Itinerant ferromagnetism in actinide 5*f*-electron systems: Phenomenological analysis with spin fluctuation theory

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(Received 11 April 2017; published 14 July 2017)

We have carried out an analysis of magnetic data in 69 uranium, 7 neptunium, and 4 plutonium ferromagnets with the spin fluctuation theory developed by Takahashi [Y. Takahashi, J. Phys. Soc. Jpn. **55**, 3553 (1986)]. The basic and spin fluctuation parameters of the actinide ferromagnets are determined and the applicability of the spin fluctuation theory to actinide 5*f* system has been discussed. Itinerant ferromagnets of the 3*d* transition metals and their intermetallics follow a generalized Rhodes-Wohlfarth relation between $p_{\rm eff}/p_{\rm s}$ and $T_{\rm C}/T_0$, viz., $p_{\rm eff}/p_{\rm s} \propto (T_{\rm C}/T_0)^{-3/2}$. Here, $p_{\rm s}$, $p_{\rm eff}$, $T_{\rm C}$, and T_0 are the spontaneous and effective magnetic moments, the Curie temperature, and the width of spin fluctuation spectrum in energy space, respectively. The same relation is satisfied for $T_{\rm C}/T_0 < 1.0$ in the actinide ferromagnets. However, the relation is not satisfied in a few ferromagnets with $T_{\rm C}/T_0 \sim 1.0$ that corresponds to local moment system in the spin fluctuation theory. The deviation from the theoretical relation 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. The value of the spontaneous magnetic moment $p_{\rm s}$ increases linearly as a function of $T_{\rm C}/T_0$ in the uranium and neptunium ferromagnets below $(T_{\rm C}/T_0)_{\rm kink} = 0.32 \pm 0.02$, where a kink structure appears in relation between the two quantities. $p_{\rm s}$ increases more weakly above $(T_{\rm C}/T_0)_{\rm kink}$. A possible interpretation with the $T_{\rm C}/T_0$ dependence of $p_{\rm s}$ is given.

DOI: 10.1103/PhysRevB.96.035125

I. INTRODUCTION

Actinide compounds with 5*f* electrons have long attracted much attention because of their interesting magnetic and electronic properties such as heavy fermion features, unconventional superconductivity, coexistence of the superconductivity and magnetism, and physical phenomena associated with multipole degrees of freedom of the the 5*f* electrons [1–4]. Similar unusual physical properties have been extensively studied in other strongly correlated electrons systems such as oxides and organic and rare-earth compounds. It is necessary to reveal the behavior of the electrons responsible for these properties and find universality of the electronic properties in the different systems.

The peculiarities of the physical phenomena in actinide systems are ascribed to the role of the 5*f* electrons with larger spatial extent than that of the 4*f* electrons in the lanthanides. The electrons show a tendency to delocalization through strong hybridization between 5*f* orbitals and conduction states. In actinide metallic compounds, the degree of localization of the 5*f* electrons differs in different compounds, ranging from strongly localized to itinerant character. The 3*d* electrons in transition metals also show various degree of localization. Differences between the 5*f* and 3*d* electrons are the smaller sensitivity to the crystal field (CF) from ligand atoms and the stronger spin-orbit coupling in the 5*f*s. The interplay of the spin and the unquenched orbital degrees of freedom gives rise to the peculiar features of the observed physical phenomena in actinide compounds.

While many theoretical studies have been done for the role of the 5*f* electrons on their interesting physical properties [1–3], the behavior of the 5*f* electrons has not been fully elucidated yet. When the 5*f* electrons have strongly localized character at higher temperature, a Kondo-lattice picture may be appropriate for understanding the formation of the strongly correlated electric states at low temperatures as has been established in the 4*f* electrons system of the rare-earth cerium (Ce) and ytterbium (Yb) compounds [5–7]. Certainly, some actinide compounds show behaviors reminiscent of the Kondo effect such as the logarithmic temperature dependence of the electrical resistivity ($\rho \sim -\ln T$). But not all physical properties in actinide compounds have been consistently explained on this point of view. There has been no experimental report of continuous and systematic evolution from the Kondo impurity to the concentrated Kondo lattice system, in contrast to the rare-earth Ce system such as $CeAl_2$ [8], CeB_6 [9], and $CeCu_6$ [10] whose physical properties can be continuously tuned by replacing the Ce ion with La without 4*f* electron. Furthermore, there have been controversies as to whether the 5*f* electrons should be treated as being itinerant or localized in various uranium compounds. The readers refer to Refs. [1] and [11] for these issues in the uranium chalogenide compounds (USe, US, and UTe) and UAsSe [1,11]. Generally, neither theoretical models based on the assumption of the localized nor itinerant 5*f* electrons can explain the physics of actinide metallic compounds. From a different point of view, this duality in the nature of the 5*f* electrons has been positively taken as a starting point in theoretical models for the heavy fermion superconductors UPd_2Al_3 or UPt_3 [5,12–14].

In this study, we focus on the nature of the 5*f* electrons in actinide ferromagnets and present interesting views deduced from analysis of the magnetic data using spin fluctuation

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theory. Among the actinide ferromagnets, uranium ferromagnetic superconductors UGe_2, URhGe, and UCoGe have been extensively studied both theoretically and experimentally for more than 10 years with the interest arising from the same uranium $5f$ electrons underlying both long-range ordered states [15–21]. Ferromagnetic superconductivity has been an important research subject since its theoretical prediction by Ginzburg in 1956 [22]. The study of the $5f$ electrons in the uranium ferromagnetic superconductors has potential importance not only for actinide science but also for wide research fields of the superconductivity and the magnetism.

In the theoretical studies for the ferromagnetic superconductors, the ferromagnetism of the $5f$ electrons has been variously approached via mean-field theory [23], the spin-fermion model [24,25], the Hubbard, periodic Anderson or Kondo Hamiltonians [26–30], and the j-j coupling scheme [31,32]. Some of the physical properties of the ferromagnetic superconductors have been explained by these theories but there have remained unsolved problems. One source of the difficulties can be attributed to the diversity of the nature in the $5f$ electrons as mentioned above. Photoemission spectroscopy suggests $5f$ electrons' itinerant character in UGe_2, URhGe, and UCoGe [33]. But the Ising-type anisotropy in the ferromagnetic state favors a localized model of the $5f$s. The degree of the itinerancy of the $5f$s may differ between the ferromagnetic superconductors. The dualism of $5f$s in UGe_2 has been discussed in both experimental and theoretical studies [24,25,34–36]. The complexity of the $5f$s makes it difficult to find an appropriate starting point for the electronic state of these electrons in theoretical studies, making it necessary to find a method to evaluate the degree of the itinerancy of the of the $5f$s in actinide ferromagnets.

In this paper, we report results of a phenomenological analysis on actinide ferromagnets using the spin fluctuation theory that was developed to account for the ferromagnetic properties of itinerant ferromagnets in the $3d$ transition metals and their intermetallics [37]. In the spin fluctuation theory, the degree of the itinerancy of the magnetic electrons is defined by the parameter T_C/T_0, where T_0 is the width of spin fluctuation spectrum in energy space. We adapt this theory and find a theoretical relation for the basic magnetic properties that also holds for actinide $5f$ electrons ferromagnets with certain exceptions. The parameter T_C/T_0 can also here be used to estimate the degree of the itinerancy of the $5f$s. We also suggest that relevant parameters for the actinide ferromagnetism are related to the itinerancy of the $5f$s, although some properties characteristic of a localized nature may play a role in the magnetic properties.

This paper is organized as follow. In the Sec. II, we briefly summarize the history of the spin fluctuation theory and the theoretical framework for the analysis of magnetic data in actinide ferromagnets. The experimental methods and analyses are given in Sec. III. In Secs. IV and V, we present the results of our analysis on 80 actinide ferromagnets and a summarizing discussion, respectively. A summary is given in Sec. VI.

II. SPIN FLUCTUATION THEORY

A. Brief history

The theoretical study of itinerant electron ferromagnets can be traced back to works by Bloch [38], Slater [39], and Stoner [40] starting in the late 1920s. Slater discussed the ferromagnetism of nickel (Ni) with tight binding d bands with intra-atomic exchange interaction only and Stoner developed the itinerant electron theory of ferromagnetism at finite temperature on the basis of the Hartree-Fock and mean-field approximations, respectively. The early theories explain ground-state magnetic properties of itinerant ferromagnets such as not integer values of the observed spontaneous magnetic moment in the $3d$ metals and their intermetallics. But there were difficulties in explaining finite-temperature magnetic properties such as Curie-Weiss behavior of the magnetic susceptibility above the ferromagnetic transition temperature T_C. These come from mean-field treatment of the on-site Coulomb interaction U whose strength ($\sim$10 eV) is one or two orders of magnitude larger than that of the kinetic energy K of the conduction electrons. Theoretical studies have been done to overcome the difficulties.

We focus on the improved mean-field approximation which takes into account the spatial spin-density fluctuations. The collective nature of magnetic excitations was considered in Herring and Kittel's theory of the spin waves by using the random-phase approximation (RPA) [41,42]. Izuyama, Kim, and Kubo developed the RPA theory for the dynamical magnetic susceptibility taking into account dispersive collective excitations and the spin fluctuations. The theory shows the magnetic critical scattering around T_C and the spin-wave dispersion in the ferromagnetic state [43]. The RPA theory was developed into the paramagnon model for nearly ferromagnetic metals or liquid helium-3 (^{3}He) [44–46]. These RPA theories are effective at low temperatures. Moriya and Kawabara developed self-consistent renormalization (SCR) theory where the dynamical susceptibility and the free energy are renormalized in a self-consistent way [37,47,48]. Phenomenological mode-mode coupling theory, equivalent to the SCR theory at high temperatures, was proposed by Murata and Doniach [49]. These advances were followed by alternative derivations of the SCR theory [50–54] and then extended to cover antiferromagnetic metals [55]. There are two important results found: One is a substantial reduction of T_C from the value in the Stoner's mean-field theory, and the other a new mechanism for the Curie-Weiss magnetic susceptibility in a wide temperature region. Other results from spin fluctuation theory have been confirmed in a number of experimental studies such as inelastic neutron-scattering experiment on the weak itinerant ferromagnet MnSi [56].

