Theoretical Development in Itinerant Electron Ferromagnetism

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Abstract. In spite of the successful explanation of the Curie-Weiss law temperature dependence of the magnetic susceptibility, there exist some difficulties in the conventional spin fluctuation theory based on the expansion in powers of fluctuation amplitudes. They are now known to originate from the severe restriction on the magnetization curve. In this review, we show another approach based on the new ideas, i.e. the local spin amplitude conservation and the simultaneous treatment of temperature and external magnetic field dependences. We show various interesting magnetic properties are derived based on them and they are compared with experiments.

1. Introduction

Significant roles of spin fluctuations on various magnetic properties of itinerant electron magnets have been recognized since the publication of the paper by Moriya and Kawabata [1, 2] in 1973. The theory, now known as the SCR theory, has been successful in deriving the observed Curie-Weiss law temperature dependence of magnetic susceptibility of weak itinerant ferromagnets, such as $ZrZn_2$, Sc_3In, and MnSi. Experimentally, the presence of spin fluctuations in these magnets has been also suggested from the enhancement of the temperature linear coefficient of the specific heat at low temperatures, and the direct observation of the fluctuation amplitudes by using the polarized neutron scattering measurements. Around 1980, the theory has been regarded to be well established except for a trivial problem [3], i.e. the spontaneous magnetic moment does not vanish at the critical point with increasing temperature.

The purpose of this article is to make a brief review on the development of spin fluctuation theories, starting from the middle of 1980 in order to overcome the above difficulty of the SCR theory. It was found at that time [4] that it originates from the basic assumption of the theory, i.e. the linearity of the Arrott plot of the magnetization curve satisfied independent of temperature. At the same time, it seems better to rely on another approach, and new theory was proposed as an alternative candidate. It is based on the spin amplitude conservation and the non negligible effects of the zero-point spin fluctuations [4, 5]. As the consequence, no assumption on the magnetization curve becomes necessary between the external magnetic field H and the induced magnetization M. It was also quite successful in predicting various magnetic properties, for instance, the critical magnetic isotherm, $H/M \propto M^4$, in contradiction to the assumption of the SCR theory.

In section 2, three theories of itinerant electron magnetism and the magnetic properties predicted by them around the critical temperature are compared with each other. Section 3 is devoted to the explanation of spin fluctuation theory based on the local spin amplitude conservation. In section 4, theoretically derived magnetic properties are compared with experiments. Finally, a brief summary is presented in the last section.

2. Brief comparison of the consequences of three theories of itinerant magnetism

As with the other thermodynamic properties, magnetic properties are derived by the following free energy as a function of magnetization M and temperature T.

$$F(M,T) = F(0,T) + \frac{1}{2}a(T)M^2 + \frac{1}{4}b(T)M^4 + \frac{1}{6}c(T)M^6 + \cdots . \tag{1}$$

For instance, the magnetic isotherm, i.e. the relation between the external magnetic field H and its induced magnetization M, is given by

$$H = \frac{\partial F(M,T)}{\partial M} = a(T)M + b(T)M^3 + c(T)M^5 + \cdots . \tag{2}$$

The inverses of magnetic susceptibilities, $\chi^{-1}(T)$ and $\chi_z^{-1}(T)$, are also defined by

$$\begin{aligned} \chi^{-1}(T) &= \frac{H}{M} = a(T) + b(T)M^2 + c(T)M^4 + \cdots , \\ \chi_z^{-1}(T) &= \frac{\partial H}{\partial M} = a(T) + 3b(T)M^2 + 5c(T)M^4 + \cdots , \end{aligned} \tag{3}$$

corresponding to the applied magnetic field perpendicular and parallel direction to the static induced magnetic moment M in z-direction. The M-dependent terms in the above right hand side are neglected in the weak field limit. In the ordered phase below T_C, (3) is rewritten in the form,

$$\frac{H}{M} = b(T)\left[M^2 - M_0^2(T)\right] + \cdots , \quad M_0^2(T) = -\frac{a(T)}{b(T)}, \tag{4}$$

where $M_0(T)$ is the spontaneous magnetic moment.

In the following, we show how these magnetic properties are treated based on the three different theories of itinerant electron magnetism.

2.1. Stoner-Wohlfarth theory

This theory is based on the band theoretical idea. Therefore, the temperature dependences of various magnetic properties originate from that of the Fermi distribution function. According to this theory, the temperature dependence of the inverse of magnetic susceptibility, $\chi^{-1}(T)$, and the spontaneous magnetization squared, $M_0^2(T)$, are given by

$$\chi^{-1}(T) \propto (T^2 - T_C^2), \quad M_0^2(T) \propto (T_C^2 - T^2). \tag{5}$$

Around the critical temperature T_C, they are also rewritten as follows.

$$\chi^{-1}(T) \propto (T - T_C)^{\gamma}, \quad M_0^2(T) \propto (T_C - T)^{\beta}, \tag{6}$$

where $\gamma = 1$ and $\beta = 1/2$. In the same way, the magnetization curve at the critical point, $T = T_C$, is given by $H \propto M^3$, i.e. $M \propto H^{1/\delta}$ with $\delta = 3$, because $a(T_C) = 0$ is satisfied in (3). It means that the scaling law relation, $\gamma = \beta(\delta - 1)$, of the theory of phase transition is satisfied.

