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Ga NQR relaxation rates in superconductor $PuRhGa_5$

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Abstract

Nuclear quadupole resonance (NQR) measurements have been performed on the Pu-based superconductor $PuRhGa_5$ with a transition temperature of ~8.8 K. NQR lines ascribed to $^{69}Ga(2)$, $^{71}Ga(2)$, and $^{69}Ga(1)$ have been found in zero field. Spin–lattice relaxation rate $(1/T_1)$ data for these lines have been measured in the superconducting state. The temperature variation of $1/T_1$ can be interpreted in terms of a line-nodal superconducting gap. Data for both the $^{69}Ga(2)$ and $^{69}Ga(1)$ NQR lines show a constant-T_1T behavior between T_c and T^*~30 K, which suggests that a coherent Fermi-liquid state sets in below T^*. The modified Korringa relation using $1/T_1$ and Knight shift data in an applied field indicates that the normal state of $PuRhGa_5$ can be regarded as an antiferromagnetically correlated metal.

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1. Introduction

Recently, Pu-based superconductors $PuCoGa_5$ with superconducting (SC) transition temperature $T_c = 18$ K [1] and $PuRhGa_5$ with $T_c = 8.5$ K [2] have attracted a good deal of attention owing to their relatively high values of T_c. Both $PuCoGa_5$ and $PuRhGa_5$ have the tetragonal $HoCoGa_5$ (1 1 5) crystal structure.

Recently, from NMR/NQR measurements [3–7], these Pu115 superconductors have both been found to show a *d*-wave, line-nodal SC gap. In the isostructural $CeMIn_5$ (M = Co, Ir, Rh) series, unconventional *d*-wave SC states have been identified through various experiments, although their respective T_c values are less than for the Pu115 systems, e.g. $T_c = 2.3$ K for M = Co [8], $T_c = 0.4$ K for M = Ir [9] under ambient pressure, and T_c~2 K for M = Rh under about 2 GPa [10]. On the other hand, the other actinide (Th, U, Np) 115 series have not been reported to show any superconductivity so far. They are usually Pauli paramagnets or antiferromagnets. From our study of the NMR/NQR relaxation rates for these actinide 115 compounds [11], the normal states of Pu115 systems have been found to be metals with correlations intermediate between a weakly correlated Pauli paramagnet and highly correlated systems showing long-range antiferromagnetic order. In order to further elucidate the spin fluctuation behavior of Pu115 systems, we have extended our NQR measurements on $PuRhGa_5$.

In the tetragonal 115 structure, there are two crystallographically inequivalent, 4*i* and 1*c* sites for ligand Ga (or In) atoms. The 4*i*-Ga site is labeled as Ga(1), which is coordinated in the *c* plane with four nearest-neighbor (nn) actinide atoms, and the other 1*c* site is labeled as Ga(2), which is coordinated in the *a* plane with two nn actinide and two nn transition metal atoms. It should be noted that

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this Ga(2) site has lower local symmetry than the Ga(1) site. To our knowledge, in Pu115 systems all the NQR measurements have been performed using the Ga(2) resonances. The site dependence of T_1 between Ga(1) and Ga(2) should give us the information about the anisotropy of spin fluctuations. NQR measurements and analysis for Ga(1) are now in progress.

On the other hand, since the SC properties (e.g., T_c, H_{c2}, J_c, and so on) are well known vary through self-irradiation aging effects in ^{239}Pu115 systems. Recently, the rate of T_c decrease through aging has been reported to be ~ -0.39 K/month for ^{239}PuRhGa$_5$ [13,14]. Moreover, this aging effect is enhanced at low temperatures, because the damage incurred through self-irradiation is not dispersed microscopically.

In this brief paper, we report the finding of NQR line for the Ga(1) site, and the effects of aging on $1/T_1$ for Ga(2) in for PuRhGa$_5$.

2. Experimental

Single crystals of PuRhGa$_5$ were grown by the flux method [12]. The sample was prepared using pure ^{239}Pu isotope, which is an α-emitter (half life $\sim 2.4 \times 10^4$ years), giving a uranium recoil atom. The same sample batch of PuRhGa$_5$ was used for this NQR experiment as used previously for NMR/NQR measurements [5,7]. NMR/NQR measurements were performed using a standard pulsed spectrometer simultaneously with *in situ* ac susceptibility (χ) measurement using the NQR coil in order to check the T_c. NQR spectra were taken by the fast-Fourier-transform (FFT) technique of accumulated spin-echo signals. T_1 was measured by the inversion-recovery method.