In reality, most of metallic ferromagnets are located in an intermediate region between the local moment and weakly coupling limits that the early theories assumed. A unified theory interpolating the two limiting cases has been developed by Moriya and Takahashi using a functional integral method [57]. Quantitative calculations of the ferromagnetic properties with electronic band structures were performed using the functional integral method [58,59]. First-principles approaches to the electronic structure of magnetic materials have also advanced

remarkably for several decades and important progress has been made in understanding finite-temperature magnetic properties of the itinerant ferromagnets iron (Fe) and Ni with the combination of the dynamic mean-field theory (DMFT) and electric structure calculations [60–63].

From the late 1980s, the spin fluctuation theory has been extended to study the unconventional superconductivity with anisotropic superconducting order parameter in strongly correlated electron systems such as high-T_c cuprates, heavy fermion superconductors, and two-dimensional organic compounds [64,65]. A theoretically predicted linear relation between the superconducting transition temperature T_{sc} and the spread of the spin fluctuation spectrum in energy space T_0 has been confirmed in the cuprate and heavy fermion superconductors [66,67]. Moriya's SCR theory was derived using of renormalization group theory [68] and reformulated with the standard s-f model to explain the anomalous physical properties around magnetic instability in the heavy fermion systems of the rare-earth and actinide compounds [69]. The validity of the theory has been confirmed by careful experimental studies [70–72] and is regarded as one of standard views in the research field of quantum critical phenomena [73]. In this study, we apply the spin fluctuation theory to actinide ferromagnets.

B. Takahashi's spin fluctuation theory

We analyze the magnetic data of uranium (U), neptunium (Np), and plutonium (Pu) ferromagnets with the spin fluctuation theory developed by Takahashi [74–77]. The total amplitude of the local spin fluctuation $\langle S_L^2\rangle_{\rm total}$ is composed of the thermal $\langle S_L^2\rangle_T$ and the zero point fluctuations $\langle S_L^2\rangle_{\rm Z.P.}$. The Takahashi's spin fluctuation theory assumes that $\langle S_L^2\rangle_{\rm total}$ is constant as a function of temperatures. This assumption is reasonable, considering the weak temperature dependence of the spin amplitude in the order of T_C seen in neutron-scattering experiments on MnSi [78] and $Y_{0.97}Sc_{0.03}Mn_2$ [79], and theoretical calculations with the Hubbard models [80–83] and the Moriya's SCR theory [84]. The effectiveness of the theory has been confirmed in a number of experimental studies on intermetallic compounds of the 3d transition metals [85–96].

The local spin fluctuation density squared $\langle S_L^2\rangle$ is related to the imaginary part of the dynamical magnetic susceptibility $\chi(\boldsymbol{q},\omega)$ through the fluctuation and dissipation theorem. The spin fluctuation spectrum for itinerant ferromagnets is described by double Lorentzian distribution functions in small energy ω and wave vector $\boldsymbol{q}$ spaces [76],

$$\mathrm{Im}\chi(\boldsymbol{q},\omega) = \frac{\chi(0)}{1+q^2/\kappa^2}\frac{\omega\Gamma_q}{\omega^2+\Gamma_q^2}, \tag{1}$$

$$\Gamma_q = \Gamma_0 q(\kappa^2+q^2), \tag{2}$$

Here, $q \equiv |\boldsymbol{q}|$, κ represents the inverse of the magnetic correlation length, and Γ_q is the damping constant, the inverse of the life time of the fluctuation with wave vector $\boldsymbol{q}$. The spectrum is represented in a parameterized form by introducing two energy scales T_0 $(=\Gamma_0 q_B^3/2\pi)$ 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 $(=6\pi^2 N_0/q_B^3)$. These parameters represent the distribution widths of the spin fluctuation spectrum in the energy and wave-vector spaces, respectively.

In the Takahashi's spin fluctuation theory, the Landau expansion of free energy $F_m(M)$ is expressed as

$$\begin{aligned} F_m(M) &= F_m(0) + \frac{1}{2}a(0)M^2 + \frac{1}{4}b(0)M^4, \\ a(0) &= \frac{1}{(g\mu_B)^2\chi(0)}, \\ b(0) &= \frac{F_1}{(g\mu_B)^4 N_0^3}, \end{aligned} \tag{3}$$

where g represents Lande's g factor. F_1 is the mode-mode coupling term, the coefficient of the M^4 term.

The magnetization M at the ground state is expressed by the following equation:

$$H = \frac{F_1}{N_0^3(g\mu_B)^4}\left(-M_0^2+M^2\right)M, \tag{4}$$

$$F_1 = \frac{2{T_A}^2}{15cT_0}, \tag{5}$$

where $c = 1/2$ and M_0 is the spontaneous magnetic moment. The parameter F_1 is connected with T_0 and T_A through Eq. (5) and can be evaluated experimentally from the inverse slope of the Arrott plots (M^2 versus H/M plot) at low temperatures,

$$F_1 = \frac{{N_A}^3(2\mu_B)^4}{k_B\zeta}, \tag{6}$$

where N_A is Avogadoro's number, k_B is the Boltzmann constant, and ζ is the slope of the Arrott plot ($H/M - M^2$ curve) at low temperatures [97]. T_0 and T_A are expressed with F_1 in following relations:

$$\left(\frac{T_C}{T_0}\right)^{5/6} = \frac{p_s^2}{5g^2C_{4/3}}\left(\frac{15cF_1}{2T_C}\right)^{1/2}, \tag{7}$$

$$\left(\frac{T_C}{T_A}\right)^{5/3} = \frac{p_s^2}{5g^2C_{4/3}}\left(\frac{2T_C}{15cF_1}\right)^{1/2}, \tag{8}$$

where $C_{4/3}$ is a constant ($C_{4/3} = 1.006089\cdots$). p_s is the spontaneous magnetic moment and T_C is the ferromagnetic transition temperature expressed in following equations:

$$p_s^2 = \frac{12T_0}{T_A}C_{4/3}\left(\frac{T_C}{T_0}\right)^{4/3}, \tag{9}$$

$$T_C = (60c)^{-3/4}p_s^{3/2}T_A^{3/4}T_0^{1/4}. \tag{10}$$

An important consequence from the Takahashi's theory is that the parameters F_1, T_0, and T_A can be determined from experimental magnetic data only. Meanwhile, the parameters are independent in the Moriya's SCR theory and the neutron or nuclear magnetic resonance spectroscopy is necessary to determine them [37,47,48].

A ratio T_C/T_0 characterizes the degree of itinerancy of magnetic electrons in the spin fluctuation theory. At $T_C/T_0 \ll 1$, the magnetic electrons have a strong itinerant character. The value of $p_{\rm eff}/p_s$ is large and T_C/T_0 becomes small for weak ferromagnets with large spin fluctuation amplitude. Both quantities approach to unity when the degree of itinerancy of

the magnetic electrons becomes small. The local magnetic moment is responsible for the ferromagnetism when $T_C/T_0 = 1$.

In 1963, Rhodes and Wohlfarth proposed to plot the ratio p_{eff}/p_s as a function of T_C (Rhodes and Wohlfarth plot) [98,99]. The ratio p_{eff}/p_s was defined as a measure of quantification to the degree of itinerancy of the magnetic electrons. This Rhodes and Wohlfarth plot has been widely used in studies of itinerant ferromagnets for a long time. However, there is no theoretical ground to the relation between p_{eff}/p_s and T_C. In the Takahashi's theory, p_{eff}/p_s is described as a function of the parameter T_C/T_0 as follows:

$$\frac{p_{eff}}{p_s} = \left(\frac{1}{10C_{4/3}dy/dt}\right)^{-1/2}\left(\frac{T_C}{T_0}\right)^{-2/3} \simeq 1.4\left(\frac{T_C}{T_0}\right)^{-2/3}. \quad (11)$$

where y is the inverse magnetic susceptibility and t is the reduced temperature $t = T/T_0$. The quantity $[1/(10C_{4/3}dy/dt)]^{-1/2}$ is numerically estimated as $\sim$1.4 [76]. This generalized Rhodes-Wohlfarth relation between p_{eff}/p_s and T_C/T_0 has been experimentally confirmed in a number of ferromagnetic compounds in the 3*d* electrons systems [85–96]. The spin fluctuation parameters for the uranium feromagnetic superconductors UGe_2, URhGe, and UCoGe have been reported by Deguchi, Takahashi, and Sato [76,100]. In this paper, we discuss the applicability of the spin fluctuation theory to the actinide 5*f* electrons system from the analyses on the magnetic data of 80 actinide ferromagnets with the Takahashi's theory.