2.2. SCR theory of spin fluctuations
In contrast to the Stoner-Wohlfarth theory, the effect of thermal spin fluctuations is included in the free energy in this theory. Consequently, the temperature dependence of $a(T)$ in (3) is rather dominated by the effect of spin fluctuations. Therefore, the following temperature dependences of $\chi(T)$ and $M_0(T)$ have been derived around the critical point.

$$\chi^{-1}(T) \propto (T - T_C)^2, \quad M_0(T) \propto (T_C - T)^{1/2}. \tag{7}$$

Their critical indexes are given by $\beta = 2$ and $\gamma = 1/2$, respectively. On the other hand, the second coefficient $b(T)$ in the right hand side of (2) is assumed to be constant. Higher order terms than this M^3 term are also neglected in this theory. The exponent of the critical magnetic isotherm, $M \propto H^{1/\delta}$, is given by $\delta = 3$, just as in the Stoner-Wohlfarth theory. Therefore, the scaling law relation between these indexes is violated.

The above argument clearly indicates the necessity that the effect of spin fluctuations on the magnetization curve has to be taken into account. The exponent $\delta = 3$ between H and M should be modified, and rather $H \propto M^5$ is satisfied in contradiction to $b(T) > 0$ at the critical point. It also means that the linearity of the Arrott plot of the magnetization curve is not generally satisfied.

2.3. Spin fluctuation theory based on TAC and GC
This theory has been proposed to overcome the difficulty of the SCR theory. As for the temperature dependence, the same dependences as (7) derived by the SCR theory are satisfied. However, the different critical exponent $\delta = 5$ is obtained for the magnetic isotherm, $M \propto H^{1/\delta}$. The scaling law relation is therefore satisfied in this case.

In summary, both the temperature and the magnetic field dependence of spin fluctuation amplitudes have to be treated simultaneously in order to satisfy the scaling law relation. In other words, temperature dependence of the magnetization curve is not so simple, contrary to the expectations of the SCR theory.

3. Development beyond the SCR Theory
In this section, after the origin of the difficulty of the SCR theory is briefly explained, another approach based on the ideas of TAC and GC is presented.

3.1. Difficulty of the SCR theory
As shown in (2) and (3), the magnetization curve, the relation between M and H, is generally given by

$$H = a(T)M + b(T)M^3 + \cdots, \quad \therefore \ \frac{H}{M} = a(T) + b(T)M^2 + \cdots. \tag{8}$$

In terms of above equations, the assumption of the SCR theory is stated as follows.

(i) The temperature dependence of the coefficient $b(T)$ in (8) is neglected.
(ii) The higher order expansions in powers of M than those explicitly presented in (8) are assumed to be absent or neglected.

Main interest of this theory is focused on the first coefficient $a(T)$.

As will be shown later, the thermal spin fluctuation amplitudes is regarded as a function of temperature T, and the magnetic susceptibilities, i.e. $\chi(M,T)$ and $\chi_z(M,T)$ of the perpendicular and the parallel components with respect to the static magnetic moment. Around the critical point, they are dominated by the dependence proportional to $\chi^{-1/2}(M,T)$ and $\chi_z^{-1/2}(M,T)$. Then the M^2 term in $\chi_z^{-1}(M,T) \simeq \chi^{-1}(M,T) + 2bM^2 \to 2bM^2$ in the limit of $T \to T_C$, gives rise to the M linear term in the thermal fluctuation amplitude. It is known that this M-linear term is the origin of the non vanishing spontaneous magnetization at $T = T_C$. In other words, the difficulty is closely related to the temperature independent assumption of $b(T)$ in this theory.

3.2. Theory based on the spin amplitude conservation

In order to overcome the difficulties of the SCR theory, another spin fluctuation theory was proposed [4, 5]. It is based on the following two basic ideas [6].

(i) The total spin amplitude conservation (TAC)
It means that the average of the local spin amplitude squared on each magnetic site remains constant independent of temperature, and whether in the presence or absence of the external magnetic field.

(ii) The global consistency (GC) in the H-M space
No assumption is made on the magnetization curve in this theory, in contrast to (8) with T-independent $b(T)$ in the SCR theory. To find the magnetization curve rather becomes of the target of this theory. This is what the global consistency (GC) means.

Thermal average of spin amplitude squared on each magnetic atom or ion is generally written as the sum of three components,

$$\langle \mathbf{S}_{\rm loc}^2 \rangle = \sigma^2 + \langle \delta \mathbf{S}_{\rm loc}^2 \rangle_T + \langle \delta \mathbf{S}_{\rm loc}^2 \rangle_Z \,, \tag{9}$$

where $\mathbf{S}_{\rm loc}$ is the local spin operator in units of $2\mu_{\rm B}$ and the first term in the right hand side represents the spontaneous local spin amplitude squared. The last two terms represent the averages of thermal and zero-point fluctuation amplitudes. In the paramagnetic phase for simplicity, according to the fluctuation-dissipation theorem of statistical mechanics, the equal-time autocorrelation of the local spin amplitude of the left hand side of (9) is represented by

$$\langle \mathbf{S}_{\rm loc}^2 \rangle = \frac{3}{N_0^2} \sum_{\mathbf{q}} \int_0^\infty \frac{{\rm d}\omega}{\pi} \coth(\omega/2k_{\rm B}T) {\rm Im}\chi(\mathbf{q}, \omega). \tag{10}$$

The imaginary part of the dynamical magnetic susceptibility in units of $(2\mu_{\rm B})^2$ in the above integrand is given in the following double Lorentzian form:

$$\begin{aligned} {\rm Im}\chi(\mathbf{q}, \omega) &= \chi(\mathbf{q}, 0) \frac{\omega \Gamma_q}{\omega^2 + \Gamma_q^2}, \quad \chi(\mathbf{q}, 0) = \chi(0, 0) \frac{\kappa^2}{q^2 + \kappa^2}, \\ \Gamma_q &= \Gamma_0 q(\kappa^2 + q^2) = 2\pi T_0 x(y + x^2), \quad y = \frac{\kappa^2}{q_B^2}, \; x = \frac{q}{q_B}, \\ T_0 &= \Gamma_0 q_B^3/2\pi, \quad \frac{N_0}{\chi(q_B, 0)} = \frac{N_0(q_{\rm B}^2 + \kappa^2)}{\kappa^2 \chi(0, 0)} = \frac{N_0(1 + y)}{y\chi(0, 0)} \simeq 2T_A. \end{aligned} \tag{11}$$

