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Favorable magnetic fluctuation anisotropy for unconventional superconductivity in f-electron systems

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We report spin-lattice relaxation time measurements in isostructural ($HoCoGa_5$-type) actinide compounds with different ground states (paramagnet, antiferromagnet, and superconductor) using single-crystal samples. The anisotropy of local magnetic fluctuations due to LS coupling on the actinide (magnetic) site is evaluated. In the superconductor $PuRhGa_5$, the XY-type magnetic anisotropy is strong compared with nonsuperconducting compounds. Favorable magnetic anisotropy for d-wave superconductivity in f-electron systems is discussed.

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Recently, new superconductors have been found among Pu-based compounds. Since T_c values in these compounds are quite high [T_c=18 K in $PuCoGa_5$ (Ref. 1) and 9 K in $PuRhGa_5$ (Ref. 2)], the origin of the high T_c attracts a great deal of interest. In strongly correlated electron systems such as 4f- and 5f-electron heavy-fermion compounds and 3d-electron high-T_c cuprates, unconventional superconductivity is considered to be mediated by magnetic fluctuations.[3] In fact, NMR studies have indicated d-wave superconductivity in $PuCoGa_5$ (Ref. 4) and $PuRhGa_5$,[5] and a correlation between the characteristic magnetic fluctuation energy and T_c has been found among f-electron heavy-fermion superconductors ($T_c \sim 1$ K), including the Pu-based ones ($T_c \sim 10$ K), and cuprates ($T_c \sim 10^2$ K).[4]

However, in order to create a unified picture of magnetically mediated superconductivity in the d- and f-electron systems, the anisotropy of local magnetic fluctuations needs to be addressed as an important difference between the f-electron and d-electron systems, in addition to the scaling between the characteristic magnetic fluctuation energy and T_c. Since the LS coupling is weak in 3d systems, orbital and spin degrees of freedom are usually well separated. In fact, from neutron scattering studies in tetragonal cuprate superconductors, e.g., $La_{2-x}Sr_xCuO_4$, the spin variables are well decoupled from the orbital variables, and the local (Cu site) magnetic (spin in this case) fluctuations are almost isotropic in the paramagnetic state.[6] On the other hand, in f-electron heavy-fermion superconductors the LS coupling is strong, leading to an anisotropic magnetic fluctuation reflecting the lattice anisotropy, although there has been no systematic study on the relation between superconductivity and the anisotropy of local magnetic fluctuations up to now.

Monthoux and Lonzarich[7] discussed a qualitative relation between the anisotropy of local magnetic fluctuations and magnetically mediated superconductivity, indicating that Ising-type anisotropy is favorable for superconductivity via ferromagnetic fluctuations, whereas, in contrast, isotropic fluctuations are favorable for supercondctivity via antiferromagnetic fluctuations. This discussion indicates that the anisotropy of the fluctuations is considered to be an important factor for superconductivity mediated by magnetic fluctuations, although no actual cases are considered in their discussion.

In this Rapid communication, we evaluate the relation between local magnetic anisotropy and superconductivity in f-electron systems, and discuss possibly favorable magnetic fluctuation anisotropy for d-wave superconductivity in strongly correlated f-electron superconductors.

Compounds with the $HoCoGa_5$ (115) structure are good systems for investigating the relation between magnetism and superconductivity, since various ground states are found to occur: superconductors with low T_c ($\sim$1 K) in 4f Ce 115 systems,[8] and, in contrast, magnetically ordered systems in 5f U and Np 115 systems,[9] in addition to the Pu 115 superconductors with high T_c values. In this report, we have estimated systematically the anisotropy of magnetic fluctuation energies in actinide (A) 5f 115 compounds with different ground states: antiferromagnetic (AFM) $UPtGa_5$ (T_N=25 K), $NpCoGa_5$ (T_N=47 K), and $NpFeGa_5$ (T_N=115 K), paramagnetic (PM) $UFeGa_5$, and superconducting (SC) $PhRhGa_5$ (T_c=9 K).