III. METHODS OF EXPERIMENT AND ANALYSIS

We have examined the 69 uranium, 7 neptunium, and 4 plutonium ferromagnets listed in Tables I, II, and III. The uranium ferromagnets are divided into two categories, group I and group II. We have obtained the basic magnetic and the spin fluctuation parameters from the analyses of our experimental data for the uranium ferromagnets in group I. The single crystal samples of group I were grown by the Czochralski crystal pulling method. The magnetic measurements have been done with a commercial superconducting quantum interference device (SQUID) magnetometer. The parameters for the uranium ferromagnets in group II and neptunium and plutonium ferromagnets are obtained from the analyses of experimental data taken from the literature.

We selected the ferromagnetic compounds that show a single ferromagnetic phase transition and excluded ferromagnets like $AnFe_2$ (An: U, Np, and Pu) where the magnetic moment of the 3*d* transition metal has an important contribution in the magnetic property at zero magnetic field [101]. Spin fluctuation theory assumes a simple ferromagnetic state. We excluded ferromagnets with complex magnetic structures such as U_3P_4 and U_3As_4 [1]. Generally, the ferromagnetic state has magnetic anisotropy in actinide compounds. We have analyzed the magnetic data of the actinide ferromagnets obtained using single crystal samples in applied magnetic field along the magnetic easy axis. There are some exceptions as will be mentioned in the next section.

The parameter F_1 is determined from the slope ζ of the Arrott-plot (H/M-M^2 curve) at T^* with the Eq. (6). Then the spin fluctuation parameters T_0 and T_A can be estimated with the value of p_s using Eqs. (7) and (8). We confirm that the transition temperature T_C determined experimentally is reproduced from the values of p_s, T_0, and T_A using Eq. (10). The effective magnetic moment p_{eff} is determined from the slope of the inverse magnetic susceptibility $1/\chi$ from the Curie-Weiss (CW) law $\chi = C/(T-\theta)$. Here C is the Curie constant and θ the paramagnetic Curie temperature. The effective magnetic moment, p_{eff}, per magnetic atom is estimated from $C = N_0\mu_B^2 p_{eff}^2/3k_B$. In some ferromagnets, $1/\chi$ is not linear in temperature. The magnetic susceptibility χ was then analyzed with the modified Curie-Weiss (mCW) law, $\chi = C/(T-\theta)+\chi_0$. χ_0 is a temperature-independent term which may arise from the density of states at the Fermi energy other than 5*f* electrons.

IV. RESULTS

A. Basic magnetic and spin fluctuation parameters and generalized Rhodes-Wohlfarth plot

Table I shows the basic magnetic and spin fluctuation parameters of the uranium ferromagnets in group I. These have been determined from the analysis of our experimental data. We used published data of UGe_2 [102,103], URhGe [103], UIr [104], UGa_2 [105], $URhGe_2$ [106], UCu_2Ge_2 [107], and $URh_{1-x}Ir_xGe$ [108]. Unpublished data were analyzed for the ferromagnets URh, URh_6Ge_4, URhSi, URhAl, and $URh_{1-x}Co_xGe$ marked with an asterisk * in the table. The detailed studies of the physical properties in the latter compounds are now in progress using high-quality single crystal samples and part of the magnetic data were used for this study. Among them, URh_6Ge_4 is a new ferromagnet with $T_C = 14.8$ K that crystalizes in the hexagonal $LiCo_6P_4$-type structure. The ferromagnetic property in URh, isostructural to UIr, has been previously studied using polycrystal samples [109,110]. We have studied the anisotropic ferromagnetic properties in URh using single crystal samples. URhSi and URhAl are known ferromagnets [111–113]. The basic magnetic properties of the ferromagnets such as the value of T_C or p_s in our single crystal samples are consistent with those in previous studies. We are interested in doping effect on the uranium ferromagnetic superconductors URhGe and UCoGe. The systematic study of the magnetic properties in the series of $URh_{1-x}Ir_xGe$ and $URh_{1-x}Co_xGe$ are underway with high-quality single crystal samples. The doping dependencies of the spin fluctuations parameters are shown in this paper. Note that the basic magnetic properties in the series are consistent with those in previous studies using polycrystal samples [114,115].

Figure 1(a) shows the temperature dependencies of the magnetization M and (b) the inverse of the magnetic susceptibility $1/\chi$ for several uranium ferromagnets UCu_2Ge_2, URhAl, URh_6Ge_4, URhSi, and URhGe in the group I in magnetic fields of 0.5, 0.2, 0.1, 0.1, and 0.1 T, respectively, applied along the magnetic easy axes of the ferromagnets. The Curie temperature T_C denoted by an arrow is determined from the temperature dependencies of the specific heat, the electrical resistivity, and the magnetization in zero or low magnetic field. The effective magnetic moment p_{eff} is determined from the

TABLE I. Basic magnetic and spin fluctuation parameters for uranium ferromagnets in group I. These parameters are obtained with present authors' experimental data taken on single crystalline samples and unpublished data are used for the ferromagnets marked with an asterisk *. The spontaneous magnetic moment p_s and the effective magnetic moment p_{eff} are estimated from the magnetization at T^* and the magnetic susceptibility, respectively. The spin fluctuation parameters, F_1, T_0, and T_A are estimated with Takahashi's spin fluctuation theory. The mode-mode coupling term F_1 is determined from the magnetization data at T^*. Abbreviation CW or mCW in column $\chi(T)$ denotes that the magnetic susceptibility has been analyzed the Curie-Weiss or modified Curie-Weiss law, respectively.

	T_C (K)	p_{eff} (μ_B/U)	p_s (μ_B/U)	p_{eff}/p_s	F_1 (K)	T_0 (K)	T_A (K)	T_C/T_0	$\chi(T)$	T^* (K)	Ref.
Uranium compounds: group I											
UGe_2	52.6	3.00	1.41	2.13	554	92.2	442	0.571	CW	2.0	[102,103]
UIr	46.0	3.40	0.492	6.91	1.97×10^3	440	1.80×10^3	0.105	CW	2.0	[104]
UGa_2	123	3.30	2.94	1.12	273	94.8	311	1.12	CW	2.0	[105]
$URhGe_2$	30.0	3.06	0.768	3.99	517	170	574	0.176	CW	2.0	[106]
UCu_2Ge_2	109	2.93	1.74	1.69	521	187	605	0.582	CW	2.0	[107]
URhGe	9.47	1.75	0.407	4.30	1.10×10^3	78.4	568	0.121	mCW	2.0	[103]
URh*	57	2.26	0.652	3.47	1.52×10^3	367	1.45×10^3	0.155	mCW	2.0	
URh_6Ge_4*	14.8	3.58	1.39	2.58	560	13.2	167	1.15	CW	2.0	
URhAl*	26.2	2.50	1.05	2.37	428	71.8	340	0.365	mCW	2.0	
URhSi*	10.5	2.94	0.571	5.15	520	64.5	354	0.163	CW	2.0	
$URh_{1-x}Co_xGe$*											
$x = 0.2$	13.7	1.86	0.450	4.13	1.22×10^3	104	691	0.131	mCW	1.87	
$x = 0.6$	19.7	1.91	0.498	3.83	1.01×10^3	164	788	0.120	mCW	1.87	
$x = 0.7$	18.6	1.94	0.416	4.65	945	239	921	0.0777	mCW	1.87	
$x = 0.8$	15.0	1.92	0.293	6.56	1.11×10^3	358	1.22×10^3	0.0419	mCW	1.87	
$x = 0.9$	7.0	1.94	0.127	15.3	2.93×10^3	439	2.19×10^3	0.0160	mCW	1.87	
$URh_{1-x}Ir_xGe$*											[108]
$x = 0.15$	9.3	1.75	0.392	4.47	1.22×10^3	78.3	599	0.119	mCW	1.87	
$x = 0.43$	6.0	1.73	0.292	5.92	1.27×10^3	76.6	605	0.0783	mCW	1.87	

fit to the magnetic susceptibility χ with the Curie-Weiss or modified Curie-Weiss law as shown in lines in Fig. 1(b). The obtained values of the parameters, T_C, p_{eff}, and p_s are listed in Table I. Generally, the values of p_s and p_{eff} are smaller than those expected for $5f^2$ (U^{4+}, $p_{eff} = 3.58\mu_B$/U) and $5f^3$ (U^{3+}, $p_{eff} = 3.62\mu_B$/U) configurations, which can be understood as manifestation of the itinerancy of the $5f$ electrons.

Figure 2 shows the M^2 versus H/M plot (Arrott plots) at $T^* = 2.0$ K for UCu_2Ge_2, URhAl, URh_6Ge_4, URhSi, and URhGe in applied magnetic field along the magnetic easy axes of the ferromagnets. We estimate F_1 from the slope ζ of Arrot plots shown as lines in the figure. The obtained values of F_1 are 521, 428, 560, 520, and 1.10×10^3 K, respectively. Then, the spin fluctuation parameters T_0 and T_A are estimated with Eqs. (7) and (8) using the values of T_C, F_1, and p_s. These obtained parameters are also listed in the Table I. We have estimated the parameters for the rest of the uranium ferromagnets in the group II in the same ways and show them in the Table II. The spin fluctuation parameters for UGe_2 and URhGe in this study are consistent with those determined previously [76,100].

