Phenomenological approach to study the degree of the itinerancy of the 5f electrons in actinide ferromagnets with spin fluctuation theory

Naoyuki Tateiwa*, Jiří Pospíšil b, Yoshinori Haga a, Hironori Sakai a, Tatsuma D. Matsuda a and Etsuji Yamamoto a

a Japan Atomic Energy Agency, 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki-ken, 319-1195, Japan; b Charles University in Prague, Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Ke Karlovu 5, 121 16 Prague 2, Czechia; c Department of Physics, Tokyo Metropolitan University, Hachioji, Tokyo, 192-0397, Japan

Actinide compounds with 5f electrons have been attracting much attention due to their interesting magnetic and electronic properties such as heavy fermion state, unconventional superconductivity, co-existence of the superconductivity and magnetism. Recently, we have reported a phenomenological analysis on 80 actinide ferromagnets with the spin fluctuation theory originally developed to explain the ferromagnetic properties of itinerant ferromagnets in the 3d transition metals and their intermetallics (N. Tateiwa et al., Phys. Rev. B 96, 035125 (2017)). Our study suggests the itinerancy of the 5f electrons in most of the actinide ferromagnets and the applicability of the spin fluctuation theory to actinide 5f system. In this paper, we present a new analysis for the spin fluctuation parameter obtained with a different theoretical formula not used in the reference. We also discuss the results of the analysis from different points of views.

Keywords: actinide ferromagnets; spin fluctuation theory; spin fluctuation parameters; uranium; neptunium; plutonium

1. Introduction

The nature of the 5f electrons in actinide compounds has been extensively studied for many years from both theoretical and experimental points of views [1-3]. Despite numerous theoretical studies, the behavior of the 5f electrons has not been fully understood yet. One of remaining issues is whether the 5f electrons should be treated as being itinerant or localized in various actinide intermetallic compounds.

Recently, we have reported the result of analyses of magnetic data in 69 uranium, 7 neptunium and 4 plutonium ferromagnets with the spin fluctuation theory developed for the magnetic properties in the 3d metals and their intermetallics [4]. The basic and spin fluctuation parameters of the actinide ferromagnets have been determined using our experimental data as well as those from literature. The analysis suggests the itinerancy of the 5f electrons in most of the actinide ferromagnets and the applicability of the spin fluctuation theory to the actinide 5f system.

In this paper, we discuss the result of the analysis from different points of views. In addition, we show a new analysis for the spin fluctuation parameters obtained with a different theoretical expression in the spin fluctuation theory not used in the reference.

2. Results and discussions

2.1. Spin fluctuation theory

We briefly summarize the spin fluctuation theory developed by Takahashi [5-7]. The spin fluctuation spectrum for itinerant ferromagnets is described by double Lorentzian distribution functions in small energy $\omega$ and wave vector $q$ spaces.

$$\text{Im} \chi(q,\omega) = \frac{\chi(0)\omega q^2}{1 + q^2/\kappa^2 + \omega^2 + \Gamma q^2}$$

Here, $q = |\mathbf{q}|$, $\kappa$ represents the inverse of the magnetic correlation length, and $\Gamma_A (= \Gamma_d(\kappa^2+q^2)$) is the damping constant. The spectrum is represented in a parameterized form by introducing two energy scales $T_0 (= \Gamma_d q_B^2/2\pi)$ and $T_A (=N_0 q_B^2/(2\chi(0)\kappa^2))$, where $q_B$ is the zone-boundary wave vector for the crystal with $N_0$ magnetic atoms with the volume $V (=6\pi^2N_0/q_B^3)$. The parameters $T_0$ and $T_A$ represent the distribution widths of the spin fluctuation spectrum in the energy and wave-vector spaces, respectively.

