Lecture note Magnetism (14)

13th July (2022) Shingo Katsumoto, Institute for Solid State Physics, University of Tokyo

Let us go into **self-consistent renormalization** (SCR) **spin fluctuation** (SF) theory, the entrance of which we finally reached last week. As mentioned in the last lecture, SCR-SF theory approaches the problem with improving the approximation not taking completely different ways. The theory then gained great successes[1]. We have used two weeks to reach the entrance. It is impossible to introduce the details of the first paper[1] in a single lecture. Here I would like to introduce only the framework of the theory along with resumes like [2, 3].e Those who wish to know the details are requested to refer to monographs of this topic[4, 5, 6], and to original papers [*1].

6.5.1 Weak metallic ferromagnetism, paramagnonr

Kawabata, one of the founders of SCR-SP theory, says a strong motivation of the theory is the finding of weak metallic ferromagnetism like $ZrZn_2$, Sc_3In, Ni_3Al[2], Also in the textbook[4] by Moriya, who is synonymous with SCR-SF theory, paramagnon theory by Izuyama and Kubo[7] is introduced as a preliminary form of SCR-SF theory. Let us then consider paramagnons, which are spin waves in non-magnetically-ordered metals with strong effective interactions between spins. Such metals are considered to be marginal to ferromagnetism (anti-ferromagnetism). Palladium (Pd) tends to form ferromagnetic alloys with non-magnetic elements and is considered to be close to ferromagnetism[8]. Helium-3, which is not a metal but has a nuclear spin, can also be mentioned. It is long since the magnon (paramagnon) system of superfluid helium-3 was found to cause BEC[9, 10]. In high-$T_{\rm C}$ cuprates close to anti-ferromagnetic order[11], also in iron-based high-$T_{\rm C}$ superconductors[12], existences of paramagnons have been confirmed and the dispersion relations etc. have been determined.

We saw in the last lecture that in RPA, which is the mean field approximation on dynamic susceptibility, magnon states may be magnetically ordered ground states. This suggests that spin-fluctuations may be considered as excited states in the magnon-system picture even when the ground state is paramagnetic. It also suggests that the existence of such collective excitations may greatly lower the energy of excited states compared with those in static HF approximations. In spite of such signs of improvement, the simple RPA does not solve the problems of mean field theory in temperature dependence of susceptibility etc. This is probably due to ignoring spin fluctuation in finite temperature thermal equilibrium state. Then in the paramagnon theory we consider the spin fluctuation in paramagnetic state.

Hellmann-Feynman theorem

First we prove the following general theorem. We consider a Hamiltonian with parameter p

$$\mathscr{H}(p) = \mathscr{H}_0 + \mathscr{H}_1(p) \tag{6.95}$$

with normalized eigenstate $|p, n\rangle$ and the eigen energy $E_n(p)$. A change in an eigenstate due to an infinitesimally small change δp of p $\mathscr{H}(p+\delta p)$ is expressed as a linear sum of the original eigenstates as

$$|p+\delta p, n\rangle = |p, n\rangle + \sum_m C_m |p, m\rangle . \tag{6.96}$$

[*1] As far as I checked, most of textbooks written in English, do not even refer to the SCR-SF theory, whose logic is even difficult to be followed. They usually introduce the simple HF approximation, then shift to DFT or GGA, in which the approximation is improved. These ab initio calculation may doing similar improvement for the HF as SCR-SF though the details cannot be known.

If we assume linear approximation $C_m = c_m \delta p$, from

$$\langle p+\delta p, n | p+\delta p, n\rangle = |1 + c_n \delta p|^2 \langle p, n|p, n\rangle + \sum_{m\neq n} |c_m|^2 |\delta p|^2 \langle p, m|p, m\rangle \,,$$

the condition of normalization leads to $c_n = 0$, that is, within linear approximation in δp, $C_n = 0$. Namely, within linear approximation of δp,

$$\langle p+\delta p|\mathscr{H}(p)|p+\delta p\rangle = \langle p|\mathscr{H}(p)|p\rangle = E_n(p). \tag{6.97}$$

Then we can write the shift in the eigenenergy as

$$\begin{aligned} E_n(p+\delta p) &= \langle p+\delta p, n|\mathscr{H}(p+\delta p)|p+\delta p, n\rangle \\ &= \left\langle p+\delta p, n \middle| \mathscr{H}(p) + \delta p \frac{\partial \mathscr{H}(p)}{\partial p} \middle| p+\delta p, n \right\rangle \\ &= E_n(p) + \delta p \left\langle p, n \middle| \frac{\partial \mathscr{H}(p)}{\partial p} \middle| p, n \right\rangle. \end{aligned} \tag{6.98}$$

We can thus conclude

$$\frac{dE_n(p)}{dp} = \left\langle p, n \middle| \frac{\partial \mathscr{H}_1(p)}{\partial p} \middle| p, n \right\rangle. \tag{6.99}$$

This is called **Hellmann-Feynman theorem**.