Figure 1 shows the T dependence of the static susceptibility along the c axis $\chi_c(0)$. In the AFM systems $UPtGa_5$,[10] $NpFeGa_5$, and $NpCoGa_5$,[9] $\chi_c(0)$ in the paramagnetic state exhibits Curie-Weiss (CW) behavior at high temperatures,

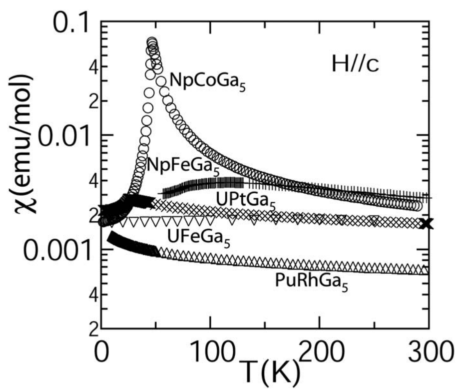

FIG. 1. T dependence of the static susceptibility $\chi_c(0)$ for $H \parallel c$ in actinide 115 compounds.

which is expected for localized-moment systems. In contrast, the ordered moment is considerably smaller, i.e. $0.3\mu_B$ for $UPtGa_5$, $0.86\mu_B$ for $NpFeGa_5$, and $0.7\mu_B$ for $NpCoGa_5$,[11] reflecting their itinerant nature. These facts imply that these AFM compounds have localized character at high temperature, becoming itinerant at low temperature, and finally having an itinerant ordered state.[12] The magnetic structure of the ordered state has a propagation vector $Q=(0,0,1/2)$, with an ordered moment parallel to [001] in $UPtGa_5$ and $NpCoGa_5$, while, in contrast, in $NpFeGa_5$ $Q=(1/2,1/2,0)$ with an ordered moment parallel to [110].[11] This difference of the ordered structures is also reflected in the paramagnetic state fluctuations as discussed below.

In PM $UFeGa_5$,[13] $\chi_c(0)$ is small and has the weak T dependence expected for an itinerant Pauli paramagnet.

In SC $PuRhGa_5$,[14] it is difficult to estimate the intrinsic T dependence of $\chi_c(0)$, particularly at low temperatures, owing to the presence of magnetic impurities. In any case, CW behavior is observed above 30 K.

The preparation and bulk property measurements of the samples have been presented previously.[9,10,13] Of the two crystallographically inequivalent Ga sites, Ga(1) (1c) and Ga(2) (4i),[5] NMR results for the Ga(2) sites are discussed in this paper. ^{69,71}Ga NMR measurements have been performed using a conventional pulsed spectrometer with a 12 T superconducting magnet. Spin-lattice relaxation time (T_1) data were also obtained using digital averaging of spin-echo signals. The relaxation times T_1 have been determined with applied magnetic field $H\|[001]$ and $H\|[100]$. From comparison of T_1 for the ^{69}Ga and ^{71}Ga isotopes, the fluctuations are known to be magnetic in the paramagnetic state up to 300 K in all the compounds. Hereafter, ^{69}Ga(2) NMR results are presented.

Since the principal axis $\vec{n}_{ZZ}$ of the electric field gradient is perpendicular to the [010] plane at the Ga(2) site, when $H\|[100]$ there are two magnetically inequivalent Ga(2) sites: the Ga(2a) site ($\vec{n}_{ZZ}\|H$) and Ga(2b) site ($\vec{n}_{ZZ}\perp H$). The Knight shift K and $1/T_1T$ have been measured for both sites, i.e., K_α and $1/T_1T_{H\|\alpha}$ [$\alpha=a$ for the Ga(2a) site and $\alpha=b$ for the Ga(2b) site], in addition to K_c and $1/T_1T_{H\|c}$ for the case of $H\|[001]$ ($\alpha=c$).

We define the spin-lattice relaxation rate $R_\alpha \equiv \gamma_n^2 \Sigma_q A_\alpha(q)^2 \mathrm{Im}\,\chi_\alpha(q,\omega_n)/\omega_n$ ($\alpha=a,b,c$). Then $1/(T_1T)_{H\|a} = R_b+R_c$, $1/(T_1T)_{H\|b}=R_c+R_a$, $1/(T_1T)_{H\|c}=R_a+R_b$, where γ_n is the gyromagnetic ratio, $A(q)$ is the hyperfine coupling constant, and $\mathrm{Im}\,\chi_\alpha(q,\omega_n)$ is the dissipative component of dynamical susceptibility at NMR frequency ω_n. Using the above three equations, $R_{a,b,c}$ are then obtained from experimental data for $1/(T_1T)_{H\|a,b,c}$. Magnetic fluctuations at the A site dominate $\mathrm{Im}\,\chi_\alpha(q,\omega_n)$, which affects the Ga site through the transferred hyperfine coupling constant $A(q)$, as discussed below. Therefore, the magnetic fluctuations at the A site can be characterized through the Ga NMR measurements.