After 14 months of aging, the sample batch was reannealed just before this NQR experiment. By this annealing, the onset T_c was fully recovered to 8.8 K. For example, in the previous NQR experiments (after one month from synthesis), the onset T_c was 8.5 K. After these NQR experiments, when about three months had elapsed after the annealing, the onset T_c had decreased to $\sim$7.7 K by the self-irradiation effect.

3. Results and discussions

Fig. 1 shows the temperature dependence of the NQR frequency (ν_{NQR}) for ^{69}Ga(1) and ^{69,71}Ga(2) in PuRhGa$_5$. During our NMR experiment in Ref. [5], the ^{69,71}Ga(1) NMR lines were very weak under applied field. Moreover, the center lines could not be distinguished from other NMR lines coming from residual Ga flux, while the satellite lines were barely observable. So we could not determine precise values for the ^{69,71}Ga(1) NQR parameters from previous NMR experiments. One reason why Ga(1) NMR signals were weak may be due to a fast spin-spin relaxation rate ($1/T_2$). However, in this experiment, the NQR line has been found near the frequency estimated roughly from NMR satellite lines. It is very likely that this NQR line near $\sim$13 MHz can be ascribed to ^{69}Ga(1). As shown in Fig. 1, the T-variation of ν_{NQR} is basically similar to the other NQR lines for PuRhGa$_5$, i.e., each NQR frequency decreases slightly as temperature increases. ν_{NQR} seems to vary with temperature as $T^{3/2}$, which is often observed in paramagnetic metals. Moreover, $1/T_1$ for this line is confirmed to decrease considerably in the SC state.

The inset of Fig. 1(a) shows the putative ^{69}Ga(1) line at 10 K in the normal state of PuRhGa$_5$. The signal-to-noise ratio was quite low compared with the ^{69,71}Ga(2) NQR lines observed at higher frequencies, as shown in the inset of Fig. 1(b) and (c). These assignments of ^{69,71}Ga(2) are also cross-checked by NMR under applied field. Further, the ratio of NQR frequencies is found to be consistently equal to the ratio of nuclear quadrupole moments (Q) in the whole temperature range.

Next, let us discuss the T_1 in the SC state of PuRhGa$_5$. Fig. 2 shows the $1/T_1$ *vs.* T plot for ^{69}Ga(2) NQR. Our

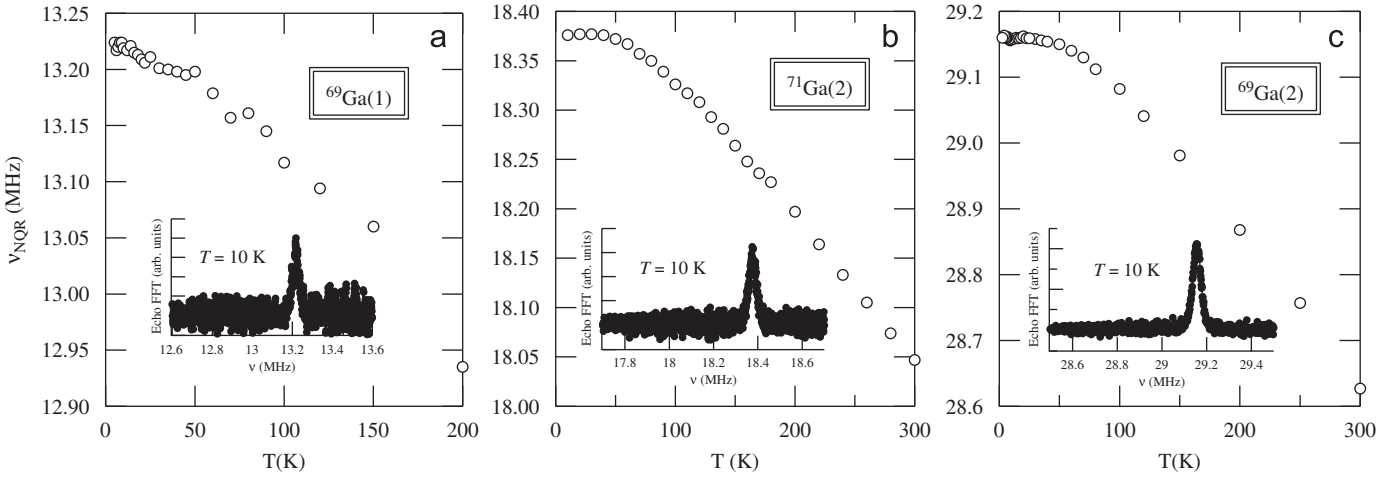

Fig. 1. Temperature dependence of NQR frequency for (a) ^{69}Ga(1), (b) ^{71}Ga(2), and (c) ^{69}Ga(2) in PuRhGa$_5$. Each inset shows the respective NQR spectrum at 10 K in zero field.