From this theorem, e.g., the free energy $F(p)$ of the system of Hamiltonian $\mathscr{H}(p)$ can be written as

$$\frac{\partial F(p)}{\partial p} = \frac{1}{Z}\sum_n \exp\left[\frac{-E_n(p)}{k_{\rm B}T}\right] \frac{\partial E_n(p)}{\partial p}. \tag{6.100}$$

In eq. (6.95), $\mathscr{H}_1(p)$ can be an interaction Hamiltonian $\mathscr{H}_I$ of interaction constant I. From Hellmann-Feynman theorem, we can introduce the interaction with varying the interaction constant as $I : 0 \to I$. The correction term for free energy is given as a function of I by

$$F(I) = F(0) + \int_0^I \left\langle \frac{\partial \mathscr{H}_{I'}}{\partial I'} \right\rangle dI'. \tag{6.101}$$

From the above, we consider the contribution of spin fluctuation to the specific heat. Here Hubbard model in eq. (6.29) can be written as

$$\mathscr{H} = \sum_{\boldsymbol{k},s} \epsilon_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}s} a_{\boldsymbol{k}s} + U \sum_i^N \hat{n}_{i\uparrow}\hat{n}_{i\downarrow} = \mathscr{H}_0 + \mathscr{H}_I. \tag{6.102}$$

As in the transform of first term, an operator at lattice point $\boldsymbol{R}_i$ is Fourier-expanded as

$$c_{is} = \frac{1}{\sqrt{N}} \sum_{\boldsymbol{k}} e^{i\boldsymbol{R}_i\cdot\boldsymbol{k}} a_{\boldsymbol{k}s}. \tag{6.103}$$

And as in eq. (6.83c), we write $\mathscr{H}_I$ as

$$\mathscr{H}_I = \frac{U}{N} \sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} a^\dagger_{\boldsymbol{k}+\boldsymbol{q}\uparrow} a_{\boldsymbol{k}\uparrow} a^\dagger_{\boldsymbol{k}'-\boldsymbol{q}\downarrow} a_{\boldsymbol{k}'\downarrow}, \tag{6.104}$$

where the interaction parameter is $I = U/N$. We also change the notation in eq. (6.78) a bit to

$$\left.\begin{aligned} S_+(\boldsymbol{q}) &= \sum_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}\uparrow} a_{\boldsymbol{k}+\boldsymbol{q}\downarrow}, \\ S_-(\boldsymbol{q}) &= \sum_{\boldsymbol{k}} a^\dagger_{\boldsymbol{k}\downarrow} a_{\boldsymbol{k}+\boldsymbol{q}\uparrow}. \end{aligned}\right\} \tag{6.78}$$

From fermionic commutation relation, with exchanging the last two operators in the sum in right-hand side, eq. (6.104) is developed to

$$\begin{aligned}\mathscr{H}_I &= I\sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} a^\dagger_{\boldsymbol{k}+\boldsymbol{q}\uparrow}a_{\boldsymbol{k}\uparrow}(\delta_{\boldsymbol{k}',\boldsymbol{k}'-\boldsymbol{q}} - a_{\boldsymbol{k}'\downarrow}a^\dagger_{\boldsymbol{k}'-\boldsymbol{q}\downarrow}) \\ &= I\left[\sum_{\boldsymbol{k},\boldsymbol{k}'} a^\dagger_{\boldsymbol{k}\uparrow}a_{\boldsymbol{k}\uparrow} - \sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} a^\dagger_{\boldsymbol{k}+\boldsymbol{q}\uparrow}a_{\boldsymbol{k}'\downarrow}a^\dagger_{\boldsymbol{k}-\boldsymbol{q}\downarrow}a_{\boldsymbol{k}\uparrow}\right]. \end{aligned} \tag{6.105}$$

Wavenumber $\boldsymbol{q}$, over which the sum is taken, transform $\boldsymbol{q}\to-\boldsymbol{q}+\boldsymbol{k}'-\boldsymbol{k}$ results in $\boldsymbol{k}+\boldsymbol{q}\to\boldsymbol{k}-\boldsymbol{q}+\boldsymbol{k}'-\boldsymbol{k}=\boldsymbol{k}'-\boldsymbol{q}$, and similarly $\boldsymbol{k}'-\boldsymbol{q}\to\boldsymbol{k}+\boldsymbol{q}$. Then from eq. (6.78), we can write

$$\begin{aligned}\mathscr{H}_I &= I\left[\sum_{\boldsymbol{k},\boldsymbol{k}'} a^\dagger_{\boldsymbol{k}\uparrow}a_{\boldsymbol{k}\uparrow} - \sum_{\boldsymbol{k},\boldsymbol{k}',\boldsymbol{q}} a^\dagger_{\boldsymbol{k}'-\boldsymbol{q}\uparrow}a_{\boldsymbol{k}'\downarrow}a^\dagger_{\boldsymbol{k}+\boldsymbol{q}\downarrow}a_{\boldsymbol{k}\uparrow}\right] \\ &= I\left[\sum_{\boldsymbol{k},\boldsymbol{k}'} a^\dagger_{\boldsymbol{k}\uparrow}a_{\boldsymbol{k}\uparrow} - \sum_{\boldsymbol{q}} S_+(-\boldsymbol{q})S_-(\boldsymbol{q})\right] = I\left[\sum_{\boldsymbol{k},\boldsymbol{k}'} a^\dagger_{\boldsymbol{k}\uparrow}a_{\boldsymbol{k}\uparrow} - \sum_{\boldsymbol{q}} S_+(\boldsymbol{q})S_-(-\boldsymbol{q})\right]. \end{aligned} \tag{6.106}$$

