Beyond Gaussian approximation in the spin-fluctuation theory of metallic ferromagnetism

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Abstract. A characteristic feature of the Gaussian spin-fluctuation theory is the jump transition into the paramagnetic state. We eliminate the jump and obtain a continuous second-order phase transition by taking into account the high-order terms of the free energy of electrons in the fluctuating exchange field. The third-order term of the free energy yields a renormalization of the mean field, and fourth-order term, responsible for the interaction of the fluctuations, gives a renormalization of the spin susceptibility. The extended theory is applied to the calculation of magnetic properties of Fe-Ni Invar.

1. Introduction
Conventional spin-fluctuation theory (SFT) uses a self-consistent quadratic approximation to the free energy of electrons in a fluctuating exchange field (see, e.g., [1] and references therein). However, calculations showed that for strong ferromagnets the quadratic approximation is insufficient to give satisfactory temperature dependence of magnetic characteristics. Static SFT gives the magnetization that is not in good agreement with the experiment at low temperatures, gives a small effective moment in the Curie-Weiss law, etc. In dynamic SFT the agreement with experiment at finite temperatures is better but a sharp increase of the fluctuations at high temperatures leads to a discontinuous change in magnetic characteristics [2].

In the present paper, using the Hubbard model with five degenerate bands and the first-principle density of states, we develop an extension of the SFT that takes into account high-order terms of the free energy. First, in the fourth-order Taylor expansion of the free energy \( F(V) \), we take a partial average with respect to \( \Delta V \) in the third- and fourth-order terms replacing them by a linear and quadratic term, respectively. Adding the obtained corrections to the Taylor terms of the first- and second-order, we come to the ‘extended’ function \( F(V) \). The best quadratic (Gaussian) approximation is constructed as in [3] with the help of the free energy minimum principle [4], but using the first and second order derivatives of the extended function \( F(V) \). This yields a Gaussian approximation different from the one obtained in [3], namely: the third-order term renormalizes the value of the magnetization, and the fourth-order term renormalizes the spin susceptibility. The fundamental difference between our treatment of the high-order terms and the previous ones is that our approach changes the ferromagnetic state selfconsistently (for treatments of the fourth-order term in the paramagnetic state see, e.g., [5] and references therein).

The extended SFT is applied to the calculation of magnetic properties of \( \text{Fe}_{0.65}\text{Ni}_{0.35} \) Invar.
2. Quadratic approximation taking account of the high-order terms

The free energy of the electrons in the exchange field \( \tilde{V} = (V_1, V_2, ...), V_j = \sum_\alpha V_j^\alpha(\tau)\sigma^\alpha \), fluctuating in space and in ‘time’ \( \tau \in [0, 1/T] \) (\( j \) is the site number, \( \sigma^\alpha \) is the Pauli matrix, \( \alpha = x, y, z \)) is given by the formula

\[
F(\tilde{V}) = T \frac{V^2}{2UT} + \ln G(\tilde{V}). \tag{1}
\]

Here the terms independent of \( V \) are omitted, \( T \) is the temperature (in energy units), \( T \)r is the full quantum-mechanical trace, \( U \) is the single-site electron-electron interaction constant, and

\[
G(\tilde{V}) = (z + \mu - H_0 - \tilde{V})^{-1}
\]

is the Green function of non-interacting electrons in the field \( \tilde{V} \), where \( z \) is the energy variable, \( \mu \) is the chemical potential, and \( H_0 \) is the single-electron Hamiltonian.

We expand the function (1) in Taylor series to the fourth order in \( \Delta V = V - \bar{V} \):

\[
F(\tilde{V}) \approx F(\bar{V}) + T \left( \frac{\bar{V} \Delta V}{UT} + G(\bar{V}) \Delta V \right) + \frac{1}{2} T \left( \frac{\Delta V^2}{UT} + (G(\bar{V}) \Delta V)^2 \right) + \frac{1}{3} T \left( G(\bar{V}) \Delta V \right)^3 + \frac{1}{4} T \left( G(\bar{V}) \Delta V \right)^4. \tag{2}
\]

Taking partial averages over \( \Delta V \), we transform the third- and fourth-order terms in the expansion (2) into corrections to the first- and second-order terms, respectively. Then, constructing the best quadratic approximation along the lines of [4], we average the obtained expression for \( F(\tilde{V}) \) over \( \bar{V} \) (everywhere but in \( \Delta V \)), and keep only the second-order term:

\[
F(\tilde{V}) \approx F(\bar{V}) = \frac{1}{2} T \left( \frac{\Delta V^2}{UT} + \langle G(\bar{V}) \Delta VG(\bar{V}) \Delta V \rangle \right) + 3 \langle G(\bar{V}) \Delta V G(\bar{V}) \Delta V G(\bar{V}) \Delta V \rangle. \tag{3}
\]