Extensive experimental studies have been carried out on many actinide ferromagnets more than five decades. To examine applicability of the spin fluctuation theory to the actinide system from a wider viewpoint, we decided to analyze the magnetic data of actinide ferromagnets reported in literature. We selected ferromagnets according to the criteria mentioned in the previous section and analyzed the magnetic data from litereture. The basic magnetic and the spin fluctuation parameters are obtained for 52 uranium ferromagnets, 7 neptunium, and 4 plutonium ferromagnets as listed in Tables II and III. Note that the parameters in UCoGe determined previously are cited from Refs. [76,100]. As can be seen from the Table I, II, and III, T_C/T_0 in the actinide ferromagnets shows a wide range of values from $T_C/T_0 = 0.0065$ for UCoGe to 1.70 for U_3TiSb_3. The degree of itinerancy of $5f$ electrons in the actinide ferromagnets largely differ, depending on each ferromagnet. It is interesting to compare the values of T_C/T_0 in the actinide ferromagnets with those of well-known itinerant ferromagnets in the $3d$ system such as Ni_3Al ($T_C/T_0 = 0.015$), MnSi (0.13), $ZrZn_2$ (0.053), $Y(Co_{1-x}Al_x)_2$ ($0.0036 \sim 0.018$), Ni (0.237), and Fe (1.05) [74,76].

Figure 3 shows the double logarithmic plot of p_{eff}/p_s and T_C/T_0 (the generalized Rhodes-Wohlfarth plot) for all actinide ferromagnets analyzed. The data points for the uranium, neptunium, and plutonium ferromagnets are shown in closed circles, squares, and triangles, respectively. Solid line is the theoretical relation [Eq. (11): $p_{eff}/p_s = 1.4(T_C/T_0)^{-2/3}$] in the Takahashi's spin fluctuation theory. For comparison, we plot the data of the $3d$ transition metals and their intermetallics such as Ni_3Al, $Ni_3Al_{1-x}Ga_x$ ($0 \leqslant x \leqslant 0.33$), $(Fe_{1-x}Co_x)_3Mo_3N$, Sc_3In, $Y(Co_{1-x}Al_x)Co_2$ ($0.13 \leqslant x \leqslant 0.19$), MnSi, $ZrZn_2$, $Fe_xCo_{1-x}Si$ ($0.36 \leqslant x \leqslant 0.91$), Ni_xRh_{1-x} ($0.72 \leqslant x \leqslant 0.90$), $Pt_{1-x}Ni_x$ ($0.429 \leqslant x \leqslant 0.502$), Y_2Ni_x ($6.7 \leqslant x \leqslant 7.0$), $YNi_{2.9}$, YNi_3, Y_2Ni_{17}, Y_2Ni_{15}, Fe, and Ni [74,76,92,96].

TABLE II. Basic magnetic and spin fluctuation parameters for uranium ferromagnets in group II. These parameters are estimated from the analysis of experimental data from the literature. The meanings of the other notations are the same as in Table I.

	T_C (K)	p_{eff} (μ_B/U)	p_s (μ_B/U)	p_{eff}/p_s	F_1 (K)	T_0 (K)	T_A (K)	T_C/T_0	$\chi(T)$	T^* (K)	Ref.
Uranium compounds: group II											
UPt	28.6	3.58	0.822	4.36	566	127	519	0.226	CW	5.0	[116]
US	177	2.35	1.51	1.56	1.71×10^3	275	1.34×10^3	0.643	mCW	4.05	[117,118]
USe	160	2.50	1.72	1.45	823	271	914	0.591	mCW	4.2	[119]
UTe	104	2.70	1.90	1.42	537	139	528	0.751	mCW	1.5	[120]
U_5Sb_4	86	2.98	1.63	1.83	416	171	517	0.503	CW	1.5	[121]
UAsS	124	3.34	1.17	2.85	1.08×10^3	383	1.25×10^3	0.323	CW	4.2	[122,123]
UAsSe	113	3.41	1.29	2.64	957	281	1.00×10^3	0.403	CW	4.2	[122,123]
UAsTe	66	3.34	1.29	2.59	343	222	534	0.298	CW	4.2	[122,123]
UPS	118	2.57	1.04	2.47	1.16×10^3	454	1.40×10^3	0.260	CW	5.0	[124]
UPSe	55	3.17	1.03	3.06	363	271	607	0.203	CW	5.0	[125]
UPTe	85	2.83	1.37	2.06	421	252	631	0.337	CW	5.0	[125]
USbSe	128	3.08	1.68	1.83	905	189	802	0.676	CW	5.0	[125]
USbTe	127	3.18	1.92	1.65	1.11×10^3	120	705	1.06	CW	5.0	[125]
USeTe	69	3.04	1.59	1.91	197	201	385	0.343	mCW	5.0	[126]
USTe	85	2.92	1.54	1.89	325	223	522	0.381	mCW	5.0	[126]
$UCu_{0.9}Sb_2$	113	3.10	1.67	1.88	414	252	93.5	0.448	CW	1.9	[127]
$UCo_{0.5}Sb_2$	65	2.80	1.11	2.52	429	271	660	0.240	CW	1.9	[128]
$UAuBi_2$	22.5	3.30	1.25	2.64	223	55.3	215	0.407	CW	1.8	[129]
$UCuAs_2$	133	2.68	1.27	2.11	5.29×10^3	136	1.64×10^3	0.979	mCW	4.2	[130]
$UCuP_2$	74.5	2.21	0.993	2.23	1.09×10^4	63.3	1.61×10^3	1.18	mCW	4.2	[130]
UCu_2P_2	216	2.26	1.80	1.25	2.15×10^3	219	1.33×10^3	0.987	mCW	4.2	[131]
U_3TiSb_5	160	2.81	1.65	1.70	3.07×10^3	135	1.25×10^3	1.18	CW	5.0	[132]
U_3ScSb_5	130	2.86	1.46	1.95	688	318	906	0.409	CW	5.0	[132]
$U_3Cu_4Ge_4$	73.0	2.99	1.68	1.77	183	200	370	0.365	mCW	2.0	[133]
$U_3Fe_4Ge_4$	18.0	1.93	0.402	4.80	1.27×10^3	207	993	0.0871	mCW	2.0	[134]
U_2Fe_3Ge	55	2.52	0.488	5.16	4.54×10^3	362	2.48×10^3	0.152	mCW	2.0	[135]
U_2RhSi_3	24.0	2.50	0.707	3.53	635	128	552	0.187	mCW	1.72	[136]
$U_3Co_2Ge_7$	40	2.41	1.07	2.25	900	87.2	543	0.459	mCW	2.0	[137]
$U_4(Ru_{1-x}Os_x)_7Ge_6$											[138]
$x = 0$	12.0	1.38	0.206	6.71	8.79×10^3	170	2.36×10^3	0.0708	mCW	2.0	
$x = 0.1$	9.0	1.36	0.148	9.20	8.20×10^3	246	2.75×10^3	0.0365	mCW	2.0	
$x = 0.2$	7.0	1.35	0.118	11.4	7.88×10^3	289	2.92×10^3	0.0312	mCW	2.0	
UCoGa	47	2.40	0.638	3.76	1.44×10^3	295	1.26×10^3	0.159	CW	4.2	[139,140]
UPtAl	43.5	2.85	1.38	2.06	615	67.8	395	0.642	CW	4.2	[141]
UIrAl	62	2.20	0.960	2.30	820	241	861	0.257	CW	4.2	[142]
$UCoAl_{0.75}Sn_{0.25}$	5.5	2.66	0.169	15.7	924	300	1.02×10^3	0.0183	mCW	2.0	[143]
$UCo_{1-x}Ru_xAl$											[144,145]
$x = 0.005$	4.5	1.87	0.322	5.81	1.24×10^3	39.0	426	0.115	mCW	1.8	
$x = 0.01$	16	1.83	0.365	5.02	2.19×10^3	156	1.13×10^3	0.103	mCW	1.8	
$x = 0.62$	38.0	1.90	0.428	4.44	2.54×10^3	396	1.94×10^3	0.0972	mCW	1.8	
$x = 0.70$	17.0	2.20	0.169	13.0	3.93×10^3	766	3.36×10^3	0.0222	mCW	1.8	
$x = 0.74$	6.5	2.00	0.0732	27.3	8.78×10^3	759	5.00×10^3	0.00856	mCW	1.8	
$UCo_{1-x}Os_xAl$											[146]
$x = 0.02$	26	1.91	0.394	4.85	1.74×10^3	324	1.45×10^3	0.0802	mCW	2.0	
$x = 0.1$	48	1.95	0.518	3.766	1.84×10^3	433	1.73×10^3	0.111	mCW	2.0	

The data points for the actinide ferromagnets in Fig. 3 are distributed overwider parameter ranges of T_C/T_0 and p_{eff}/p_s. This suggests that the $5f$ electrons in the actinide ferromagnets show the various degree of itinerancy. The data for the actinide ferromagnets are plotted close to those of the itinerant ferromagnets in the $3d$ transition metals and their intermetallics. The relations between the two quantities T_C/T_0 and p_{eff}/p_s in the actinide ferromagnets follow the relation in the Takahashi's spin fluctuation theory for $T_C/T_0 < 1.0$. This suggests itinerant character of the $5f$ electrons in most of the actinide ferromagnets. Similarities of the ferromagnetic properties between the $3d$ and $5f$ electrons systems are evident

TABLE III. Basic magnetic and spin fluctuation parameters for uranium ferromagnets in group II and neptunium and plutonium ferromagnets. These parameters are estimated from the analysis of experimental data from the literature. The parameters are estimated with data taken on polycrystalline samples for the ferromagnets marked with a dagger †. An indicates actinide atom U, Np, or Pu. ⋆The value of $p_{\rm eff}$ is assumed to be 1.90 in $UCo_{1-x}Fe_xGe$ (see text). ‡The parameters in UCoGe are cited from Refs. [76,100]. The meanings of the other notations are the same as in Table I.