Next, in paramagnetic states, $\mathscr{H}_I$ does not change with spin inversion, thus, we add the expression of $\uparrow\leftrightarrow\downarrow$ and divide by two to obtain

$$\mathscr{H}_I = \frac{N_eU}{2} - \frac{I}{2}\sum_{\boldsymbol{q}}\{S_+(\boldsymbol{q}),S_-(-\boldsymbol{q})\}_+, \tag{6.107}$$

where N_e is the number of electrons, $\{A,B\}_+ = AB+BA$ represents anti-commutation relation. In eq. (6.106), sums over $\boldsymbol{k}$, $\boldsymbol{k}'$ produce N_e and N respectively. The latter is because the sum comes from that over $\boldsymbol{R}_i$. Accordingly, the variation in the free energy due to introduction of interaction is given by

$$\varDelta F = \frac{N_eU}{2} - \frac{1}{2}\sum_{\boldsymbol{q}}\int_0^I dI' \left\langle\{S_+(\boldsymbol{q}),S_-(-\boldsymbol{q})\}_+\right\rangle. \tag{6.108}$$

For usage of fluctuation-dissipation theorem, remember eq. (6.74):

$$\mathcal{G}^+_{QP}(\omega) = \sum_{n,m}\langle n|Q|m\rangle\langle m|P|n\rangle\frac{e^{-\beta E_n}-e^{-\beta E_m}}{E_n-E_m+\hbar\omega+i\eta}. \tag{6.74}$$

If we use the lower half of complex plane by replacing $\eta\to-\eta$, we obtain a parallel representation for $\mathcal{G}^-_{QP}(\omega)$. From these two, we can write

$$\mathcal{G}^+_{QP}(\omega) - \mathcal{G}^-_{QP}(\omega) = -2i\mathrm{Im}[\chi_{QP}(\omega)],$$

to further obtain

$$\mathcal{S}_{QP}(\omega) = \frac{2}{1-e^{-\beta\hbar\omega}}\mathrm{Im}[\chi_{QP}(\omega)]. \tag{6.109}$$

We also rewrite eq. (6.80) as

$$\chi_{+-}(\boldsymbol{q},\omega) = -(g\mu_{\mathrm{B}})^2\frac{i}{\hbar}\int_0^\infty dt\,\langle[S_-(-\boldsymbol{q}),S_+(\boldsymbol{q},t)]\rangle\,e^{i\omega t}. \tag{6.110}$$

Let $|n\rangle$ and E_n be a many-body eigenstate and its eigenenergy of Hamiltonian in eq. (6.102), then the imaginary part of $\chi_{+-}(\boldsymbol{q},\omega)$ is given by

$$\mathrm{Im}[\chi_{+-}(\boldsymbol{q},\omega)] = \frac{\pi(g\mu_{\mathrm{B}})^2}{\hbar}\sum_{m,n}(\rho_m-\rho_n)\delta(\omega-\varDelta E_{mn}/\hbar)\langle n|S_-(-\boldsymbol{q})|m\rangle\langle m|S_+(\boldsymbol{q})|n\rangle, \tag{6.111}$$

where

$$\text{Boltzmann factor: } \rho_n = \frac{1}{Z}\exp\left[-\frac{E_n}{k_{\mathrm{B}}T}\right],\quad \varDelta E_{mn} = E_m - E_n.$$

Multiply both sides of eq. (6.111) by $\coth(\beta\omega\hbar/2)$, and integrate over ω to obtain

$$\begin{aligned}
&\int_{-\infty}^{\infty} d\omega \mathrm{Im}\chi_{+-}(\boldsymbol{q},\omega)\coth\left(\frac{\hbar\omega}{2k_{\mathrm{B}}T}\right) \\
&\quad = \frac{\pi(g\mu_{\mathrm{B}})^2}{\hbar}\sum_{m,n}(\rho_m-\rho_n)\coth\left(\frac{\Delta E_{nm}}{k_{\mathrm{B}}T}\right)\langle n|S_-(-\boldsymbol{q})|m\rangle\langle m|S_+(\boldsymbol{q})|n\rangle \\
&\quad = \frac{\pi(g\mu_{\mathrm{B}})^2}{\hbar}\langle\{S_-(-\boldsymbol{q}),S_+(\boldsymbol{q})\}_+\rangle. \qquad (6.112)
\end{aligned}$$

Then we can write the variation of free energy in eq. (6.108) as

$$\Delta F = \frac{N_e U}{2} - \sum_{\boldsymbol{q}}\int_0^I dI' \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega \coth\left(\frac{\hbar\omega}{2k_{\mathrm{B}}T}\right)\mathrm{Im}[\chi_{+-}(\boldsymbol{q},\omega)]. \qquad (6.113)$$

By applying the RPA formula in eq. (6.87), the above is approximated as

$$\Delta F = \frac{N_e U}{2} + \sum_{\boldsymbol{q}}\frac{1}{\pi}\int_0^{\infty} d\omega \coth\left(\frac{\hbar\omega}{k_{\mathrm{B}}T}\right)\mathrm{Im}\{\log[1-2U\chi^{(0)}(\boldsymbol{q},\omega)]\}. \qquad (6.114)$$

For calculation of specific heat, only temperature-dependent parts in ΔF are necessary.

$$\coth\frac{\hbar\omega}{k_{\mathrm{B}}T} = 1 + \frac{2}{e^{\hbar\omega/k_{\mathrm{B}}T}-1}. \qquad (6.115)$$

In the above, the first term "1" in the right hand side is the zero-point motion of paramagnons and ignored in the calculation of specific heat. In eq. (6.114), we need to consider the contribution of $\chi^{(0)}(q,\omega)$, susceptibility of non-interacting system. In (q,ω)-plane shown in Fig. 6.4, the largest contribution is given in region-III, in which we can expand $\chi^{(0)}(q,\omega)$ as

$$\chi^{(0)}(q,\omega) = \frac{1}{2}\rho(\epsilon_{\mathrm{F}})\left[1 - A_0\left(\frac{q}{k_{\mathrm{F}}}\right)^2 + iC_0\frac{\hbar\omega}{\epsilon_{\mathrm{F}}}\frac{k_{\mathrm{F}}}{q}\right]. \qquad (6.116)$$