Figure 2 shows the temperature dependence of the spin-lattice relaxation rate R_a and R_c in the paramagnetic state of actinide 115 AFM, PM, and SC compounds.

In AFM $UPtGa_5$, $NpFeGa_5$, and $NpCoGa_5$, $R_{a,c}$ is large, and the static susceptibility shows CW-like behavior. This means that the dynamical susceptibility has no strong q dependence in the paramagnetic state at high temperature, which indicates localized behavior for these compounds.

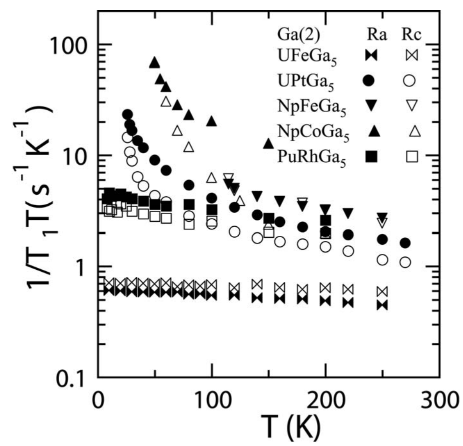

FIG. 2. T dependence of spin-lattice relaxation rates R_a and R_c at ^{69}Ga(2) site in the paramagnetic state of actinide 115 compounds.

In PM $UFeGa_5$, $R_{a,c}$ have small T dependences compared with the other compounds, i.e., $T_1T\sim$const, which is usually expected for itinerant systems. The extended Korringa relation for ligand sites,[15] $1/T_1T=4\pi k_B\hbar^{-1}n^{-1}(\gamma_n/\gamma_e)^2K(\alpha)K_s^2$, where K_s is the spin part of the Knight shift, and $n\sim 4$ is the number of neighboring magnetic sites for the Ga(2) site. The exchange enhancement parameter $K(\alpha)$ is estimated to be ~ 1, if the observed Knight shift K is used as K_s. Principally, $K(\alpha)>1$ for AFM systems, but $K(\alpha)<1$ for a FM one. As the orbital part K_{orb} of the Knight shift cannot be separated precisely from the total observed shift $K=K_{orb}+K_s$,[16] it is difficult to distinguish AFM from FM correlations here. In any case, $UFeGa_5$ appears to have a typical itinerant ground state with weak magnetic correlations.

Estimates suggest $K(\alpha)\gg 1$ for $PuRhGa_5$,[17] indicating considerable AFM correlations in this compound. In addition, the T dependence of $R_{a,c}$ in SC $PuRhGa_5$ can be regarded as intermidiate behavior between those of AFM and PM, indicating that the superconductivity is considered to appear on the border between strongly magnetic localized and weakly magnetic itinerant systems.

In the present paper, we focus on T_1 for the Ga(2) site, since fluctuations over a wider extent of q space can be detected compared with Ga(1) or the transition metal sites, owing to the lower symmetry of the Ga(2) site.[18] In addition, we focus on the paramagnetic state where $\mathrm{Im}\,\chi_\alpha(q,\omega_n)$ has no strong q dependence.[18] We therefore ignore the q dependence of $\overline{A(q)^2 \mathrm{Im}\,\chi_\alpha(q,\omega_n)}$, taking $\Sigma_q A(q)^2 \mathrm{Im}\,\chi_\alpha(q,\omega_n) \sim [A(0)^2/n]\mathrm{Im}\,\chi_\alpha(q,\omega_n)$ (the overbar means the q average) for the Ga(2) site, where $A(0)$ has been estimated from the slope of the so-called K-χ plot. Then, using the usual Lorentzian approximation for $\mathrm{Im}\chi_\alpha(q,\omega_n)$ with the magnetic fluctuation energy $\Gamma(q)$, $\mathrm{Im}\chi_\alpha(q,\omega_n)/\omega_n=\chi_\alpha(q)/\Gamma_\alpha(q)$, the relation for the strongly correlated limit approximation,[19] $2\pi\chi_\alpha(q)\Gamma_\alpha(q)\sim 1$, and $n\sim 4$, we find the relation between R_α and the q-averaged (local) magnetic fluctuation energy at the A site,

TABLE I. Hyperfine coupling constants $A_\alpha(0)$ at the Ga(2) site ($\alpha=a,b,c$ axes) in kOe/μ_B and anisotropy of Γ_α at 150 K in actinide 115 compounds. For comparison, the estimated Γ_c/Γ_a in $CeCoIn_5$ at 10 K based on ^{115}In NMR results is shown (Ref. 17).