	T_C (K)	$p_{\rm eff}$ (μ_B/An)	p_s (μ_B/An)	$p_{\rm eff}/p_s$	F_1 (K)	T_0 (K)	T_A (K)	T_C/T_0	$\chi(T)$	T^* (K)	Ref.
Uranium compounds: group II											
$UCo_{1-x}Ru_xGe$											[145,147]
$x = 0.03$	6.50	1.88	0.108	17.4	7.96×10^3	316	3.07×10^3	0.0206	mCW	1.8	
$x = 0.10^\dagger$	8.62	1.86	0.111	16.8	1.38×10^4	337	4.18×10^3	0.0256	mCW	1.8	
$x = 0.12$	7.46	1.88	0.149	12.6	5.54×10^3	226	1.44×10^3	0.0330	mCW	1.8	
$x = 0.23^\dagger$	4.68	1.83	0.0544	34.6	2.43×10^4	495	6.72×10^3	0.00945	mCW	1.8	
$UCo_{1-x}Fe_xGe^{\dagger,\star}$											[148]
$x = 0.025$	7.73	1.9	0.156	12	3.99×10^3	251	1.94×10^3	0.0308	mCW	2.0	
$x = 0.05$	8.35	1.9	0.206	9.2	2.00×10^3	229	1.31×10^3	0.0364	mCW	2.0	
$x = 0.075$	8.55	1.9	0.310	6.1	606	183	645	0.0466	mCW	2.0	
$x = 0.10$	6.90	1.9	0.229	8.3	1.07×10^3	157	875	0.0389	mCW	2.0	
$x = 0.125$	7.02	1.9	0.220	8.8	763	263	869	0.0267	mCW	2.0	
$x = 0.15$	6.40	1.9	0.0980	19	3.25×10^3	805	2.85×10^3	0.00894	mCW	2.0	
$UCoGe^\ddagger$	2.4	1.93	0.039	49.5	2.87×10^4	362	5.92×10^3	0.0065	mCW	0.1	[76,100]
Neptunium compounds											
$NpAl_2$	56.0	2.40	1.21	1.99	884	113	613	0.494	CW	4.2	[149,150]
$NpOs_2$	7.50	3.10	0.361	8.58	589	105	481	0.0715	CW	4.2	[149,150]
$NpNi_2$	32.0	2.42	0.756	3.19	519	195	616	0.164	CW	5.0	[151]
$Np_2C_3^\dagger$	109	2.40	0.920	2.61	1.53×10^3	452	1.61×10^3	0.241	CW	4.2	[152]
$NpSb_2$	47.0	2.52	1.57	1.60	282	90.0	309	0.522	CW	5.0	[153]
$NpNiSi_2$	51.5	2.16	1.13	1.92	848	120	617	0.427	CW	5.0	[154]
$NpFe_4P_{12}$	23	1.55	1.35	1.15	1.33×10^3	16.4	285	1.40	mCW	5.0	[155]
Plutonium compounds											
PuAs	125	1.00	0.655	1.53	2.85×10^4	232	4.98×10^3	0.557	CW	4.2	[156]
PuP	126	1.06	0.618	1.72	6.04×10^4	164	6.09×10^3	0.770	mCW	4.2	[11,157]
$PuGa_3^\dagger$ ($R-3m$)	20	0.77	0.197	3.91	5.92×10^4	136	5.49×10^3	0.147	mCW	5.0	[158]
$Pu_2Pt_3Si_5$	58.0	0.78	0.305	2.56	6.95×10^4	249	7.79×10^3	0.242	mCW	5.0	[159]

and suggest that the spin fluctuation theory can be applied to the 5f actinide ferromagnets. This finding is surprising since there are differences in the nature of the 3d and 5f electrons as mentioned in the Introduction.

Around $T_C/T_0 \sim 1$, the data points of U_3TiSb_5, $UCuP_2$, $UCuAs_2$, and URh_6Ge_4 deviate from the theoretical relation. The situation $T_C/T_0 = 1$ corresponds to the local moment system in spin fluctuation theory and the deviation of the data points may be due to characteristic features of the localized 5f electrons not included in the theory as will be discussed in the next section.

We look for other systematic tendencies among the basic and the spin fluctuation parameters. Figure 4 shows relation between T_C/T_0 and the spontaneous magnetic moment p_s and for the uranium, neptunium, and plutonium ferromagnets. The relation in the 3d system is also plotted. There is a general tendency of a positive correlation between the two quantities. The data points of the 3d systems are comparably scattered but there seems to be a linear relation between p_s and T_C/T_0 in the uranium and neptunium ferromagnets. It is reasonable that p_s increases with increasing degree of localization of the 5f electrons. The value of p_s in the uranium and neptunium ferromagnets increases as a function of T_C/T_0 followed by a kink structure in the relation between the two quantities at $(T_C/T_0)_{\rm kink} = 0.32 \pm 0.02$ as denoted by an arrow in the figure. The bold dotted line is a fit to the data of the uranium and neptunium ferromagnets for $T_C/T_0 < (T_C/T_0)_{\rm kink}$ with the function $p(T_C/T_0) = a(T_C/T_0)^{-n}$ where a and n are fitting parameters. The values of a and n are determined as 3.05 and 0.838, respectively. p_s increases more weakly above $(T_C/T_0)_{\rm kink}$. The data points are scattered around $T_C/T_0 \sim 1$, similar to the ones between T_C/T_0 and $p_{\rm eff}/p_s$ shown in Fig. 3. It is interesting to note that the extrapolated value of p_s for $T_C/T_0 \to 1$ with the fitted line is close to the values of effective Bohr magneton number of the free actinide ions ($3.58\mu_B$ for U^{4+}: f^2 configuration, $3.62\mu_B$ for U^{3+} and Np^{4+}: f^3 configuration, $2.83\mu_B$ for Np^{3+} and Pu^{4+}: f^4 configuration). This is naturally expected since the value of p_s in the local moment system ($T_C/T_0 = 1$) should be equal to the free actinide ion ones. Meanwhile, actual data points are smaller than the extrapolated line above $(T_C/T_0)_{\rm kink}$, suggesting a spin-compensating mechanism that

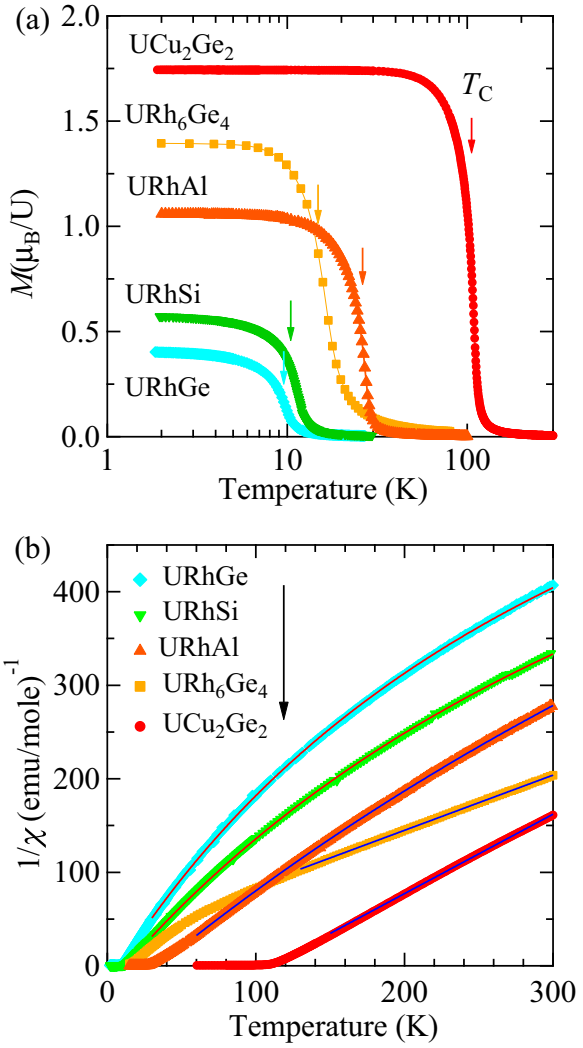

FIG. 1. Temperature dependencies of (a) the magnetization M under magnetic field and (b) the inverse of the magnetic susceptibility $1/\chi$ for several uranium ferromagnets UCu_2Ge_2, URhAl, URh_6Ge_4, URhSi, and URhGe in magnetic fields of 0.5, 0.2, 0.1, 0.1, and 0.1 T, respectively, applied along the magnetic easy axes. The Curie temperatures T_C are denoted by arrows. Solid lines are results of fits to the temperature dependence of the inverse of the magnetic susceptibility $1/\chi$ with the Curie-Weiss or modified Curie-Weiss law.

will be discussed in the next section. From the four data points of the plutonium ferromagnets, the value of p_s seems to approach to that ($0.84\mu_B$) of the trivalent Pu ion (Pu^{3+}: f^5 configuration).