In the Takahashi’s spin fluctuation theory, $T_0$ and $T_A$ are expressed in following relations.

$$T_0 = \frac{p_i^2}{5g^2C_{4\Delta}} \left( \frac{15\Delta}{2T_C} \right)^{1/2}$$

$$T_A = \frac{p_i^2}{5g^2C_{4\Delta}} \left( \frac{2T_C}{15\Delta} \right)^{1/3}$$

Here, $g$ represents Landé’s g factor and $C_{4\Delta}$ is a constant ($C_{4\Delta} = 1.006089\cdots$). $p_i$ is the spontaneous magnetic moment and $T_C$ is the ferromagnetic transition
temperature. $F_1$ is the mode-mode coupling term: the coefficient of the $M^4$ term in the free energy that can be evaluated experimentally from the inverse slope of the Arrott plots ($M^2$ versus $H/M$ plot) at low temperatures.

Generally, actinide ferromagnets have anisotropic magnetic properties. We analyzed the magnetic data taken on single crystal samples under magnetic field applied along the magnetic easy axis for most of the ferromagnets. But experimental data on polycrystalline samples have been used for several compounds. This could cause uncertainty in obtained parameter values that is reflected in error bars of the data points. Readers are referred to Ref. 4 for the details of our analysis.

2.2. Application of the spin fluctuation theory to actinide ferromagnets

In this subsection, we summarize our analysis on actinide ferromagnets with the spin fluctuation theory reported in Ref. 4. An important result in the Takahashi’s spin fluctuation theory is generalized Rhodes-Wohlfarth relation between $p_{\text{eff}}/p_s$ and $T_C/T_0$ [5-7].

$$p_{\text{eff}}/p_s \approx 1.4 \left(\frac{T_C}{T_0}\right)^{-2/3}$$  \hspace{1cm} (4)

Here, $p_{\text{eff}}$ is the effective magnetic moments in the magnetic susceptibility. This relation has been confirmed in itinerant ferromagnets of the 3$d$ electrons systems [5]. As shown in Figure 1, we find that the same relation holds in most of actinide ferromagnets for $T_C/T_0 < 1.0$ [4]. This suggests the itinerant nature of the 5$f$ electrons. Here, a ratio $T_C/T_0$ characterizes the degree of itinerancy of magnetic electrons in the spin fluctuation theory. At $T_C/T_0 << 1$, the magnetic electrons have a strong itinerant character. Both quantities approach unity when the degree of itinerancy becomes small. The local magnetic moment is responsible for the ferromagnetism when $T_C/T_0 = 1$. Note that several data points deviate from the relation for $T_C/T_0 \sim 1.0$. This deviation may be due to several other effects not included in the spin fluctuation theory such as the crystalline electric field effect on the 5$f$ electrons from ligand atoms. We note that the spin fluctuation theory neglects the orbital moment oriented antiparallel to the spin moment of the 5$f$ electrons. Further elaborate theoretical consideration is necessary for the present result.

2.3. Relation between $T_C$, $T_0$ and $T_A$

We show new result of the analysis not reported in Ref. 4. Figure 2 shows relations between $T_C$ and (a) $T_0$, and (b) $T_A$ for uranium, neptunium and plutonium compounds, and transition 3$d$ metals plotted as closed circles, squares, triangles and anti-triangles, respectively. The parameters for the 3$d$ systems are cited from Ref. 5. A general tendency is that the widths of spin fluctuation spectra in the actinide ferromagnets are about one order magnitude smaller than those in the 3$d$ systems. This suggests smaller energy scales of magnetic excitations for the actinide systems. This could be attributed to the larger spatial extent of the 3$d$ wave functions than that of the 5$f$ ones. An interesting feature is that the values of $T_A$ for the plutonium ferromagnets are generally larger than those of the uranium and neptunium compounds and the values are comparable to those of the 3$d$ systems. The spin fluctuation spectra for the plutonium ferromagnets spread to the higher momentum $q$ space, similar to the 3$d$ systems.