Compound	$A_a(0)$	$A_b(0)$	$A_c(0)$	Γ_c/Γ_a
$UFeGa_5$ PM (Ref. 18)	17	19	14	0.72
$UPtGa_5$ AFM (Ref. 16)	17	20	10	0.68
$NpCoGa_5$ AFM (Ref. 20)	44	38	25	0.95
$NpFeGa_5$ AFM (Ref. 21)	16	21	30	1.7
$PuRhGa_5$ SC (Ref. 17)	−3	−1.5	21	8.1
$CeCoIn_5$ SC (Ref. 22)				1.5

$$R_\alpha = \frac{\gamma_n^2 A_\alpha(0)^2}{8\pi\Gamma_\alpha^2}, \quad (1)$$

where $\Gamma_\alpha \equiv \overline{[\Gamma_\alpha(q)^2]}^{1/2}$.

It should be noted that, if we estimate Γ_α using the approximation $\chi_\alpha(q)/\Gamma_\alpha(q) \sim \chi_\alpha(0)/\Gamma_\alpha(q)$, instead of the strongly correlated limit approximation, the discussion below does not change. Although R_a is different from R_b experimentally, due to a difference between $A_a(0)$ and $A_b(0)$, Γ_a has been confirmed to be the same as Γ_b, consistent with the tetragonal local symmetry of the A site in the paramagnetic state. Therefore we discuss only R_a and R_c below.

In order to obtain the T dependence of $\Gamma_{a,c}$, $A_\alpha(0)$ has been estimated from a plot of K_α vs $\chi_\alpha(0)$ ($\alpha=a,b,c$). Since these values are considerably larger than the dipolar fields, the main contribution comes from the transferred hyperfine field due to hybridization between $5f$ and Ga $4s,4p$ orbitals. In $UFeGa_5$, $UPtGa_5$, and $NpFeGa_5$, the K_α-$\chi_\alpha(0)$ plot is linear in the paramagnetic state up to 300 K. On the other hand, in $PuRhGa_5$, the K_α-$\chi_\alpha(0)$ plot has an anomaly below 30 K, which may be due to a magnetic impurity contribution in $\chi_\alpha(0)$. We think the estimated $A(0)_\alpha$ values are correct, since they yield $\Gamma_a=\Gamma_b$ in all the compounds. The values of $A_\alpha(0)$ are presented in Table I. Since positive and negative values are found for $A_{a,b}(0)$, the Ruderman-Kittel-Kasuya-Yosida contribution to the transferred hyperfine coupling may be rather strong in these systems.

Using the value obtained for $A_\alpha(0)$, $\Gamma_{a,c}$ is estimated from $R_{a,c}$ and Eq. (1). Figure 3 shows the temperature dependence of Γ_a and Γ_c. In contrast with cuprate superconductors, in all these compounds Γ is anisotropic.

In PM $UFeGa_5$, Eq. (1) may not be appropriate, since the correlation is not strong in this compound. At least $\Gamma_{a,c}$ is large, since the exchange enhancement is small and shows a small T dependence, which is consistent with weak correlations for this compound.

In AFM $UPtGa_5$, $NpFeGa_5$, and $NpCoGa_5$, $\Gamma_{a,c}$ show only slight T dependence at high temperature (except for Γ_c of $NpCoGa_5$), and decrease due to critical slowing down toward T_N, although the T dependence near T_N may not be precise, since the q dependence of $\mathrm{Im}\chi_\alpha(q,\omega_n)$ is ignored here. The rather T-independent behavior observed below 300 K may correspond to a crossover from the Kondo screening region

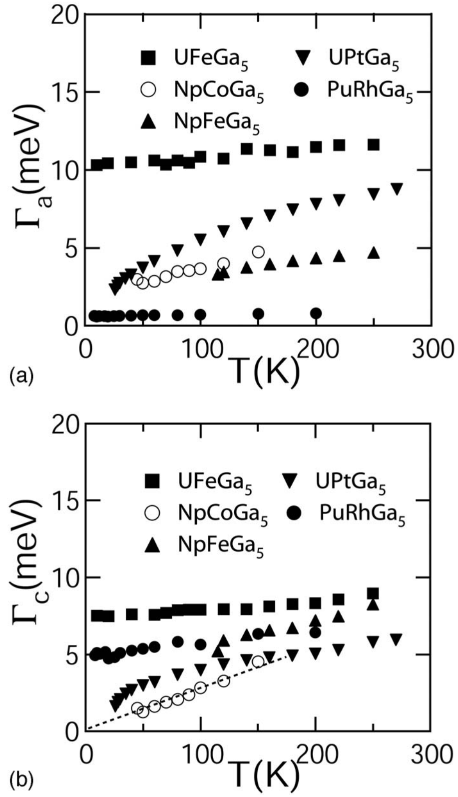

FIG. 3. T dependence of the local magnetic fluctuation energy along the a axis, Γ_a, and the c axis, Γ_c. The dashed line is a guide to the eye.