B. Some technical notes

We explain the analyses of several compounds. The analyses of most of the ferromagnets have been done using the magnetic data taken on single crystalline samples in magnetic field applied along the magnetic easy axes as mentioned in the previous section. This is because the ferromagnetic state in actinide system generally has the magnetic anisotropy. But there are some exceptions. The analysis has been done on magnetic data taken on polycrystalline samples in the literatures

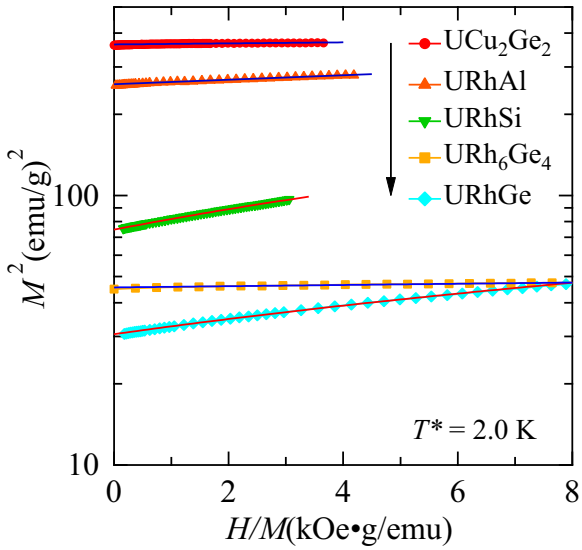

FIG. 2. M^2 versus H/M plot (Arrott plots) at $T^* = 2.0$ K for UCu_2Ge_2, URhAl, URh_6Ge_4, URhSi, and URhGe in applied magnetic field along the magnetic easy axes.

for $UCo_{1-x}Ru_xGe$ ($x = 0.10$ and 0.23), $UCo_{1-x}Fe_xGe$, Np_2C_3, and $PuGa_3$ marked with a dagger† in Tables II and III. The series of $UCo_{1-x}Ru_xGe$, $UCo_{1-x}Fe_xGe$, $URh_{1-x}Co_xGe$, $URh_{1-x}Ir_xGe$, and the ferromagnetic superconductors URhGe and UCoGe crystalize in the orthorhombic TiNiSi-type structure. The strong uniaxial anisotropy is a characteristic feature in the ferromagnetic state of this system. The spin fluctuations parameters of $UCo_{1-x}Ru_xGe$ ($x = 0.10$ and 0.23) were obtained from the analyses of the magnetization data taken on polycrystalline samples multiplied by three. Note that the parameters for $x = 0.03$, and 0.12 in $UCo_{1-x}Ru_xGe$ were obtained with the data taken on single crystalline samples. This method may be reasonable for this system considering the systematic changes of the basic and spin fluctuation parameters as a function of the concentration x in the series. The same treatment has been done in the analysis for $UCo_{1-x}Fe_xGe$ [148]. There has been no report for the effective paramagnetic moment p_{eff} in $UCo_{1-x}Fe_xGe$. In this study, the value of p_{eff} is assumed to be 1.90. The effect of the alloying does not largely affect the value of p_{eff} in the doped systems of UCoGe or URhGe. The values of p_{eff} are between 1.83 and $1.93\mu_B$/U in $UCo_{1-x}Ru_xGe$, $UCo_{1-x}Rh_xGe$, and UCoGe. This approximation of p_{eff} is reasonable but it could cause small uncertainty in p_{eff}/p_s that is reflected in error bars of the data points.

The crystal structure of Np_2C_3 is cubic [152]. Thus, the magnetic data of Np_2C_3 measured using polycrystalline sample may not largely differ from that of a single crystalline sample. We analyzed the magnet without a correction. There are two types of crystal structure for $PuGa_3$. One is trigonal type ($R-3m$) and the other is hexagonal DO19 type ($P6_3/mmc$) [158]. The former shows the ferromagnetic transition at $T_C = 20$ K and the latter the antiferromagnetic one at Neel temperature $T_N = 24$ K. The analysis was on the data taken on polycrystalline sample for the former trigonal $PuGa_3$. We estimate possible errors of the parameter shown in

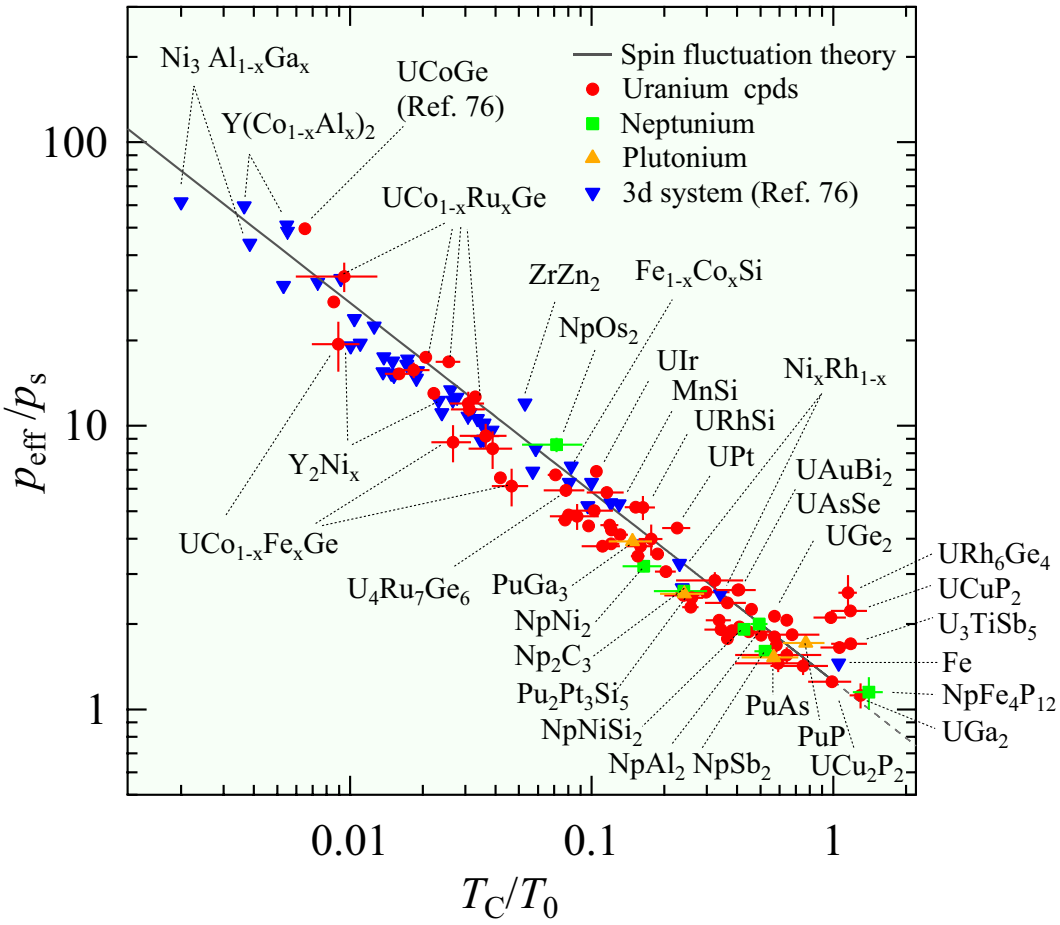

FIG. 3. Generalized Rhodes-Wohlfarth plot. Data points for uranium, neptunium, and plutonium compounds are plotted as closed circles, squares, and triangles, respectively. The data of UCoGe are cited from Ref. [76]. Data for intermetallic ferromagnetic compounds of the transition 3d metals cited from Refs. [74–76] are represented as closed antitriangles. The solid line is the theoretical relation between T_C/T_0 and p_{eff}/p_s, Eq. (11) in Takahashi's spin fluctuation theory [76].

Figs. 3 and 4. considering several factors that give uncertainty in the analyses on data taken on polycrystalline samples.

V. DISCUSSIONS

We have analyzed the basic magnetic data of the actinide ferromagnets with Takahashi's spin fluctuation theory and found that the theoretical relation in the theory applied to 3d system is satisfied also for most of the actinide ferromagnets except in several cases. This suggests that relevant factors for the magnetic properties in actinide ferromagnetism are related to the itinerancy of the 5f electrons. The deviation of some data points at $T_C/T_0 \sim 1$ is expected since the theory assumes the local moment system at $T_C/T_0 = 1$ where other factors arising from the localized character of the 5f electrons not included in the theory set in. In this section, we discuss the consequences of the present analysis from several points of views.

First we discuss the applicability of spin fluctuation theory to the actinide 5f systems. To date, there has been no experimental study for the upper limit of T_C/T_0 where the spin fluctuation theory can be applied. The result shown in Fig. 3 suggests that spin fluctuation theory is valid for $T_C/T_0 < 1$. We check the applicability of the theory more carefully. Here, we discuss the relative change between the experimental data of $(p_{eff}/p_s)_{exp}$ and theoretical value from the relation $p_{eff}/p_s = 1.4(T_C/T_0)^{-2/3}$ [Eq. (11)] in Takahashi's spin fluctuation theory. The relative change is defined as $\Delta/(p_{eff}/p_s)_{theory}$, where $\Delta = (p_{eff}/p_s)_{exp} - f(T_C/T_0)$ and $f(T_C/T_0) = 1.4(T_C/T_0)^{-2/3}$. Figure 5(a) shows the T_C/T_0 dependence of $\Delta/f(T_C/T_0)$. As can been seen from Fig. 5(a), the mean value of $\Delta/f(T_C/T_0)$ for $T_C/T_0 < 1.0$ is negative, indicating a systematic tendency that the data points of $(p_{eff}/p_s)_{exp}$ are located lower than the theoretical expected values. To see the T_C/T_0 dependence of p_{eff}/p_s more clearly, we fit the data of p_{eff}/p_s for $T_C/T_0 \ll 1$ with the function $f'(T_C/T_0) = a(T_C/T_0)^{-2/3}$, where a is a fitting parameter. The value of a is determined as 1.23. Figure 5(b) shows the T_C/T_0 dependence of $\Delta'/f'(T_C/T_0)$, where $\Delta' = (p_{eff}/p_s)_{exp} - f'(T_C/T_0)$. These results suggest that the relation $f(T_C/T_0) = 1.4(T_C/T_0)^{-2/3}$ is satisfied for $T_C/T_0 \to 1$. The 5f electrons in these ferromagnets should be basically regarded as being itinerant for $T_C/T_0 < 1.0$.