Here the expansion coefficients are

$$A_0 = \frac{1}{12}, \quad C_0 = \frac{\pi}{4}. \qquad (6.117)$$

For the simplest approximation we use this expression of $\chi^{(0)}(q,\omega)$. Let $\alpha \equiv U\rho(\epsilon_{\mathrm{F}})$ be the interaction constant, we reach

$$\begin{aligned}
\Delta F(T) &= \frac{N}{2}\rho(\epsilon_{\mathrm{F}})\epsilon_{\mathrm{F}}^2\int_0^{q_{\mathrm{c}}} q^2dq\int_0^{\infty} d\omega\frac{2}{e^{\beta\omega}-1}\mathrm{Im}\left[\log\left(1-\alpha+\alpha A_0 q^2 - i\alpha C_0\frac{\omega}{q}\right)\right] \\
&= -\frac{N}{2}\rho(\epsilon_{\mathrm{F}})\epsilon_{\mathrm{F}}^2\int_0^{q_{\mathrm{c}}} q^2dq\int_0^{\infty} d\omega\frac{2}{e^{\beta\omega}-1}\arctan\left[\frac{\omega}{q}\frac{C_0}{K_0^2+A_0q^2}\right]. \qquad (6.118)
\end{aligned}$$

As before, for simplicity of representation, unit of wavenumber q is taken as k_{F}, $\hbar = 1$, unit of energy ω is taken as ϵ_{F}. q_{c} is a cutoff of wavenumber. Here expansion in region-III is effective only for finite q and the cutoff is indispensable and is around 1 (k_{F}). And K_0 is

$$K_0^2 = \frac{1-\alpha}{\alpha}. \qquad (6.119)$$

We further apply low temperature approximation $\omega \ll 1$ and $\arctan x \sim x$ to obtain

$$\frac{\Delta F(T)}{N} = -\frac{2\pi^2}{3}\rho(\epsilon_{\mathrm{F}})(k_{\mathrm{B}}T)^2\frac{C_0}{2\pi A_0}\log\frac{K_0^2+A_0q_{\mathrm{c}}^2}{K_0^2}, \qquad (6.120)$$

from which we know the spin-fluctuation contribution to low temperature specific heat is proportional to T. Then if we write

$$C = \gamma T, \quad \gamma_0 \equiv \frac{2\pi^2}{3}k_{\mathrm{B}}^2\rho(\epsilon_{\mathrm{F}}), \qquad (6.121)$$

then the coefficient γ is

$$\gamma = \gamma_0 \left(1 + \frac{C_0}{\pi A_0} \log \frac{K_0^2 + A_0 q_c^2}{K_0^2}\right), \tag{6.122}$$

where γ_0 is the temperature coefficient of specific heat of electron systems without contribution from spin fluctuation. Equation (6.122) tells that the temperature coefficient of specific heat logarithmically diverges as the condition approaches Stoner ferromagnetic criterion $\alpha \to 1$, $K_0 \to 0$.

6.5.2 SCR spin-fluctuation theory

So far, Hellmann-Feynman theorem and fluctuation-dissipation theorem are used to obtain the expression for free energy of thermal equilibrium with dynamic susceptibility. By applying RPA to dynamic susceptibility, the effect of spin-fluctuation is included. However, at this stage the theory just describes low-temperature approximation of paramagnetic states. A problem here is, as is always the case of many-body problem, the effect of spin-fluctuation should be reflected on the spin-fluctuation. Hence this simply cannot be applied to ferromagnetic state with spontaneous magnetization. For that we need to consider self-consistent equation as in the case of Heisenberg model.

Then, here, we consider the free energy in the presence of magnetization M. The free energy is given as the sum of free energy $F_0(M,T)$ of non-interacting system and the quantity in eq. (6.113) by

$$F(M,T) = F_0(M,T) + \frac{N_e U}{2} - bM - \sum_{\boldsymbol{q}} \int_0^I dI' \frac{1}{2\pi} \int d\omega \coth \frac{\hbar\omega}{2k_B T} \mathrm{Im}[\chi_{+-}(M, I'; \boldsymbol{q}, \omega)]. \tag{6.123}$$

$-bM$ is the Zeeman term. Here, as a notation, the magnetization M and the interaction parameter I are specified in the dynamic susceptibility χ_{+-}.

As we saw in the GL theory static spontaneous magnetization M is given by extremum condition of F as

$$\frac{\partial F(M,T)}{\partial M} = 0. \tag{6.124}$$

This can be viewed as **magnetic equation of state**.