The number of magnetic atoms, the correlation wave-number squared, and the damping constant are, respectively, denoted by N_0, κ^2, and Γ_q. The zone boundary wave-number is denoted by q_B. As the measures of the distribution widths of fluctuation amplitudes in frequency and wave-vector space, two temperature scales, T_0 and T_A, are also introduced. From the decomposition, $\coth(\omega/2k_{\rm B}T) = 1+2n(\omega)$, the total amplitude in (10) is split into the thermal and the zero-point amplitudes as given by

$$\begin{aligned} \langle \mathbf{S}_{loc}^2 \rangle_{\rm T} (y, t) &= \frac{6}{N_0^2} \sum_q \int_0^\infty \frac{{\rm d}\omega}{\pi} n(\omega) {\rm Im}\chi(\mathbf{q}, \omega) = \frac{9T_0}{T_A} A(y, T), \\ A(y, t) &= \int_0^1 {\rm d}x x^3 \left[\log u - \frac{1}{2u} - \psi(u) \right], \quad u = x(y + x^2)/t, \\ \langle \mathbf{S}_{loc}^2 \rangle_{\rm Z} (y) &= \frac{3}{N_0^2} \sum_q \int_0^\infty \frac{{\rm d}\omega}{\pi} {\rm Im}\chi(\mathbf{q}, \omega) = \langle \mathbf{S}_{loc}^2 \rangle_{\rm Z} (0) - \frac{9T_0}{T_A} c\, y, \end{aligned} \tag{12}$$

where $\psi(u)$ in the integrand of the above second line represents the digamma function. Both amplitudes depend on the parameter y through its dependence of $\chi(\mathbf{q}, 0)$ and Γ_q in (11). The thermal amplitude has an extra temperature dependence through the Bose distribution function $n(\omega)$.

3.3. Basic Equation for magnetic properties

The condition of the local spin amplitude conservation is regarded as the basic equation of the TAC-GC theory. If we compare the condition of (9) at any temperature and under the external magnetic field with the reference condition at the critical point with no external magnetic field, the following equation is derived by substituting (12).

$$\begin{aligned} &2A(y,t) + A(y_z,t) - c(2y + y_z) + \lambda\sigma^2 = 3A(0,t_c), \quad \lambda = \frac{T_A}{3T_0}, \\ &y_z(\sigma,t) = y(\sigma,t) + \sigma\frac{\partial y(\sigma,t)}{\partial\sigma}, \end{aligned} \tag{13}$$

The inverses of the perpendicular and the parallel components of magnetic susceptibilities are denoted by $y \propto H/M$ and $y_z \propto \partial H/\partial M$, respectively, with respect to the direction of the spontaneous moment. The spin moment per magnetic atom is denoted by σ in units of $2\mu_{\mathrm{B}}$. The important point of (13) is that it is regarded as an ordinary differential equation of $y(\sigma, t)$ as the function of σ. Therefore the σ dependence of $y(\sigma, t)$ is obtained by solving this equation. There is also no need to restrict the highest power of $y(\sigma, t)$ as a function of σ. The σ-dependence of $y(\sigma, t)$ is determined in this way by the global condition (GC) of (13) over the wide range of the variable σ.

In the absence of spontaneous or induced magnetic moment by the external magnetic field, both $y_z = y$ and $\sigma = 0$ are satisfied simultaneously. Then the temperature dependence of the magnetic susceptibility above T_c as well as the spontaneous magnetic moment below T_c are also obtained by solving the equation.

4. Magnetic Properties in More Detail

Let us show in this section, various interesting magnetic properties are derived by solving our basic equation (13). They are classified into two groups, i.e. properties that depend on external magnetic field or temperature.

4.1. Magnetic Field dependent properties

First of all, we show how magnetic isotherms in the ground state and at the critical temperature are derived as the solutions of (13). These results are also compared with experiments.

4.1.1. Magnetization curve at $T = 0$ determined by Zero-Point Spin Fluctuations In the ground state, the following differential equation is satisfied, because of the absence of the thermal spin fluctuation amplitudes in (13).

$$-c(2y + y_z) + \lambda\sigma^2 = 3A(0,t_c), \quad y_z = y + \sigma\frac{\partial y}{\partial\sigma} \tag{14}$$

The σ dependence of $y(\sigma, t)$ is easily obtained by assuming

$$y(\sigma, 0) = y_1(0)[\sigma^2 - \sigma_0^2(0)]. \tag{15}$$

Two parameters, $\sigma_0(0)$ and $y_1(0)$, correspond to the spontaneous magnetic moment $M_0(T)$ and the expansion coefficient $b(T)$, respectively, in (4) at $T = 0$. After substitution of (15) into (14), they are determined as follows.

$$y_1(0) = \frac{\lambda}{5c} = \frac{T_A}{15cT_0}, \quad \sigma_0^2(0) = \frac{1}{cy_1(0)}A(0,t_c) = \frac{5T_0}{T_A}C_{4/3}t_c^{4/3}, \tag{16}$$

where $A(0,t_c) \simeq C_{4/3}t^{4/3}/3$, with $C_{4/3} = 1.006089\cdots$, is satisfied for small t_c. As for $\sigma_0^2(0)$, the same result has been also derived by the SCR theory.