We discuss the coefficient $[1/(10C_{4/3}dy/dt)]^{-1/2}$ in the relation between p_{eff}/p_s and $(T_C/T_0)^{-2/3}$ expressed by Eq. (11). In the Takahashi's spin fluctuation theory, dy/dt is weakly

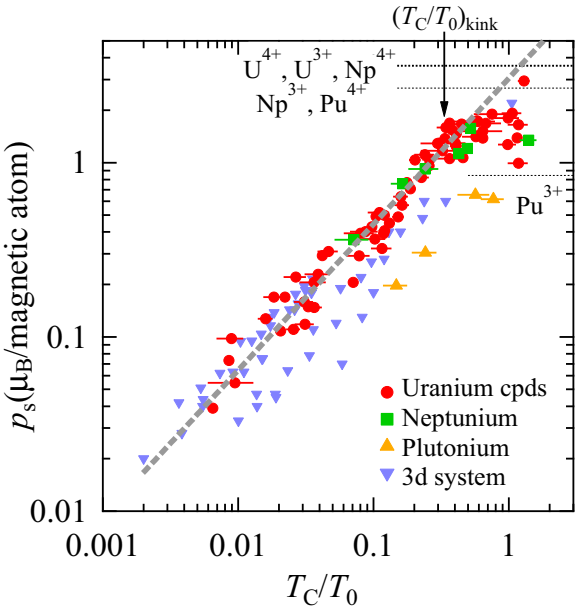

FIG. 4. Relations between the spontaneous magnetic moment p_s and T_C/T_0 for uranium, neptunium, and plutonium compounds plotted as closed circles, squares, and triangles, respectively. Data points for intermetallic ferromagnetic compounds of the transition 3*d* metals represented as antitriangles are cited from Refs. [74–76]. The dotted line is a fit to the data of the uranium and neptunium ferromagnets for $T_C/T_0 < (T_C/T_0)_{kink} = 0.32 \pm 0.02$ with the function $p(T_C/T_0) = a(T_C/T_0)^{-n}$ where a and n are fitting parameters.

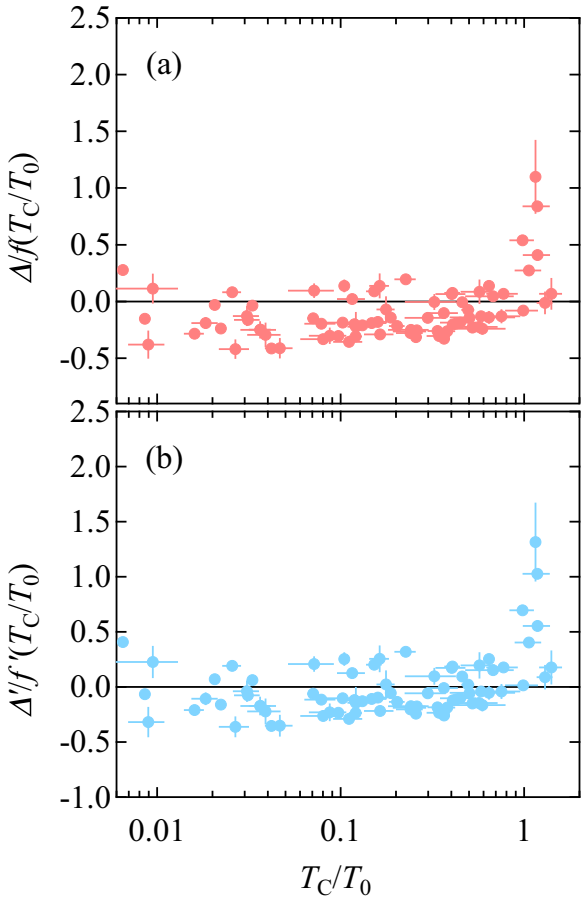

FIG. 5. (a) Relative change $\Delta/f(T_C/T_0)$ as a function of T_C/T_0 for the actinide ferromagnets. $\Delta = (p_{eff}/p_s)_{exp} - f(T_C/T_0)$, where $f(T_C/T_0) = 1.4(T_C/T_0)^{-2/3}$ [Eq. (11)] from the Takahashi's spin fluctuation theory. (b) Relative change $\Delta'/f'(T_C/T_0)$ as a function of T_C/T_0 where $\Delta' = (p_{eff}/p_s)_{exp} - f'(T_C/T_0)$ and $f'(T_C/T_0) = a(T_C/T_0)^{-2/3}$. The value of a is determined from the fit with the function $f'(T_C/T_0)$ to the data points of $(p_{eff}/p_s)_{exp}$ for $T_C/T_0 < 1.0$.

temperature dependent, which gives the Curie-Weiss like behavior of the magnetic susceptibility χ. The coefficient weakly depends on magnetic properties specific to ferromagnets and temperature regions where the effective magnetic moment p_{eff} is determined from the temperature dependence of χ. As mentioned in the previous section, the value of the coefficient is estimated as 1.4 from comparisons between the theory and experimental data on itinerant ferromagnets in the 3*d* systems [76,77]. The results shown in Figs. 5(a) and 5(b) suggest that the coefficient in actinide 5*f* systems is smaller than that in the 3*d* systems.

One important consequence of the present study is that Takahashi's spin fluctuation theory can be applied to the actinide ferromagnets whose spontaneous magnetic moment p_s is in the order of 1 μ_B/U. This may be reasonable because the theory assumes that the mean-square amplitude of the local spin fluctuation $\langle S_L^2 \rangle_{total}$ is constant as a function of temperature. This is contrary to the early spin fluctuation theories whose applicability is limited to the weak coupling limit. The assumption here is of the constant $\langle S_L^2 \rangle_{total}$ may be effective to the *f*-electron system of the rare-earth and the actinide compounds where the intra-atomic Coulomb interaction between the *f* electrons is quite large. As mentioned in the introduction, Moriya's SCR theory was later extended to the *f* electron system by the application of the constant $\langle S_L^2 \rangle_{total}$ on the standard *s*-*f* model [57]. The validity of the theory has been confirmed in a number of experimental studies [70–73].

Figures 6(a) and 6(b) show the relation between p_{eff}/p_s and T_C/T_0 in two uranium ferromagnetic systems: (a) U*TX* (*T*: Co, Rh, Ir, and Pt, *X*: Al, Ga, and Sn) series with hexagonal ZrNiAl-type crystal structure and (b) ferromagnetic superconductors URhGe and UCoGe and related doping systems with orthorhombic TiNiSi-type structure. The two systems have been extensively studied for the effect of alloying on the transition metal site that strongly affects the basic magnetic properties such as the spontaneous magnetic moment p_s or the Curie temperature T_C. A magnetic to nonmagnetic transition occurs at around $x = 0.77$ in $UCo_{1-x}Ru_xAl$ [144], $x = 0.31$ in $UCo_{1-x}Ru_xGe$ [145], and $x = 0.22$ in $UCo_{1-x}Fe_xGe$ [148,160], where anomalous physical properties around the ferromagnetic instability have been reported. It should be stressed here that the basic ferromagnetic properties are changed by the doping following the Takahashi's spin fluctuation theory similar to itinerant ferromagents in the 3*d* system $Y(Co_{1-x}Al_x)_2$ [85,86], $Ni_3Al_{1-x}Ga_x$ [92], $(Fe_{1-x}Co_x)_3Mo_3N$ [96], $Fe_xCo_{1-x}Si$ [88], and Ni_xRh_{1-x} [76] as shown in Fig. 6(c). This is in contrast with the rare-earth Ce or Yb 4*f* systems where competition between the magnetic intersite RKKY interaction $T_{RKKY} \propto J_{cf}{}^2 D(\epsilon_F)$ and the demagnetizing Kondo effect $T_K \propto \exp[-1/J_{cf} D(\epsilon_F)]$ has

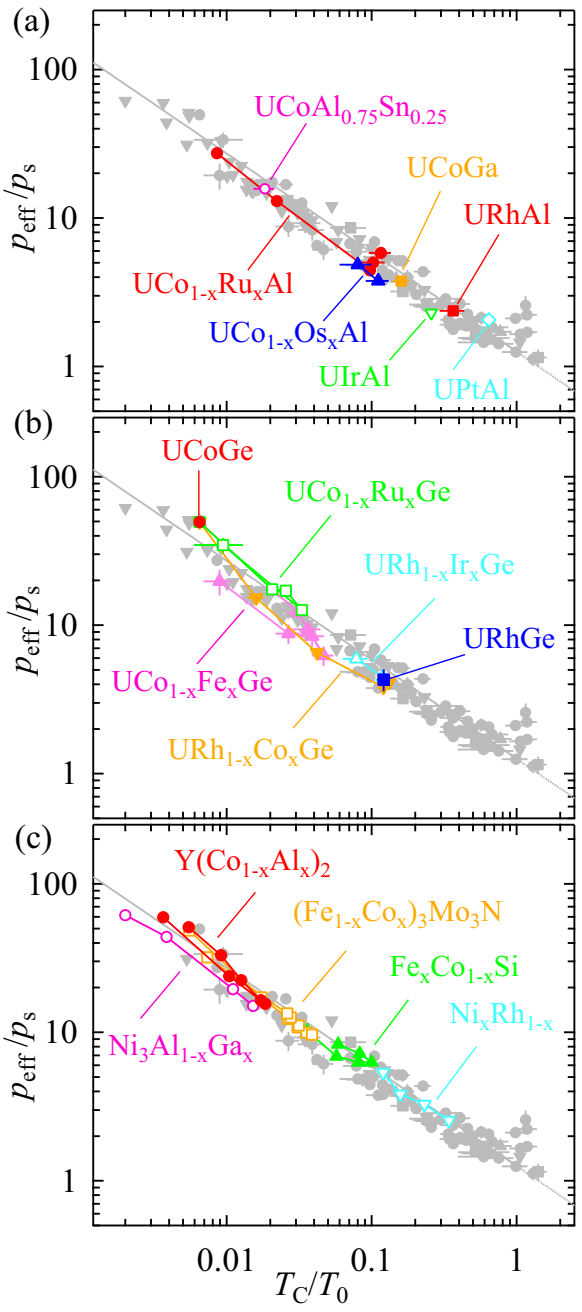

FIG. 6. Relation between p_{eff}/p_s and T_C/T_0 in (a) a UTX (T: Co, Rh, Ir, and Pt, X: Al, Ga, and Sn) series with hexagonal ZrNiAl-type crystal structure, (b) ferromagnetic superconductors URhGe and UCoGe and related doping systems with orthorhombic TiNiSi-type structure, and (c) itinerant ferromagents in the 3d system $Y(Co_{1-x}Al_x)_2$ [85,86], $Ni_3Al_{1-x}Ga_x$ [92], $(Fe_{1-x}Co_x)_3Mo_3N$ [96], $Fe_xCo_{1-x}Si$ [88], and Ni_xRh_{1-x} [76].

been described by the Doniach diagram [161]. Here J_{cf} is the exchange constant between the f and conduction electrons.