We restart with eq. (6.113), which does not contain any approximation. In HF mean-field approximation, integrand of the second term in right-hand side is replaced with the one without interaction ($I = 0$), thus the integration with dI' over region $[0, I]$ is replaced with simple product of I.

$$\Delta F_{\rm HF} = \frac{N_e U}{2} - I \sum_{\boldsymbol{q}} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \coth\left(\frac{\hbar\omega}{2k_B T}\right) \mathrm{Im}[\chi_{+-}(M, 0; \boldsymbol{q}, \omega)]. \tag{6.125}$$

From the Hubbard model in eq. (6.102), we consider the quantity:

$$\begin{aligned}\left\langle \frac{\partial \mathscr{H}}{\partial I} \right\rangle_{I=0} &= N \sum_i^N \langle n_{i\uparrow} n_{i\downarrow} \rangle_{I=0} = N \sum_i^N \langle n_{i\uparrow} \rangle \langle n_{i\downarrow} \rangle \\ &= \frac{N^2}{4}(n_+^2 - n_-^2) = \frac{N^2}{4}[n^2 - (2m)^2] = \frac{N_e^2}{4} - M^2. \end{aligned} \tag{6.126}$$

In the above, small characters n, m are the numbers per sites, and

$$n_+ = n_\uparrow + n_\downarrow, \quad n_- = n_\uparrow - n_\downarrow, \quad m = \frac{n_-}{2},$$

where the unit of magnetization is taken as $g\mu_B$, the spin is 1/2. We take eq. (6.126) multiplied by I as the I-proportional term in (6.125). Then we can write formally

$$\begin{aligned} F(M,T) = &F_0(M,T) + I\left(\frac{N_e^2}{4} - M^2\right) - bM && \text{: HF Approximation} \\ &- \sum_{\boldsymbol{q}} \int_0^I dI' \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \coth \frac{\hbar\omega}{k_B T} \mathrm{Im}[\chi_{+-}(M, I'; \boldsymbol{q}, \omega) - \chi_{+-}(M, 0; \boldsymbol{q}, \omega)] && \text{: Correction.} \end{aligned} \tag{6.127}$$

As is in paramagnon theory, we apply RPA(eq. (6.87)) to $\chi_{+-}(M, I'; \boldsymbol{q}, \omega)$. Then the integration with I' can be performed and the result is

$$F(M,T) = F_0(M,T) + I\left(\frac{N_e^2}{4} - M^2\right) - bM$$
$$- \sum_{\boldsymbol{q}} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \coth\frac{\hbar\omega}{k_{\rm B}T} {\rm Im}[\log\{1 - 2U\chi^{(0)}(M;\boldsymbol{q},\omega)\} + 2U\chi^{(0)}(M;\boldsymbol{q},\omega)], \quad (6.128)$$

where we write

$$\chi^{(0)}(M;\boldsymbol{q},\omega) = \frac{1}{2N}\chi_{+-}(M,0;\boldsymbol{q},\omega). \quad (6.129)$$

We obtain the following magnetic equation of state by calculating magnetization-derivative of eq. (6.124) in eq. (6.128).

$$\frac{\partial F_0}{N\partial m} - 2Um - b - \frac{1}{N}\sum_{\boldsymbol{q}} \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega \coth\frac{\hbar\omega}{2k_{\rm B}T} {\rm Im}\left[\frac{2U\chi^{(0)}(M;\boldsymbol{q},\omega)}{1-2U\chi^{(0)}(M;\boldsymbol{q},\omega)} 2U\frac{\partial\chi^{(0)}(M;\boldsymbol{q},\omega)}{\partial m}\right] = 0, \quad (6.130)$$

which is given as an equation for magnetization per site $m = M/N$.

Here we formally define magnetization per site in paramagnetic states as

$$\chi = \frac{\partial m}{\partial b}, \quad \frac{1}{\chi} = \frac{\partial b}{\partial m}. \quad (6.131)$$

We also write susceptibility per site for non-interacting system as χ_0. Free energy of the system with Zeeman term $F_0 - bm$ gives a magnetic equation of state as

$$\frac{\partial F_0}{N\partial m} - b = 0. \quad (6.132)$$

By applying eq. (6.124) to the above with transposition of b, we obtain

$$\frac{\partial^2 F_0}{N\partial m^2} = \frac{1}{\chi_0}. \quad (6.133)$$

Then in eq. (6.130) for the case of paramagnetic state, substituting the above gives

$$\frac{1}{\chi} = \frac{1}{\chi_0} - 2U$$
$$- \frac{1}{N}\sum_{\boldsymbol{q}}\frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega \coth\frac{\hbar\omega}{2k_{\rm B}T}(2U)^2$$
$$\times\,{\rm Im}\left[\chi(\boldsymbol{q},\omega)\left.\frac{\partial^2\chi^{(0)}(\boldsymbol{q},\omega)}{\partial m^2}\right|_{m=0} + \chi^2(\boldsymbol{q},\omega)\left\{\frac{1}{\chi^{(0)}}(\boldsymbol{q},\omega)\left.\frac{\partial\chi^{(0)}(\boldsymbol{q},\omega)}{\partial m}\right|_{m=0}\right\}^2\right], \quad (6.134)$$

the last part of which is calculated as follows. Equation (6.87) of RPA and eq. (6.129) lead to

$$\chi_(\boldsymbol{q},\omega) = \frac{\chi^{(0)}(\boldsymbol{q},\omega)}{1-2U\chi^{(0)}(\boldsymbol{q},\omega)}. \quad (6.135)$$

We then apply this to obtain

$$\frac{\partial\chi}{\partial m} = \frac{\partial\chi}{\partial\chi^{(0)}}\frac{\partial\chi^{(0)}}{\partial m} = \frac{1}{(1-2U\chi^{(0)})^2}\frac{\partial\chi^{(0)}}{\partial m} = \chi^2\frac{1}{\chi^{(0)2}}\frac{\partial\chi^{(0)}}{\partial m}.$$