According to the thermodynamic relation in (2), we can find the magnetic free energy $F_m(M,T)$ as the function of M. If we note the definition, $y \propto H/M$, then (15) is rewritten as the relation between H and M, as shown in the first equation of (8). By integrating the thus obtained relation of H vs M with respect to M, the following free energy $F_m(M,T)$ is derived.

$$F_m(M,T) = F_m(0,T) + N_0\left[\frac{N_0}{2\chi(T)}m^2 + \frac{F_1}{4}m^4 + \cdots\right], \quad F_1 = \frac{2T_A^2}{15cT_0}, \tag{17}$$

where $m = M/(2\mu_B N_0)$. It is very interesting that the fourth expansion coefficient F_1 with respect to m is determined by using only two spectral parameters T_0 and T_A. The reason is that the magnetic isotherm in this case is determined by the effect of zero-point spin fluctuations. On the contrary, F_1 is regarded to be determined by the density of states of conduction electrons and its derivatives at the Fermi energy in the Stoner-Wohlfarth theory and in the SCR theory.

As an application of the relation of F_1 in (18), we can estimate the values of spectral parameters T_0 and T_A, only by using the results of magnetic measurements. Note the following relations are satisfied for σ_0^2 in (16) and F_1.

$$F_1 = \frac{2T_A^2}{15cT_0}, \quad \sigma_0^2 \simeq \frac{5T_0}{T_A}C_{4/3}\left(\frac{T_C}{T_0}\right)^{4/3}. \tag{18}$$

By eliminating either T_A or T_0 from the above relations in (18), the following two results are obtained.

$$\left(\frac{T_C}{T_0}\right)^{5/6} = \frac{\sigma_0^2}{5C_{4/3}}\left(\frac{15cF_1}{2T_C}\right)^{1/2}, \quad \left(\frac{T_C}{T_A}\right)^{5/3} = \frac{\sigma_0^2}{5C_{4/3}}\left(\frac{2T_C}{15cF_1}\right)^{1/3}. \tag{19}$$

These ratios of T_0/T_C and T_A/T_C in the left hand side are, therefore, estimated from (19) because only the values of σ_0, T_C, and F_1 are included in both of the right hand sides. To check the validity of F_1 in (18), values of $4T_A^2/15T_0$ are compared in Table 1 with those of F_1 estimated from the slopes of the Arrott plots of magnetization measurements. Parameters T_0 and T_A are estimated either by inelastic neutron scattering experiments or by NMR measurements. These two values in the fourth and fifth columns seem to be in fairly good agreement with each other.

4.1.2. Critical magnetic isotherm At the critical temperature $T = T_C$, our basic equation (13) is well approximated, for $y \ll 1$, by

$$\begin{aligned} 2[A(y,t_c) - A(0,t_c)] + [A(y_z,t_c) - A(0,t_c) - c(2y + y_z) + \lambda\sigma^2 \\ \simeq -\frac{\pi t_c}{4}(2\sqrt{y} + \sqrt{y_z}) + \lambda\sigma^2 = 0, \end{aligned} \tag{20}$$

because of the critical thermal amplitude $A(y,t)$ given by

$$A(y,t) - A(0,t) \simeq -\frac{\pi t}{4}\sqrt{y}. \tag{21}$$

Table 1. Comparison of values of $4T_A^2/15T_0$ and F_1.

Compounds	T_0(K)	T_A(K)	$4T_A^2/15T_0$(K)	F_1(K)	Ref.
MnSi	231	2.08×10^3	$5.0\ \times10^3$	9.7×10^3	neutron [7]
	171	2.11×10^3	6.94×10^3	–	NMR [8]
Ni_3Al	3590	3.09×10^4	0.71×10^5	–	neutron [9]
$Ni_{74.7}Al_{25.3}$	2860	4.05×10^4	1.53×10^5	1.0×10^5	NMR [10]
Sc_3In	565	2.00×10^5	0.66×10^5	2.0×10^5	NMR [11]
$ZrZn_2$	321	8.83×10^3	1.05×10^4	1.3×10^4	NMR [12]
$Y(Co_{1-x}Al_x)_2$					NMR [13]
$x = 0.13$	2290	1.16×10^4	1.57×10^4	2.1×10^4	
$x = 0.15$	2119	6.34×10^3	0.51×10^4	1.0×10^4	
$x = 0.17$	2093	7.03×10^3	0.63×10^4	1.6×10^4	

Both the y- and the y_z-linear terms in (20), originated from the zero-point fluctuations, are also neglected in this limit. The solution of (20) is easily obtained by assuming the σ dependence proportional to σ^4, as given by

$$y(\sigma, t_c) = y_c\sigma^4, \quad y_c = \left[\frac{4\lambda}{\pi t_c(2+\sqrt{5})}\right]^2. \tag{22}$$

The observed magnetization curve of the MnSi [14] will be a good candidate to confirm the above σ dependence. The linearity of the Arrott plot of this compound was known not to be satisfied around the critical temperature as shown in Figure 2 (right). Its origin was sometimes