Next, we discuss the deviations of the data points between T_C/T_0 and p_{eff}/p_s in U_3TiSb_5, $UCuP_2$, $UCuAs_2$, and URh_6Ge_4 with $T_C/T_0 \sim 1.0$. The values of p_{eff}/p_s of these ferromagnets are larger than unity. As mentioned before, spin fluctuation theory assumes localized magnetism at $T_C/T_0 = 1$ and the deviations may be due to the characteristic feature of the localized 5f electrons not included in the theory. We suggest a crystalline electric field effect (CEF) from surrounding ligand atoms on the 5f electrons can play an important role for the deviations as follows. The anisotropic magnetic property in UCu_2P_2, $UCuP_2$, and $UCuAs_2$ has been explained by the CEF effect on the localized 5f electrons [130,162]. Let us discuss the effect of the CEF potential on the 5f electrons of the actinide ion based on the LS coupling scheme [1–3,7]. The CEF potential splits the degenerate J-multiplet of the actinide ions U^{4+} (5f^2 configuration, total angular momentum $J = 4$), U^{3+} and Np^{4+} (5f^3, $J = 9/2$), Np^{3+} and Pu^{4+} (5f^4, $J = 4$), and Pu^{3+} (5f^5, $J = 5/2$) into several separated energy levels (CEF level). The CEF effect depends on the site symmetry of the actinide ion in a crystal and surrounding ligand atoms. Generally, the value of the spontaneous magnetic moment p_s in the ferromagnetic state at $T = 0$ K corresponds the expected value of gJ_z of the CEF ground state and is smaller than that of the free actinide ion without the CEF effect. When the temperature is raised, the excited states of the CEF levels are populated and contribute to the magnetic property at finite temperature. Naturally, the effective magnetic moment p_{eff} at the higher temperature region is larger than p_s. Of course, the ratio p_{eff}/p_s depends on the CEF level scheme of each ferromagnet. This may be a reason of the deviations of the data points for $T_C/T_0 \sim 1.0$. The scatterings of the data between T_C/T_0 and p_s at around $T_C/T_0 \sim 1.0$ shown in Fig. 4 may be also attributed to the CEF effect. There is no large deviation of the data from the theoretical relation for $T_C/T_0 < 1$, suggesting that CEF effects on the 5f electrons becomes substantially weaker. There has been no report of observation of CEF excitation in the actinide ferromagnets for $T_C/T_0 < 1$.

The numbers of the actinide ferromagnets close to $T_C/T_0 \sim 1.0$ is only about 10% of the total being analyzed in this study and the other ferromagnets follow the spin fluctuation theory. This fact suggests that the localized character of the 5f electrons is rare among the actinide ferromagents. It is noted that a CEF excitation in the inelastic neutron-scattering spectrum, evidence of the localized character of the 5f electrons, has been observed in only a few compounds such as UPd_3 [163], UPdSn [164], UGa_2 [165], and $U_3Pd_{20}Si_6$ [166–168]. It is interesting to consider the "localized 5f electrons state" from experimental studies on UGa_2 and $U_3Pd_{20}Si_6$. The former is the ferromagnet analyzed in this study and the latter shows successive antiferromagnetic and ferromagnetic transitions at low temperatures [166]. Although the localized character of the 5f electrons has been suggested from the observations of the CEF excitations in the inelastic neutron-scattering studies, peak intensities are generally weak compared with rare-earth compounds [165,168]. The overall features of the nuclear spin-lattice relaxation rate $1/T_1$ in NMR experiments cannot be interpreted based on the localized moment system for both compounds [169,170], suggesting that the low-energy spin dynamics due to the hybridization between the 5f and conduction electrons. The nature of the localized state of the 5f electrons state may differ from that of the rare-earth 4f electrons systems.

A number of actinide compounds show physical properties reminiscent of the Kondo effect as mentioned in the Introduction. Among the actinide ferromagnets analyzed in this paper,

the Kondo-like logarithmic temperature dependence of the resistivity ($\rho \sim -\ln T$) has been reported in $UCo_{0.5}Sb_2$($T_C/T_0 = 0.240$) [171], UPS(0.260) [124], $UAuBi_2$(0.407) [129], $UCu_{0.9}Sb_2$(0.448) [127], USbSe(0.676) [172], UTe(0.751) [173], $UCuAs_2$(0.979) [130,174], USbTe(1.06) [172], UGa_2(1.12) [105], $UCuP_2$(1.18) [130], $NpNiSi_2$(0.427) [154], and $NpFe_4P_{12}$ (1.40) [155]. The coexistence of the Kondo effect and the ferromagnetic order in actinide systems has been theoretically studied with $S = 1$ underscreened Anderson and Kondo lattice models [29,30]. The minimum value of T_C/T_0 among those of the ferromagnets is 0.240 for $UCo_{0.5}Sb_2$. It is interesting to note that this value is comparably close to $(T_C/T_0)_{\rm kink}(= 0.32 \pm 0.02)$ where the kink is located in the relation between T_C/T_0 and p_s as shown in Fig. 4. For $(T_C/T_0)_{\rm kink} < T_C/T_0$, p_s increases more weakly as a function of T_C/T_0 and the values of p_s are smaller than extrapolated ones with the fitted line in the uranium and neptunium ferromagnets. One possible interpretation is that p_s is suppressed by a Kondo-like spin-compensating mechanism. Some of the physical properties of the actinide compounds have been partly explained by taking into account Kondo or the CEF effect, but the two effects have not been well established theoretically in actinide metallic systems as mentioned in the Introduction. The main message from this analysis is that the $5f$ electrons in the actinide ferromagnets should be treated as being itinerant for $T_C/T_0 < 1.0$. Further theoretical study is necessary to understand the dual nature of the $5f$ electrons in actinide systems.

VI. SUMMARY

We have analyzed the magnetic data of 69 uranium, 7 neptunium, and 4 plutonium ferromagnets with Takahashi's spin fluctuation theory. The analysis has been carried out using our experimental data for 17 uranium ferromagnets (group I). Data taken from the literature were analyzed for the remaining uranium (group II), neptunium, and plutonium ferromagnets. We have determined the basic and the spin fluctuation parameters of the ferromagnets and discuss the applicability of spin fluctuation theory to the actinide $5f$ electrons system. The ratio of the effective magnetic moment and the spontaneous one, $p_{\rm eff}/p_s$, follows the generalized Rhodes-Wohlfarth relation, viz. $p_{\rm eff}/p_s \propto (T_C/T_0)^{-3/2}$, predicted by Takahashi's spin fluctuation theory in the actinide ferromagnets for $T_C/T_0 < 1.0$, similarly to itinerant ferromagnets in the $3d$ transition metals and their intermetallics. This result suggests that the itinerant nature of the $5f$ electrons in the actinide ferromagnets and that the magnetic properties of the ferromagnets can be basically understood in the framework of the spin fluctuation theory. Meanwhile, data points between T_C/T_0 and $p_{\rm eff}/p_s$ deviate from the theoretical relation in several ferromagnets with $T_C/T_0 \sim 1.0$, which may be due to the effect of the CEF on the $5f$ electrons. The value of the spontaneous magnetic moment p_s increases linearly as a function of T_C/T_0 in the uranium and neptunium ferromagnets below $(T_C/T_0)_{\rm kink} = 0.32 \pm 0.02$ where a kink structure appears in relation between the two quantities. p_s increases more weakly above $(T_C/T_0)_{\rm kink}$. A possible interpretation with the T_C/T_0-dependence of p_s is given in terms of the Kondo effect.

ACKNOWLEDGMENTS

The authors thank Prof. A. V. Andreev and Dr. D. Gorbunov for providing their unpublished data of some uranium ferromagnets and Prof. Y. Takahashi for helpful advice on spin fluctuation theory. We acknowledge discussions with Dr. K. Kaneko, Dr. N. Metoki, Dr. S. Kambe, Dr. K. Deguchi, Prof. N. K. Sato, and Prof. H. Yamagami. We also thank Prof. Z. Fisk for discussions and his editing of this paper. This work was supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grants No. 16K05463, No. 16K05454, No. 16KK0106, No. JP15H05884 (J-Physics), and No. 26400341.

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