Equation (6.134) tells the existence of correction, which is expressed as an integral with ω, to the susceptibility from the term indicated as “Correction” in eq. (6.127). In the correction, inside ${\rm Im}[\cdots]$, within two terms in $\cdots$, the first term is linear in spin-fluctuation, the second is in the second order. Here we only consider the linear term. We further approximate the first term by replacing $\partial^2\chi^{(0)}/\partial m^2|_{m=0}$ with the value for $q = 0$, $\omega = 0$.h Then by writing that

$$g = -(2U)^2\chi_0 \left.\frac{\partial^2\chi^{(0)}(\boldsymbol{q},\omega)}{\partial m^2}\right|_{m=0,q=0,\omega=0}, \quad (6.136)$$

we obtain the following.

$$\frac{\chi_0}{\chi} = 1 - 2U\chi_0 + \frac{g}{N}\sum_{\boldsymbol{q}} \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega \coth\frac{\hbar\omega}{2k_{\mathrm{B}}T}\mathrm{Im}[\chi(\boldsymbol{q},\omega)]. \tag{6.137}$$

We further ignore the temperature dependence of susceptibility of non-interacting system χ_0. This should be taken into account, e.g., in comparison with experiments etc., though here our purpose is to see the essential structure of SCR-SF theory. However, we still have the problem of inconsistency in that application of RPA (eq. (6.135)) to the result of (6.137) results in a disagreement of the divergent point of dynamic susceptibility $\omega \neq 0$ and that of static one ($\omega = 0$). In spite of the progress of approximation, a simple RPA still cannot satisfy the self-consistent condition.

The difficulty is avoided in the SCR-SF theory in the following way. We write the susceptibility at absolute zero as

$$\frac{\chi_0}{\chi(T=0)} = 1 - 2U\chi_0 + \frac{g}{N}\sum_{\boldsymbol{q}} \frac{1}{\pi}\int_0^{\infty} d\omega\mathrm{Im}[\chi(\boldsymbol{q},\omega)]_{T=0}. \tag{6.138}$$

The third term in the rhs represents the contribution of spin-fluctuation with $q \neq 0$ at absolute zero, namely the zero-point motion. As can be seen here, the effect of spin-fluctuation enters into the denominator of RPA representation making the condition of ferromagnetism difficult to be satisfied.

We take this as an RPA modified in the simplest way by the effect of spin-fluctuation. As is in the case of eq. (6.115), we again ignore the temperature dependence of zero-point motion in magnon spectrum and take the difference between eq. (6.137) and eq. (6.138). The approximation gives

$$\frac{\chi_0}{\chi} = \frac{\chi_0}{\chi(T=0)} + \frac{g}{N}\sum_{\boldsymbol{q}} \frac{1}{\pi}\int_0^{\infty} d\omega\frac{2}{e^{\hbar\omega\beta}-1}\mathrm{Im}[\chi(\boldsymbol{q},\omega)]. \tag{6.139}$$

Here an expansion of $\chi^{-1}(\boldsymbol{q},\omega)$ around $(q,\omega)=(0,0)$ is possible as in eq. (6.116). The functional form is

$$\frac{\chi_0}{\chi(\boldsymbol{q},\omega)} = \frac{\chi_0}{\chi(+0,+0)} + A\left(\frac{q}{k_{\mathrm{F}}}\right)^2 - iC\frac{\omega}{\epsilon_{\mathrm{F}}}\frac{k_{\mathrm{F}}}{q}. \tag{6.140}$$