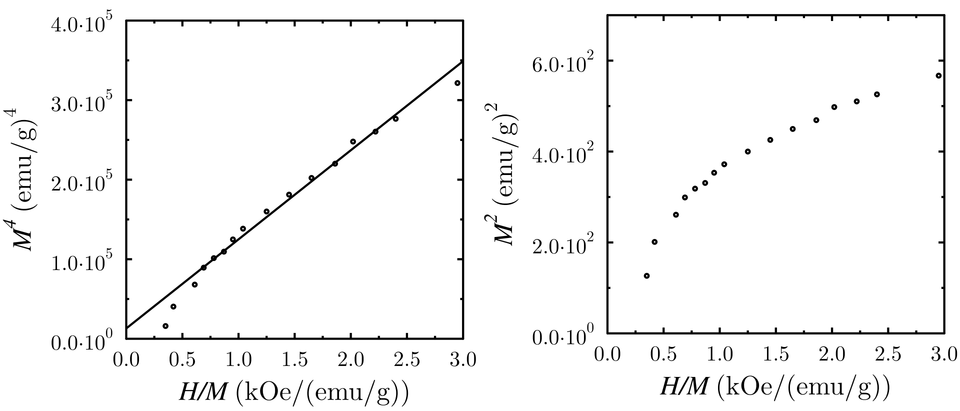
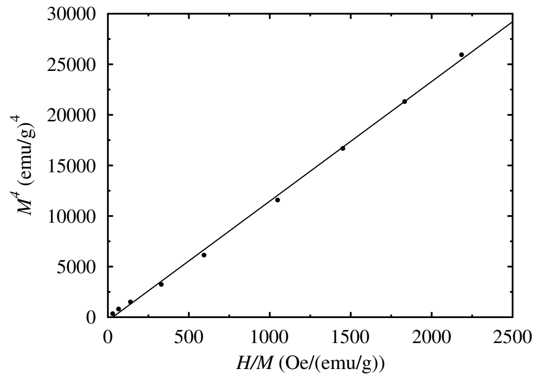
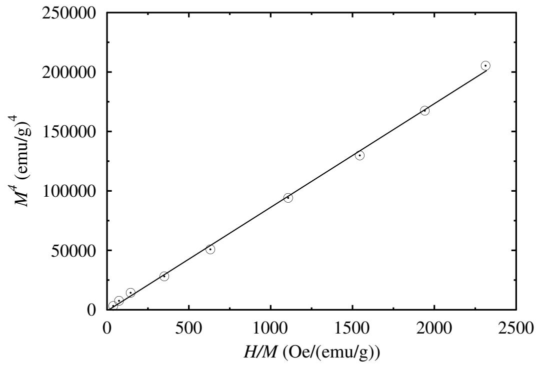


Figure 1. Magnetization curve of MnSi plotted by M^4 vs H/M (left) and M^2 vs H/M (right) at $T = 29$ K.

ascribed to the relatively larger magnitude of spontaneous magnetic moment of this compound. Therefore, it was only later that the critical M^4 vs H/M relation was found to be well satisfied as shown in the left figure. Recently good linear relations are observed for Ni [15] and the Heusler compound Co_2CrGa [16] by Nishihara *et al* , for instance, as shown in Figure 2.

Figure 2. Critical magnetic isotherms of Ni (left) and Co_2CrGa (right) by Nishihara *et al* .





For convenience of the quantitative comparison of the theoretical prediction with experiments, the critical dependence of (22) is also written as follows.

$$\left(\frac{M}{M_g}\right)^4 = 1.20 \times 10^6 \frac{T_C^2}{w_A T_A^3 p_s^4} \frac{H}{M_g}, \tag{23}$$

where w_A is the molar weight per magnetic atom or ion, and H and M_g are measured in units of kOe and emu/g, respectively. From the experimentally observed slope of the M^4 vs H/M plot, we can estimate the value of T_A with using the values of T_C and p_s. The values of $T_A^{(c)}$ estimated in this way are shown in Table 2 for MnSi [14] and (FeCo)Si by Shimizu *et al* [17]. They are fairly in good agreement with $T_A^{(g)}$ estimated from the magnetization curve in the ground state.

Table 2. Comparison of the values of T_A estimated by (23) and those in the ground state.

Compound	$T_A^{(g)}$ (10^4 K)	$T_A^{(c)}$ (10^4 K)	Ref.
MnSi	0.218	0.129	[14]
$Fe_xCo_{1-x}Si$			[17]
$x = 0.36$	1.179	0.727	
0.48	0.998	0.727	
0.67	0.987	0.725	
0.77	1.209	0.824	
0.88	1.518	0.917	
0.91	2.273	1.268	

As the last example, the Arrott plot of numerically evaluated magnetization curves are shown in Figure 3. As a general tendency, the almost linear relations between M^2 and H/M are observed in this figure. Initial slope of curves, however, increases towards the critical temperature, and it finally diverges to infinity.

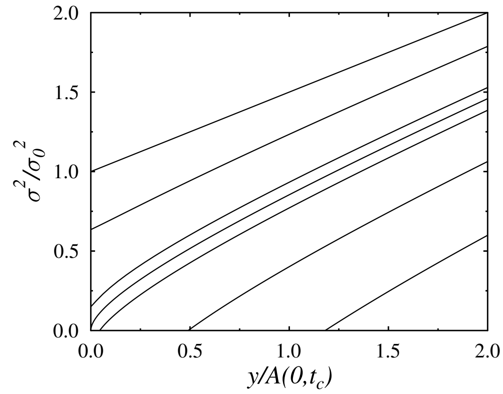


Figure 3. Arrott plot of numerically evaluated magnetization curves for $T/T_C = 0,\ 0.5,\ 0.9,\ 1,\ 1.1,\ 1.5,\ 2$ from the top.

4.2. Temperature Dependent Properties

As the typical properties of temperature dependence, the magnetic susceptibility in the paramagnetic phase and the spontaneous magnetization below the critical temperature are treated in this section.

