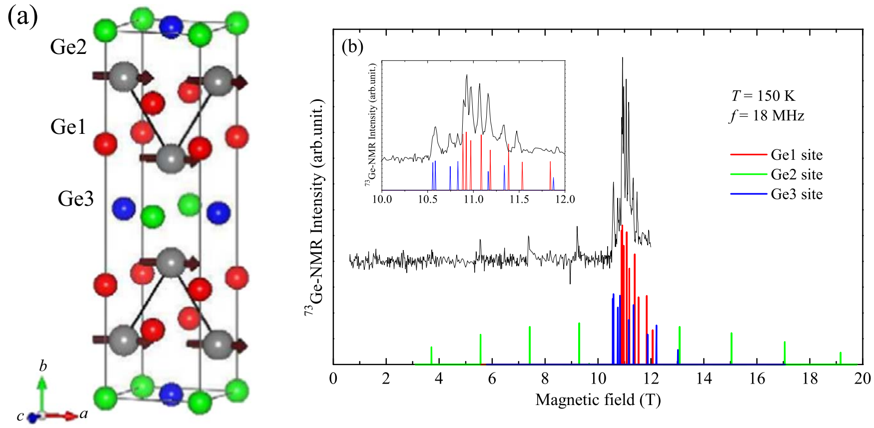

Fig. 1. (a)The crystal structure of UGe_2. Equivalent U atoms from the zigzag chain along the *a* axis, and the ordered moments are directed along the *a* axis. The Ge atoms are divided into crystallographically inequivalent three Ge sites. (b) ^{73}Ge-NMR field-swept spectrum of UGe_2 for the sample oriented along $H_0||a$ at $T = 150$ K. The inset is the enlarged figure of the spectrum in the field range between 10–12 T. The colored lines indicate the simulation for the each Ge site. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

Table 1
The directions of the principal axes and the values of η and ν_Q and Knight shift at each Ge site from the band calculation and experiment.

			LDA calculation					Experiment		
Site	Wyckoff	Local symmetry	V_{zz}	V_{yy}	V_{xx}	ν_Q (MHz)	η	ν_Q (MHz)	η	Knight shift (%)
Ge1	4*i*	*m2m*	*b*	*c*	*a*	2.36	0.953	2.29	0.967	0.059
Ge2	2*a*	*mmm*	*a*	*b*	*c*	3.48	0.678	3.11	0.818	0.07
Ge3	2*c*	*mmm*	*c*	*b*	*a*	3.63	0.719	3.29	0.915	0.047

orthothombic one with the space group *Cmmm*. UGe_2 has three crystallographically inequivalent Ge sites and the number of Ge1 sites is twice as large as the others sites in one unit cell. The respective values of the quadrupole frequency ν_Q and the asymmetry parameter η at Ge1, Ge2, and Ge3 sites were calculated through an full-potential LAPW (linear augmented plane wave) calculation within the LDA (local density approximation), shown in Table 1. The direction of the maximum principal axis of the electric field gradient (EFG), V_{zz}, is different depending on the Ge site, as shown in Table 1. It should be noted that V_{zz} is parallel to the easy *a* axis only at the Ge2 site. Fig. 1(b) shows the field-swept NMR spectrum measured for $H_0||a$ at 150 K up to 12 T. For $I = 9/2$, nine peaks are observed for one Ge site, and thus 27 peaks are expected to appear. Most of the resonances are observed between 10.5 – 12 T, while the equally separated three or four peaks are seen below 10 T. It is considered that those peaks are arised from the transitions for the Ge2 site, because the satellite transitions are clearly separated when $H_0||V_{zz}$. The transitions for the Ge1 and Ge3 sites are expected to cause the complicated spectrum in the vicinity of 11 T due to $H_0 \perp V_{zz}$. Taking the directions of the principle axes of EFG into account, we reproduced the experimental spectrum as shown in Fig. 1(b). Here, we used the EFG parameters for three Ge sites, which have already been determined from the NQR experiments shown in Table 1. [15] The good reproduction of the spectrum ensures our site assignment and the validity of the calculation.