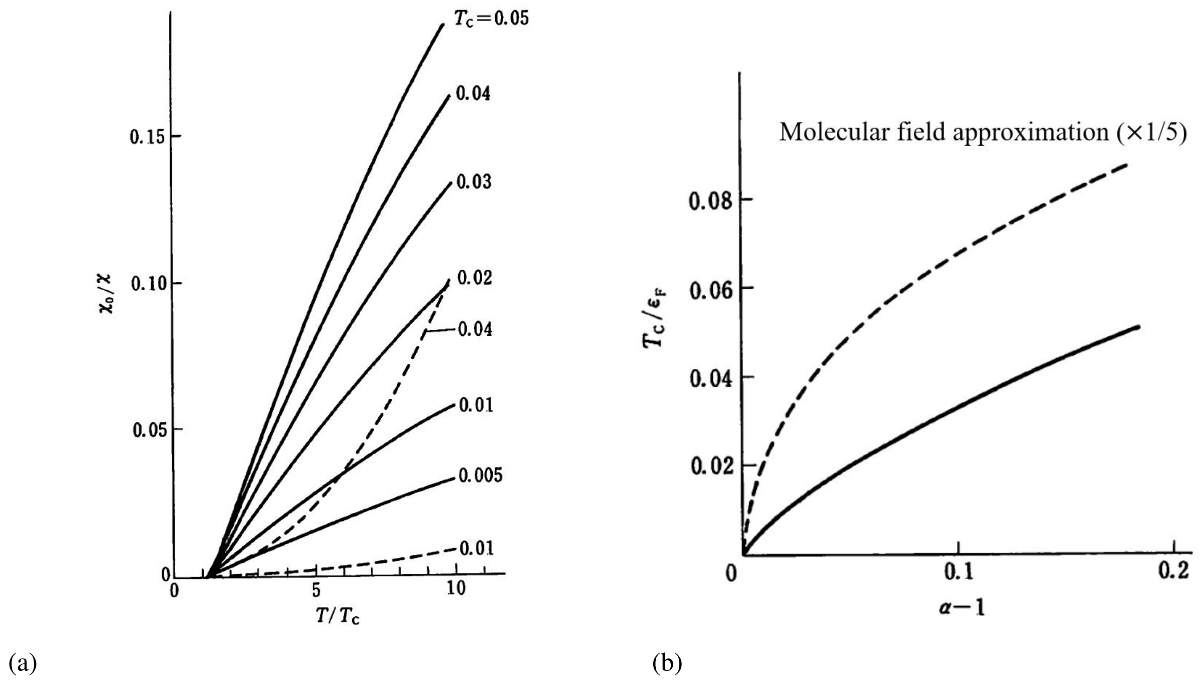
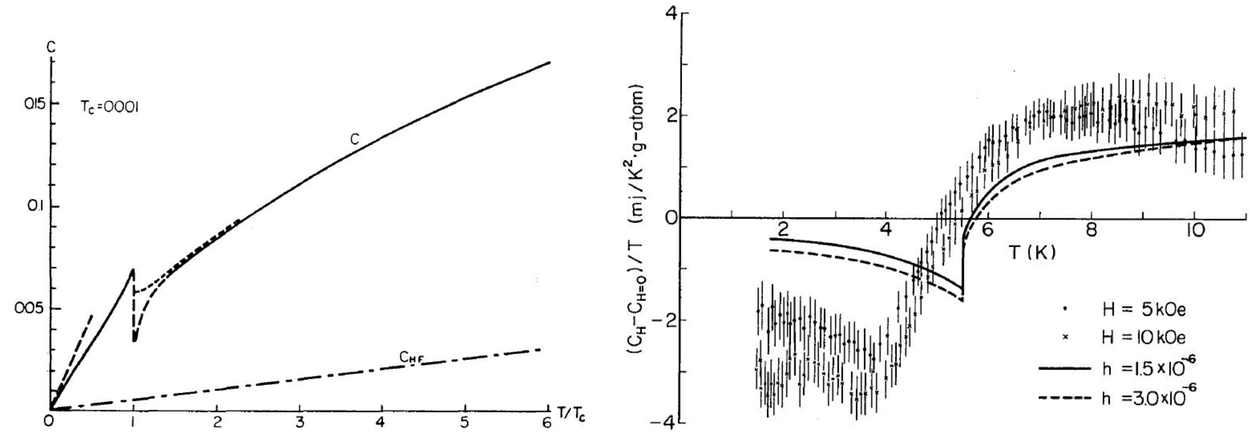


Fig. 6.6 (a) Temperature dependence of susceptibility in a ferromagnet calculated in the SCR-SF theory. The parameter here is the interaction parameter, which determines T_{C}. The broken lines are by the simple molecular field theory. (b) Critical temperature as a function of Stoner parameter $\alpha = I\mathscr{D}(\epsilon_{\mathrm{F}})$ calculated by the SCR-SF theory. The broken line is 1/5 of the result in the simple molecular field theory. From [1].



(a) (b)

Fig. 6.7 (a) Specific heat in the vicinity of ferromagnetic transition point calculated with the SCR-SF theory. From [13]. (b) Specific heat of itinerant electron weak ferromagnet Sc_3In under magnetic field around $T_{\rm C}$. The measured values in zero-magnetic field is subtracted. Solid and broken lines are results of numerical calculation. From [14].

This form requires agreement of $\chi(\boldsymbol{q},\omega)$ with the static susceptibility for $q\to+0$, $\omega\to+0$.

In summary, calculation of $\chi(\boldsymbol{q},\omega)$ from (6.139) and (**??**) results in a self-consistent solution. The above is the essential framework of the SCR-SF theory though realistic calculations are far more complicated even for simplified band structure with many expansion coefficients[1].

In Fig. 6.6, we show the temperature dependence of susceptibility calculated with the SCR-SF theory for a simple parabolic band structure and the critical temperature $T_{\rm C}$ as a function of interaction strength $\alpha = I\mathscr{D}(\epsilon_{\rm F})$ (Stoner parameter) [1]. In Fig. 6.6(a), compared with the simplest molecular field theory (broken lines), the SCR-SF theory gives much better linearity in a wide temperature range, which indicates the Curie-Weiss law. Also as in Fig. 6.6(b), the energy of paramagnetic state largely lowers due to the contribution of spin-fluctuation in thermal equilibrium, leading to correction of overestimation in stability of ferromagnetic state. As a result, $T_{\rm C}$ largely lowers from the results of the molecular field theory.

On the other hand, there still exist many problems and open questions. As shown in Fig. 6.7, there is a large difference between the calculated behavior of specific heat and those in experiments. Studies on these points have long history and they are summarized e.g., in [5].

Appendix 14A: Calculation in (6.111)

The calculation shown in the following, very often appears, e.g, in usage of Green's function (calculation of (6.74) is almost the same). For your convenience, I would like to show a little of calculation in (6.111). It is probably enough to see

$$\frac{i}{\hbar}\int_0^\infty dt\,\langle S_+(\boldsymbol{q},t)S_-(\boldsymbol{q})\rangle\, e^{i\omega t} = (**).$$

This is calculated as follows.