4.2.1. Curie-Weiss law temperature dependence of magnetic susceptibility Since both of $\sigma = 0$ and $y_z = y$ are satisfied in the absence of the external magnetic field above T_C, the basic equation (13) is simply written as follows.

$$A(y,t) - c\,y = A(0,t_c). \tag{24}$$

Although it looks like the same equation of the SCR theory, the origin of the above coefficient c of the left hand side is completely different. The y-linear coefficient in this theory originates from the fourth order expansion coefficient $b(T)$ of the free energy in (1). Therefore, the almost t-linear solution of y depends on the assumption that $b(T)$ is independent of temperature. In our view, however, it depends on temperature and vanishes at the critical point, where the critical magnetic isotherm, $H \propto M^5$, is satisfied. The coefficient c in (24) rather originates from the y dependence of the amplitude of the zero-point spin fluctuations as shown in (12).

The thermal amplitude $A(y,t)$ in the left hand side of (24) increases with temperature. It is, however, compensated by the second y-linear term, to satisfy the spin amplitude conservation. The temperature dependence of the magnetic susceptibility is therefore determined as the solution $y \propto \chi^{-1}$ in (24). It is known that numerical solutions of (24) give the fairly good T-linear dependence of y, i.e. the Curie-Weiss law dependence of the magnetic susceptibility.

In the following, we show that a very interesting relation is derived from (24). If we assume the Curie-Weiss law temperature dependence of the magnetic susceptibility $\chi(T)$, its approximate T dependence is given as follows.

$$\frac{1}{N_0}\chi(T) \simeq \frac{p_{\rm eff}^2}{12(T-T_c)}, \text{ or } \frac{N_0}{\chi(T)} \simeq \frac{12(T-T_c)}{p_{\rm eff}^2}. \tag{25}$$

From the definition of $y(t)$, the following relation is also satisfied.

$$\frac{N_0}{\chi(T)} = 2T_A y(t) \simeq 2T_A\frac{{\rm d}y(t)}{{\rm d}t}(t-t_c) = \frac{2T_A}{T_0}\frac{{\rm d}y(t)}{{\rm d}t}(T-T_c). \tag{26}$$

By equating the right hand sides of $N_0/\chi(T)$ in (25) and (26), the following relation is derived.

$$\frac{6}{p_{\rm eff}^2} \simeq \frac{T_A}{T_0}\frac{{\rm d}y(t)}{{\rm d}t} \tag{27}$$

Eliminating the ratio T_A/T_0 from (27) and the second equation of (18) for σ_0^2 by substituting (27) into (18), the following relation is finally obtained [4, 5].

$$\frac{p_{\rm eff}^2}{p_{\rm s}^2} \simeq \frac{3}{10C_{4/3}{\rm d}y/{\rm d}t}\left(\frac{T_0}{T_C}\right)^{4/3}, \tag{28}$$

where $p_{\rm s} = 2\sigma_0$ is defined. According to the distinct feature of (24), the slope ${\rm d}y/{\rm d}t$ is almost independent of magnets.

Independently of the above argument, so-called Rhodes-Wohlfarth plot, i.e. $p_{\rm C}/p_{\rm s}$ vs $T_{\rm C}$ plot, was already proposed [18]. The value of $p_{\rm C}$ is defined by the relation $p_{\rm C}(p_{\rm C}+2) = p_{\rm eff}^2$. We show in Figure 4, experimentally observed ratios of magnetic moments for compounds Y_xNi_y, plotted in two different ways against $T_{\rm C}$ or $T_{\rm C}/T_0$. As shown in the right figure of Figure 4, almost all

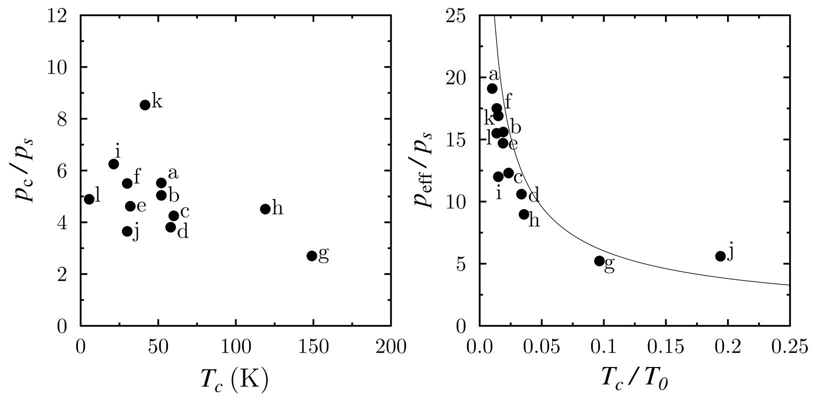


Figure 4. Rhodes-Wohlfarth plot (left) and its revised one (right) for itinerant electron ferromagnets, Y_xNi_y by Nakabayashi *et al* [19]. Solid circles of i, j, k, and l are for $ZrZn_2$, MnSi, Ni_3Al, and Sc_3In, respectively.

the ratios fall near the solid theoretical curve, confirming the validity of (28). Recently, another $\log(p_{\rm eff}/p_{\rm s})$ vs $\log(T_C/T_0)$ plot has been also proposed as shown in Figure 5.