at low temperatures to the localized moment region at high temperatures, as was proposed theoretically and observed in $CeRhIn_5$ around 20 K.[22] Since the crossover temperature is proportional to $\Gamma_{a,c}$, which is ten times larger in the present $5f$ 115 compounds, it is natural that such a crossover would take place above 300 K in the present cases. In the localized moment region at high temperatures, we expect $\Gamma_{a,c} \propto T$. Such behavior would be observed above 300 K in the present AFMs. For Γ_c of $NpCoGa_5$, this behavior is already seen below 200 K, perhaps because the localized character is strong along the c axis.

We find $\Gamma_a > \Gamma_c$ in $UPtGa_5$ and $NpCoGa_5$, but $\Gamma_a < \Gamma_c$ in $NpFeGa_5$, indicating that the $5f$ moment tends to be ordered parallel to the c axis in $UPtGa_5$ and $NpCoGa_5$, whereas it is perpendicular to the c axis in $NpFeGa_5$. This tendency is consistent with the observed orientation of the ordered moments in both compounds,[11] which justifies the present estimation of $\Gamma_{a,c}$ values using Eq. (1).

In SC $PuRhGa_5$, the weak T dependence observed for $\Gamma_{a,c}$ corresponds to the above crossover region. It is remarkable that the anisotropy of this compound is particularly large compared with the other 115 compounds (Table I). Since Γ_a is considerably smaller than Γ_c, this compound has a strong XY-type magnetic anisotropy.

Theoretically, in the strongly correlated systems, p-wave superconductivity in FM systems favors Ising-type coupling, since only longitudinal fluctuations can contribute to the attractive force. In contrast, d-wave superconductivity in AFM systems favors a more isotropic coupling, since both longitudinal and transverse fluctuations can contribute to the attractive force.[7] In fact, in the *p*-wave superconductor UGe_2, the Ising FM character is quite strong.[23] From the present study of the actinide 115 systems, *d*-wave superconductivity appears only in $PuRhGa_5$, which has a strong AFM *XY*-type anisotropy. The strong AFM *XY*-type anisotropy is also found in isostructural SC $CeCoIn_5$ at temperatures below 20 K using ^{115}In NMR results[22] (Table I). Furthermore, in the tetragonal $CePd_2Si_2$ (Ref. 24) and hexagonal UPd_2Al_3 (Ref. 25) *d*-wave superconductor, the AFM ordered moment lies in the basal plane, indicating *XY*-type magnetic anisotropy. In contrast, for Ising-type antiferromagntic compounds, e.g., tetragonal $CeRu_2Si_2$,[26] no superconductivity has been found. An exception is tetragonal $CeRh_2Si_2$,[27] which has an AFM ordered moment along the *c* axis, and shows superconductivity under pressure. However, a 4*q*-complex AFM ordered phase appears at low temperatures in this compound, which suggests a frustrated magnetic interaction. Such a complexity may be the origin of the exceptional behavior.

From these observations we can suggest that, in tetragonal and hexagonal, strongly correlated *f*-electron systems, at least the *XY*-type is more favorable than Ising type for AFM *d*-wave superconductivity. In the cuprate AFM *d*-wave superconductors, quite high T_c values are observed for the isotropic Γ case, as predicted by Monthoux and Lonzarich. An interesting question is whether a strongly *XY* type is more favorable than a nearly isotropic one in tetragonal and hexagonal *f*-electron *d*-wave superconductors. In order to answer this question, we should consider the role of orbitals exhibiting *LS* coupling explicitly.[28] In the present discussion, the dimensionality of the system has been ignored, although it should be included for a more realistic treatment.

In conclusion, the anisotropy of magnetic fluctuations has been estimated systematically for isostructural actinide 115 compounds with various ground states, using spin-lattice relaxation time measurements. Compared with the non-SC compounds, the AFM *XY*-type magnetic anisotropy is found to be quite strong in $PuRhGa_5$, which shows *d*-wave superconductivity at quite a high T_c.

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