![Figure 1. Generalized Rhodes-Wohlfarth plot. Data points for uranium, neptunium and plutonium compounds, and transition 3$d$ metals are plotted as closed circles, squares, triangles and anti-triangles, respectively (cited from Ref. 4). Solid line is theoretical relation between $p_{\text{eff}}/p_s$ and $T_C/T_0$ in the Takahashi's spin fluctuation theory [5].](image1)

![Figure 2. Relations between $T_C$ and (a) $T_0$, and (b) $T_A$ for uranium, neptunium, and plutonium compounds, and transition 3$d$ metals plotted as closed circles, squares, triangles and anti-triangles, respectively.](image2)
2.4. Uranium ferromagnetic superconductors UGe$_2$, URhGe, and UCoGe

Many studies have been done for the superconducting properties in uranium ferromagnetic superconductors UGe$_2$, URhGe, and UCoGe since the findings of the superconductivity [8-11]. Meanwhile, there have been only a few studies for the dynamical magnetic property of the ferromagnetic superconductors. Figure 3 shows relations between the spontaneous magnetic moment $p_s$ and $T_c/T_0$ for UGe$_2$, URhGe, and UCoGe. The data points for several itinerant ferromagnets in the 3d system are also plotted. In this figure, right and lower region corresponds to the local moment system, and the left and upper region does the weak ferromagnetic state. The dynamical magnetic property differs depending on each ferromagnetic superconductor. The value of UGe$_2$ is 0.571, indicating that this compound is located close to the local moment system ($T_c/T_0 = 1.0$). On the other hand, the value for UCoGe is 0.0065, suggesting the weak ferromagnetic state similar to those in Y(Co$_{1-x}$Al$_x$)$_2$. The values of the parameters ($T_0 = 362$ K and $T_A = 5.92 \times 10^3$ K) in UCoGe are significantly larger than those ($T_0 = 92.2$ K and $T_A = 442$ K) in UGe$_2$. The spin fluctuation spectrum Im$\chi(q,\omega)$ in UCoGe spreads to the higher energy and momentum spaces. URhGe is located in an intermediate region between the two limiting cases. The value of $T_c/T_0$ for URhGe is 0.121, similar to those in MnSi ($T_c/T_0 = 0.131$) and Ni ($T_c/T_0 = 0.237$).

2.5. Determination of $T_A^*$ from the $M(T)$-$T$ curve

In Ref. 4, the value of the spin fluctuation parameter $T_A$ was determined from Eqs. (2) and (3) with the mode-mode coupling term $F_1$ that is obtained from the slope of the magnetization at low temperatures [4]. Here, we estimate $T_A$ of uranium ferromagnets classified as “group I” in Ref. 4 by a different theoretical formula in the Takahashi’s spin fluctuation theory [7]. The spontaneous magnetization $M(T)$ of itinerant ferromagnets is expressed by a following relation in the theory.

$$\left(\frac{M(T)}{M_0}\right)^2 = 1 - \frac{a_0}{p_s^2} \frac{T}{T_A^*}$$  \hspace{1cm} (5),

where $M_0$ is the spontaneous magnetization at 0 K, $a_0$ is a constant 50.4, and $p_s = M_0/\mu_B$. The value of $T_A^*$ can be determined by the fit of the data $M(T)$ with Eq. (5). Figure 4 shows temperature dependencies of the magnetization for UGe$_2$, UCu$_2$Ge$_2$, URh$_2$Ge$_2$, URhAl, URhGe$_2$, URhSi, UIr, and URhGe in magnetic fields of 0.01 ~ 0.5 T applied along the magnetic easy axes. The Curie temperatures $T_C$ are denoted by arrows. Solid lines are result of fits to the data with Eq. (5). Table 1 shows the values of $T_A^*$ for the uranium ferromagnets in the group I determined in this method. The values of $T_A^0$ and $T_A^*$ determined from the mode-mode coupling term $F_1$ using Eqs. (2) and (3) are also shown in the table. Generally, the values of $T_A^0$ and $T_A^*$ obtained by the two different methods are consistent with each other. This suggests the effectiveness of the theoretical expression (Eq. (5)). However, there is difference between $T_A^0$ and $T_A^*$ for UCu$_2$Ge$_2$, URh$_2$Co$_2$Ge$_2$, and URh$_2$Co$_2$Ge$_3$ where the parameters are larger than those of the other ferromagnets. The coefficient of the $T^2$ term in Eq. (5) becomes smaller for the larger value of $T_A^*$. The discrepancy may arise from ambiguity in the determination of $T_A^*$ with Eq. (5). Note that the lowest temperature for the magnetic measurement is 2.0 K.
We discuss relations between the spontaneous magnetic moment and the magnetic excitation spectra of the actinide ferromagnets. The analysis also suggests the smaller energy scales of the electrons in most of the actinide ferromagnets.