4.2.2. Spontaneous magnetic moment In the ordered phase below T_c, the σ dependences of $y(\sigma,t)$ and $y_z(\sigma,t)$ are given by

$$y(\sigma,t) \simeq y_1(t)[\sigma^2 - \sigma_0^2(t)], \quad y_z(\sigma,t) \simeq 2y_1(t)\sigma_0^2(t) + 3y(\sigma,t), \tag{29}$$

where $\sigma_0^2(t)$ and $y_1(t)$ are the spontaneous magnetic moment squared in the absence of external magnetic field and the fourth expansion coefficient of the free energy. These parameters are determined by substituting $y(\sigma,t)$ and $y_z(\sigma,t)$ in the above (29) and σ^2 given by

$$\sigma^2 \simeq \sigma_0^2(t) + \frac{1}{y_1(t)}y(t), \tag{30}$$

into our basic equation (8). Then the following simultaneous equations for two independent parameters, $U(t) = \sigma_0^2(t)/\sigma_0^2(0)$ and $V(t) = y_z(\sigma_0(t),t)/y_z(\sigma_0(0),0)$, are obtained as the zeroth

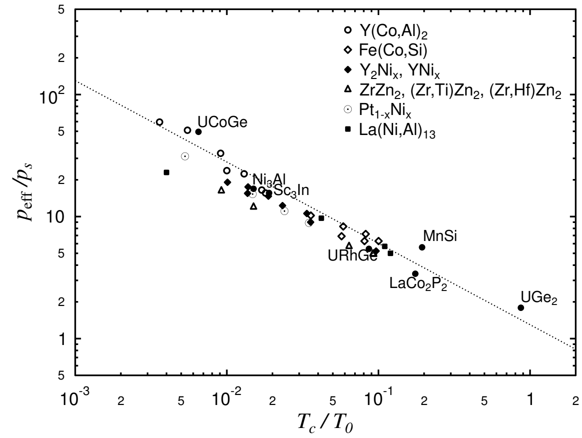


Figure 5. Deguchi-Takahashi log-log plot for various itinerant electron ferromagnets [6]. The dotted line corresponds to the relation, $p_{\text{eff}}/p_{\text{s}} \simeq 1.4(T_0/T_{\text{C}})^{2/3}$, estimated theoretically.

and the first expansion coefficient with respect to y.

$$\begin{aligned} U(t) - \frac{2}{5}V(t) + \frac{1}{5A(0,t_c)}[2A(0,t) + A(y_{z0},t)] = 0, \\ V(t)\left\{1 - \frac{1}{5c}[2A'(0,t) + 3A'(y_{z0},t)]\right\} = 0. \end{aligned} \tag{31}$$

Although it may seem to be a trivial problem, the y derivative of the perpendicular component of the thermal amplitude $A'(y,t)$ has to be well-defined at $y = 0$ [5] in the second equation of the above (31). The simple $\sqrt{y}$ dependence of $A(y,t)$ will lead to the divergent $A'(0,t)$. To find the t-dependence of $U(t)$ and $V(t)$ below T_C, we have to rely on some numerical calculations. As will be shown later, numerically calculated these parameters vanish simultaneously at the critical point.

Experimentally, the T^2-linear dependence of $\sigma_0^2(t)$ is observed for most of weak itinerant electron ferromagnets. On the other hand, almost no interest have been payed on the same dependence of $y_1(t)$, except for the analysis of Wohlfarth and de Chatel [20] in Figure 6, as well as those by Beille *et al* [21] for Ni-Pt alloys in Figure 7. If we confine our interest within these temperature ranges, analytical treatments become possible as will be shown below.

(i) In the limit of low temperature, reduced fourth expansion coefficient, $y_1(t)/y_1(0)$, and the spontaneous moment squared, $U(t) = \sigma_0^2(t)/\sigma_0^2(0)$, are given as follows.

$$\begin{aligned} \frac{y_1(t)}{y_1(0)} = \frac{V(t)}{U(t)} = 1 - \frac{c[2(\pi/2)^4 + 3]}{480A^2(0,t_c)}\left(\frac{T}{T_0}\right)^2 + \cdots \simeq 1 - \frac{b_0}{p_s^4}\left(\frac{T}{T_A}\right)^2, \\ \frac{\sigma_0^2(t)}{\sigma_0^2(0)} = U(t) = 1 - \frac{c[(\pi/2)^4 + 5(\pi/2)^2 + 4]}{360A^2(0,t_c)}\left(\frac{T}{T_0}\right)^2 + \cdots \simeq 1 - \frac{a_0}{p_s^4}\left(\frac{T}{T_A}\right)^2. \end{aligned} \tag{32}$$

where $b_0 \simeq 56.91$ and $a_0 \simeq 112.1$. These relations provide us the alternative ways to estimate the parameter T_A experimentally. For example, from the T^2-linear coefficient α_0 of $U(t)$ in (33) below, the value of T_A is estimated as follows.

$$U(t) \simeq 1 - \frac{a_0}{p_{\text{s}}^4}\frac{T^2}{T_A} = 1 - \alpha_0 T^2, \quad T_A = \frac{1}{p_{\text{s}}^2}\sqrt{\frac{a_0}{\alpha_0}} \tag{33}$$

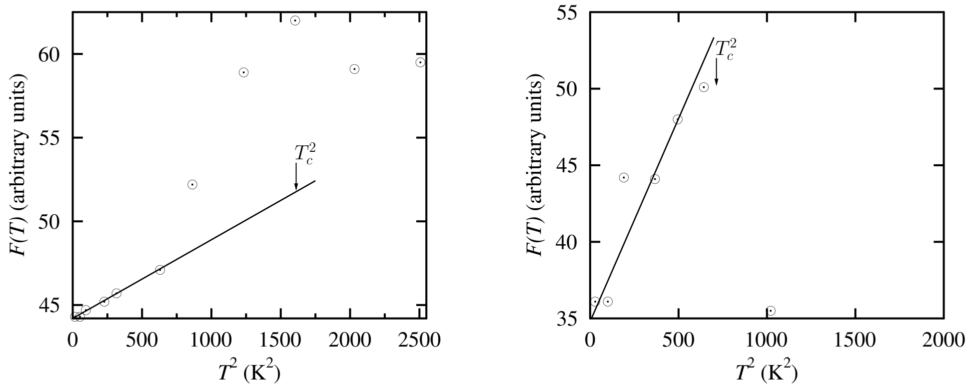


Figure 6. T^2-linear dependence of $F(T) \propto 1/y_1(t)$ for $Zr_{0.92}Ti_{0.08}Zn_2$ (left) and $ZrZn_{19}$ (right) by Wohlfarth and de Chatel (1970).