In order to clarify the magnetic fluctuations in UGe_2, the nuclear spin lattice relaxation rate $1/T_1T$ was measured for $H_0 \perp a$ and $H_0||a$. For both cases, $1/T_1T$ was measured at the position of the central transition, although the various transitions are most likely mixed at the position. It is hard to measure $1/T_1T$ at the position consisting of the separated transition due to the weakness of the signal intensity. The magnetic field wes fixed at ~ 9.0 T for $H_0||a$, and the measurement was done in a field range of 9.7 – 11.7 T for $H_0 \perp a$. In general NMR case, the nuclear spin is relaxed by using magnetic fluctuations perpendicular to the nuclear quantization axis, as follows.

$$\frac{1}{T_1} = \frac{\gamma_n^2}{2} \int_{-\infty}^{\infty} < \delta H^-(t)\delta H^+(0) > \exp(-i\omega_n t)dt, \quad (1)$$

where $<\delta H^-(t)\delta H^+(0)>$ is a time correlation function for magnetic fluctuations perpendicular to the nuclear quantization axis, and γ_n and ω_n are the gyromagnetic ratio and resonance frequency. In this NMR case, the nuclear quantization axis is almost same as the direction of the magnetic field for any Ge sites. $1/T_1T$ for $H_0||a$ corresponds to the magnetic fluctuations along the *bc* plane, while $1/T_1T$ for $H_0 \perp a$ includes the magnetic fluctuations along the *a* axis. Fig. 2 shows the temperature dependence of $1/T_1T$ under the different direction of the magnetic field. For $H_0||a$, $1/T_1T$ has a broad maximum at $T \sim 70$ K, which is crossover behavior. On the other hand, $1/T_1T$ for $H_0 \perp a$ has a sharp peak at $T \sim 50 – 60$ K. The behavior in $1/T_1T$ is highly contrast against the directions of the magnetic field, and the result suggests that the

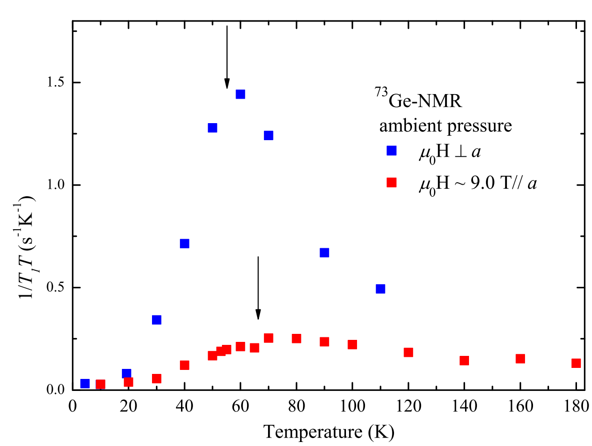

Fig. 2. Temperature dependence of $1/T_1T$ under the different direction of the magnetic field, $H_0||a$ and $H_0 \perp a$. For $H_0||a$, $1/T_1T$ has a broad maximum at $T \sim 70$ K. In contrast, $1/T_1T$ for $H_0 \perp a$ shows the sharp peak near T_{Curie}.

magnetic fluctuations along the a axis are stronger than other axes in UGe_2. $1/T_1T$ for $H_0 \perp a$ is suppressed rapidly below T_{Curie} and anisotropy are weakened below ~20 K. By contrast, $1/T_1$ for $H_0 \| a$ and b in UCoGe, which has the anisotropy of the magnetic fluctuations along c axis is nearly an order of magnitude larger than that for $H_0 \| a$ even below T_{Curie}. [11] This suggests that the anisotropy of the magnetic fluctuations deep inside the FM state in UGe_2 is weaker than that in UCoGe.

Summary

We have performed ^{73}Ge-NMR measurements for UGe_2 at ambient pressure. The spectrum for the sample oriented along the a axis is well reproduced by the simulation using the directions of principle axes deduced from the calculation and the EFG parameters were obtained in NQR measurements. $1/T_1$ for $H_0 \perp a$ is enhanced toward T_{Curie}, while that for $H_0 \| a$ is apparently smaller. This result indicates that UGe_2 has the strong magnetic fluctuations along the a axis. It is important to clarify the anisotropy of the magnetic fluctuations more accurately using a single crystal.

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