3. Conclusion

We have analyzed 80 actinide ferromagnets using the spin fluctuation theory and found the itinerancy of the 5f electrons in most of the actinide ferromagnets. The analysis also suggests the smaller energy scales of the magnetic excitation spectra of the actinide ferromagnets. We discuss relations between the spontaneous magnetic moment and the spin fluctuation parameter in uranium ferromagnetic superconductors UGe$_2$, URhGe, and UCoGe. We determine the parameter $T_A$ using a different theoretical formula not used in our previous study [4]. The obtained values of $T_A$ are basically consistent with $T_A^0$ determined using the mode-mode coupling constant $F_0$. The magnetization data at very low temperatures may be necessary for the accurate determination of $T_A$ when it is larger.

Acknowledgements

This work was supported by JSPS KAKENHI Grant Numbers 16K05463, 16K05454, 16KK0106, JP15H05884 (J-Physics) and 26400341.

References

[1] P. Santini, R. Lémanski and P. Erdös, Magnetism of actinide compounds, Adv. Phys. 48 (1999), pp. 537-653.
[2] P. Santini, S. Carretta, G. Amoretti, R. Caciuffo, N. Magnani and G. H. Lander, Multipolar interactions in f-electron systems: The paradigm of actinide dioxides, Rev. Mod. Phys. 81 (2009), pp. 807-863.
[3] K.T. Moore and G. van der Laan, Nature of the 5f states in actinide metals, Rev. Mod. Phys. 81, (2009), pp. 235-298.
[4] N. Tateiwa, J. Pospíšil, Y. Haga, H. Sakai, T.D. Matsuda and E. Yamamoto, Itinerant ferromagnetism in actinide 5f electrons system: Phenomenological analysis with spin fluctuation theory, Phys. Rev. B 96 (2017), pp. 035125-1-15.
[5] Y. Takahashi, On the origin of the Curie-Weiss law of the Magnetic susceptibility in Itinerant Electron Ferromagnetism, J. Phys. Soc. Jpn. 55 (1986), pp. 3553-3573.
[6] Y. Takahashi, Quantum spin fluctuation theory of the magnetic equation of state of weak itinerant-electron ferromagnets, J. Phys.: Condens. Matter. 13 (2001), pp. 6323-6358.
[7] Y. Takahashi, Spin Fluctuations Theory of Itinerant Electron Magnetism, Springer, New York, 2013.
[8] S.S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, G. G. Lonzarich, A. Huxley, I. Sheikin, D. Braithwaite and J. Flouquet, Superconductivity on the border of itinerant-electron ferromagnetism in UGe$_2$, Nature 406 (2000), pp. 587-592.
[9] A. Huxley, I. Sheikin, E. Ressouche, N. Kernavanois, D. Braithwaite, R. Calemczuk and J. Flouquet, UGe$_2$: A ferromagnetic spin-triplet superconductor, Phys. Rev. B 63 (2001), pp. 144519-1-13.
[10] D. Aoki, A. Huxley, E. Ressouche, D. Braithwaite, J. Flouquet, J. P. Brison, E. Lhotel and C. Paulsen, Coexistence of superconductivity and ferromagnetism in URhGe, Nature 413 (2001), pp. 613-615.
[11] N.T. Huy, A. Gasparini, D.E. de Nijs, Y. Huang, J.C.P. Klaasse, T. Gortenmulder, A. de Visser, A. Hamann, T. Görlich and H. v. Löhneysen, Superconductivity on the border of weak itinerant ferromagnetism in UCoGe, Phys. Rev. Lett. 99 (2007), pp. 067006-1-4.