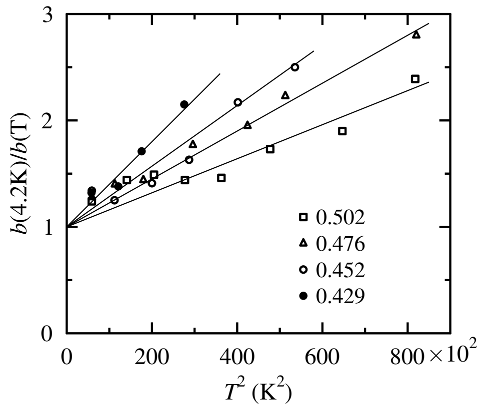


Figure 7. T^2-linear dependence of $y_1(t)$ for Ni-Pt alloys by Beille, Bloch, and Besnus (1974)

Experimentally estimated values of T_A are shown in the third column of Table 3 for several itinerant ferromagnets. They are compared well with those in the fourth column estimated from the magnetization curves in the ground state.

Table 3. Comparison of the values of T_A and $T_A^{(g)}$ estimated by two different experiments.

Compounds	α_0 (K^{-2})	T_A (K)	$T_A^{(g)}$ (K)	Ref.
$Ni_{74.7}Al_{25.3}$	2.77×10^{-3}	6.00×10^4	3.85×10^4	[22]
Ni_3Al	0.784×10^{-3}	4.51×10^4	3.09×10^4	[23]
$ZrZn_2$	2.69×10^{-3}	9.51×10^3	7.40×10^3	[24]
Y_2Ni_{15}	8.54×10^{-5}	3.41×10^4	3.51×10^4	[25]
YNi_3	1.20×10^{-3}	1.28×10^5	0.92×10^5	[26]
$Ni_{0.45}Pt_{0.55}$		1.0×10^4	0.69×10^4	[27]

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(ii) Around the critical temperature below T_C, the following temperature dependence of $U(t)$ and $V(t)/U(t)$ are derived from (31).

$$\begin{aligned} U(t) &= \frac{\sigma_0^2(t)}{\sigma_0^2(0)} \simeq a_c[1-(T/T_C)^{4/3}], \\ \frac{V(t)}{U(t)} &= \frac{y_1(t)}{y_1(0)} \simeq b_c[1-(T/T_C)^{4/3}], \end{aligned} \tag{34}$$

where $a_c = 7/5$ and $b_c \simeq \dfrac{640cC_{4/3}}{21\pi^2 t_c^{2/3}} \gg 1$ for $t_c \ll 1$.

Finally, we show in Figure 8, numerically calculated results of the temperature dependence of $\sigma_0^2(t)/\sigma_0^2(0)$ and $y_1(t)/y_1(0)$ below T_C. The steep decrease of the dashed curve of

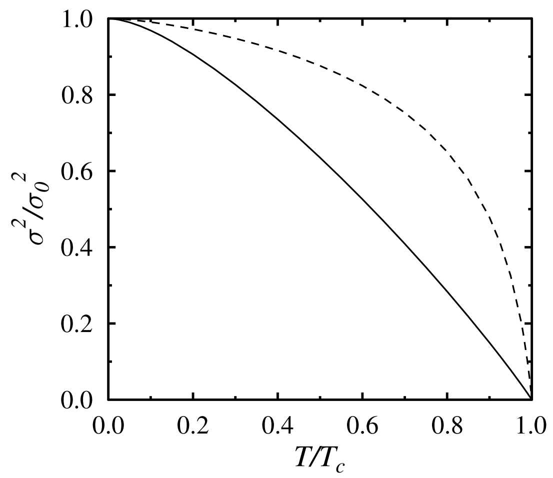

Figure 8. Numerically evaluated spontaneous magnetic moment σ^2/σ_0^2 (solid line) and the fourth expansion coefficient $y_1(t)/y_1(0)$ of the free energy (dashed line).

$y_1(t)/y_1(0)$ around the critical point results from the $1/t_c^{2/3}$ dependence of the coefficient b_c in (34).

5. Summary

Since the successful explanation of the Curie-Weiss law dependence of the magnetic susceptibility, several misleading statements have been made in the SCR theory. They are closely related to its basic concepts. Its strict condition on the magnetization curve has actually caused the problems around the critical point. The purpose of this review is to clarify the origin of these problems. On the other hand, we show how many interesting magnetic properties are derived from another theory of spin fluctuations based on the quite different basic ideas of TAC and GC.

To conclude, magnetic properties of itinerant electron ferromagnets should be understood according to the following basis.

- Spin amplitude conservation plays predominant rolls in our understanding of itinerant electron magnets.
  It implicitly implies the significant role of zero-point spin fluctuations.
- The linearity of the magnetization curve between H/M and M^2 is not generally satisfied.
  At the critical temperature, for instance, M^4 vs H/M relation is rather satisfied.

- Temperature and external field dependence of itinerant electron magnets are not so simple. They will change simultaneously with temperature or under the influence of the external magnetic field, contrary to the expectation of early studies.

Finally, as applications of the spin fluctuation theory presented in this review, the effects of spin fluctuations on the specific heat and the magneto-volume effects have also been treated [6, 28, 29]. Both dependences on temperature and external magnetic field are treated in consistent with thermodynamic relations.

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