Here the pair coupling of \( \Delta V = V - \bar{V} \) implies the Gaussian average, where the mean field \( \bar{V} = \langle \bar{V} \rangle \) is derived from the condition that the linear term annihilates:

\[
T \left( \frac{\bar{V} \Delta V}{UT} + \langle G(\bar{V}) \rangle \Delta V + \langle G(\bar{V}) \Delta VG(\bar{V}) \Delta V \rangle \right) = 0, \tag{4}
\]

identically over \( \Delta V \). (For details see our paper [6].)

3. Basic equations of the extended SFT

Practical use of the quadratic approximation (3) and (4) requires further simplifications. First, we replace the averages of the products of the Green functions \( \bar{G}(\bar{V}) \) over \( \bar{V} \) by the products of averages \( \bar{G} \equiv \langle \bar{G}(\bar{V}) \rangle \) and apply the approximate formula

\[
\text{Tr}(\bar{G} \Delta V \bar{G} \Delta V) \approx \frac{1}{2N_a N} \text{Tr}(\bar{G} \Delta V \bar{G} \Delta V) \text{Tr}(\bar{G} \Delta V) \equiv \eta \text{Tr}(\bar{G} \Delta V). \tag{5}
\]

We neglect the charge field, same as in [3].
where \( N_n \) is the number of atoms, and \( N = 5 \) is the number of (degenerate) d bands. Then following [3], we replace the Green function by its site-diagonal part: \( \bar{G}_{jj'} \approx g(\omega_n) \delta_{jj'} \) (single-site approximation) to rewrite (4) as follows:

\[
\bar{V}_z = -u(1 + \eta) s_z,
\]

Here \( \bar{V}_z = N^{-1}_a \bar{V}_{q=0} (\omega_n = 0) \) is the value of the mean field, \( u = U/N \), and

\[
s_z = N \text{Int}(g_\uparrow - g_\downarrow)/2\tag{7}
\]

is the mean spin moment per atom, where the sum over the thermodynamic frequencies \( \omega_n = (2n + 1)\pi T \) is evaluated using the integral over the energy variable \( \varepsilon \) with the Fermi function \( f(\varepsilon) = [\exp((\varepsilon - \mu)/T) + 1]^{-1}:\n\]

\[
T \sum_n ... = \frac{1}{\pi} \int \text{Im} ... f(\varepsilon) \, d\varepsilon \equiv \text{Int} ...
\]

Analogously transforming the quadratic form (3), we come to the expression

\[
F^{(2)}(V) = \frac{1}{2} \text{Tr} \left[ U^{-1} \Delta V^2 + (1 + 3\eta) T (\bar{G} \Delta V)^2 \right] = \sum_{qmn} \Delta V_{qm}^\alpha \left( u - \chi_{qm}^\alpha \right) \Delta V_{q-m}^\alpha .
\]

Here

\[
\chi_{qm}^\alpha = -\frac{N}{2} (1 + 3\eta) T \sum_{kn} \sum_{\gamma \delta} \bar{G}_{kn}^\gamma \bar{G}_{k-q,n-m}^\delta \text{Sp}(\sigma^\alpha \sigma^\gamma \sigma^\alpha \sigma^\delta), \quad \gamma, \delta = 0, z,
\]

is the spin susceptibility of non-interacting electrons, where the wavevector \( k \) runs over the Brillouin zone, \( \omega_n = 2m\pi T \), and \( \text{Sp} \) is the trace over spin indices. Thus \( \Delta V_{qm}^\alpha \) are independent Gaussian variables, and the mean squares of the fluctuations (fluctuations’, for short) are given by the formula

\[
\zeta^\alpha = \frac{1}{N_a} \sum_{qmn} \langle |\Delta V_{qm}^\alpha |^2 \rangle = \frac{1}{N_a} \sum_{qmn} \frac{T}{2(u - 1 - \chi_{qm}^\alpha)} . \tag{9}
\]

Further summation over \( q \) and \( m \), which accounts for the dynamic nonlocal character of the spin fluctuations, follows [3]. Finally, conservation of the total number of electrons \( N_e \) yields

\[
N \text{Int}(g_\uparrow + g_\downarrow) = N_e/N_a . \tag{10}
\]

Four nonlinear equations (7), (9) and (10) on the chemical potential \( \mu \), mean spin moment \( s_z \), and fluctuations \( \zeta^\uparrow \) (= \( \zeta^\uparrow \)) and \( \zeta^z \) form the system of equations of the extended SFT.

4. Renormalization of the mean field and susceptibility

Final computational formulae of the extended SFT differ from the ones in [3] by the renormalizations of the mean field (6) and the susceptibility (8) that depend on the coefficient \( \eta \) defined in (5) (with \( \eta = 0 \) the system of equations reduces to the one in [3]). Using the single-site and ‘static’ approximations, we come to the following expression [6]:

\[
\eta \approx \frac{1}{\pi T} \left[ 2\zeta^z \int (\text{Re} \, g_\uparrow \text{Im} \, g_\downarrow + \text{Re} \, g_\downarrow \text{Im} \, g_\uparrow) \, d\varepsilon + \zeta^z \int (\text{Re} \, g_\uparrow \text{Im} \, g_\uparrow + \text{Re} \, g_\downarrow \text{Im} \, g_\downarrow) \, d\varepsilon \right] \tag{11}
\]

The impact of the corrections due to the third- and fourth-order terms becomes critical at high temperatures. Therefore, coefficient \( \eta \) can be estimated by the reduced formula

\[
\eta \approx \frac{6\zeta}{\pi T} \int (\text{Re} \, g^0 \text{Im} \, g^0) \, d\varepsilon \equiv \frac{c}{T} \zeta , \tag{12}
\]

where \( \zeta = (2\zeta^z + \zeta^x)/3 \) is the mean fluctuation, and \( g^0 = [g_\uparrow + g_\downarrow]/2 \). In the ferromagnetic region, formula (12) follows from the initial formula (11) in the approximation \( g_\uparrow = g_\downarrow \). In the paramagnetic region, formulae (11) and (12) coincide.
5. Results and discussion
The extended SFT is investigated in the dynamic nonlocal approximation (DNA) [3] by the example of the Invar alloy Fe$_{0.65}$Ni$_{0.35}$. The initial non-magnetic density of states (see figure 2 in [2]) is formed from the two spin-polarized densities obtained from the self-consistent calculation for the disordered Fe$_{0.65}$Ni$_{0.35}$ [7]. The experimental value of the spin magnetic moment per atom $m_{\text{exp}}^\text{spin} = 1.70 \mu_B$ (where $\mu_B$ is the Bohr magneton), used to determine the effective interaction constant $u$, is taken from [8].

Figure 1 presents the results of the calculations of basic magnetic characteristics for the Invar Fe$_{0.65}$Ni$_{0.35}$ obtained using the correction coefficient $\eta$ defined by (12) with $c = -0.015W^{-1}$ ($W = 9.70$ eV is the bandwidth). In this case it is possible to obtain nearly full agreement with experiment for the Curie temperature: $T_C = 1.02T_C^\text{exp}$ ($T_C^\text{exp} = 520K$[8]), for the paramagnetic Curie point: $\Theta_C = 1.067T_C^\text{exp}$, for the effective magnetic moment: $m_{\text{eff}} = 0.90m_{\text{eff}}^\text{exp}$ ($m_{\text{eff}}^\text{exp} = 3.3 \mu_B$[9]) and for the local magnetic moment $m_L(T)$ (see the discussion in [10]). As can be seen from figure 1, a sharp increase of the fluctuations and sharp decrease in magnetization at high temperatures, which occurred in [2], disappear in the extended SFT. On the whole the curve for the magnetization fits the experimental one well enough. However, the inflection of the curve $m(T)$ cannot be removed entirely, when $\eta$ is calculated by (12). Only the application of the expression (11) alternating in $T$ in a self-consistent way yields a curve without the inflection [6].

![Figure 1](image-url)  
**Figure 1.** The magnetization $m(T)/m(0)$ (—— calculation, ooooo experiment [8]), the mean square of spin fluctuations $\zeta^x$ (---) and $\zeta^z$ (—-—) in units of $um(0)/(2\mu_B)$, the reciprocal paramagnetic susceptibility $\chi^{-1}(T)$ (—-—) in units of $T_C^\text{exp}/\mu_B^2$, and the local magnetic moment $m_L(T)/m(0)$ (· · · · · ·) of the Invar Fe$_{0.65}$Ni$_{0.35}$ calculated in the DNA of the extended SFT as functions of the reduced temperature $T/T_C^\text{exp}$.

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