$$\begin{aligned}
(**) &= \frac{i}{\hbar Z}\int_0^\infty dt\mathrm{Tr}\left[e^{-\beta\mathscr{H}}e^{(i/\hbar)\mathscr{H}t}S_+(\boldsymbol{q})e^{-(i/\hbar)\mathscr{H}t}S_-(\boldsymbol{q})\right]e^{i\omega t}\\
&= \frac{i}{\hbar Z}\int_0^\infty dt\sum_{n,m}\langle n|\,e^{-\beta\mathscr{H}+(i/\hbar)\mathscr{H}t}S_+(\boldsymbol{q})\,|m\rangle\,\langle m|\,e^{-(i/\hbar)\mathscr{H}t}S_-(\boldsymbol{q})\,|n\rangle\,e^{i\omega t}\\
&= \frac{i}{\hbar Z}\int_0^\infty dt\sum_{n,m}e^{(-\beta+(i/\hbar)t)E_n}\,\langle n|\,S_+(\boldsymbol{q})\,|m\rangle\,\langle m|\,S_-(-\boldsymbol{q})\,|n\rangle\,e^{-(i/\hbar)E_m t}e^{i\omega t}.
\end{aligned}\tag{14A.1}$$

Here we add a infinitesimal real part to the pure imaginary argument of exponential function, which is a trick often appears in Fourier integration over half-infinite region. Rigorous proof is of course possible with ϵ-δ logics. Below, we omit to write but η implicitly means taking the limit $\eta \to +0$.

$$\begin{aligned}
(**) &= \sum_{n,m} \langle n| S_+(\boldsymbol{q}) |m\rangle \langle m| S_-(-\boldsymbol{q}) |n\rangle \frac{ie^{-\beta E_n}}{\hbar Z} \int_0^\infty dt e^{(i/\hbar)(E_n - E_m + \hbar\omega + i\eta)t} \\
&= \sum_{n,m} \langle n| S_+(\boldsymbol{q}) |m\rangle \langle m| S_-(-\boldsymbol{q}) |n\rangle \rho_n \left[\frac{e^{(i/\hbar)(\hbar\omega - \Delta E_{mn} + i\eta)t}}{\hbar\omega - \Delta E_{mn} + i\eta} \right]_0^\infty \\
&= \sum_{n,m} \langle n| S_+(\boldsymbol{q}) |m\rangle \langle m| S_-(-\boldsymbol{q}) |n\rangle \frac{-\rho_n}{\hbar} \frac{1}{\omega - \Delta E_{mn}/\hbar + i\eta} \\
&= \sum_{n,m} \langle n| S_+(\boldsymbol{q}) |m\rangle \langle m| S_-(-\boldsymbol{q}) |n\rangle \frac{-\rho_n}{\hbar} \left[\frac{1}{\omega - \Delta E_{mn}/\hbar} - i\pi\delta(\omega - \Delta E_{mn}/\hbar) \right].
\end{aligned} \tag{14A.2}$$

Then eq. (6.111) is obtained. Here in the last part we have used

$$\lim_{\eta \to +0} \frac{1}{x \pm i\eta} = \frac{1}{x} \mp i\pi\delta(x). \tag{14A.3}$$

This equation can be shown from an expression of δ-function as a limit of function:

$$\lim_{\eta \to +0} \frac{1}{\pi} \frac{\eta}{x^2 - \eta^2} = \delta(x).$$

References

[1] Tôru Moriya and Arisato Kawabata. Effect of spin fluctuations on itinerant electron ferromagnetism. *Journal of the Physical Society of Japan*, Vol. 34, No. 3, pp. 639–651, March 1973.

[2] 川畑有郷. 電子相関 (パリティ物理学コース クローズアップ). 丸善, 3 1992.

[3] 上田和夫. 磁性入門 (物性科学入門シリーズ). 裳華房, 9 2011.

[4] Toru Moriya. *Spin Fluctuations in Itinerant Electron Magnetism (Springer Series in Solid-State Sciences, 56)*. Springer, 10 2012.

[5] 高橋慶紀, 吉村一良. 遍歴磁性とスピンゆらぎ. 内田老鶴圃, 単行本, 4 2012.

[6] Yoshinori Takahashi. *Spin Fluctuation Theory of Itinerant Electron Magnetism (Springer Tracts in Modern Physics)*. Springer, 5 2015.

[7] Takeo Izuyama and Ryogo Kubo. Some considerations on the magnetic carriers in ferromagnetic transition metals. *Journal of Applied Physics*, Vol. 35, No. 3, pp. 1074–1081, 1964.

[8] R. Doubble, S. M. Hayden, Pengcheng Dai, H. A. Mook, J. R. Thompson, and C. D. Frost. Direct observation of paramagnons in palladium. *Phys. Rev. Lett.*, Vol. 105, p. 027207, Jul 2010.

[9] A. S. Borovik-Romanov, Y. M. Bunkov, V. V. Dmitriev, and Y. M. Mukharskiy. *JETP Lett.*, Vol. 40, p. 1033, 1984.

[10] G. E. Volovik. Twenty years of magnon bose condensation and spin current superfluidity in 3he-b. *Journal of Low Temperature Physics*, Vol. 153, No. 5-6, pp. 266–284, October 2008.

[11] Jagat Lamsal and Wouter Montfrooij. Extracting paramagnon excitations from resonant inelastic x-ray scattering experiments. *Phys. Rev. B*, Vol. 93, p. 214513, Jun 2016.

[12] M. C. Rahn, K. Kummer, N. B. Brookes, A. A. Haghighirad, K. Gilmore, and A. T. Boothroyd. Paramagnon dispersion in β-fese observed by fe l-edge resonant inelastic x-ray scattering. *Phys. Rev. B*, Vol. 99, p. 014505, Jan 2019.

[13] Kenji Makoshi and Tôru Moriya. Effect of spin fluctuations on the specific heat of weakly and nearly ferromagnetic metals. *Journal of the Physical Society of Japan*, Vol. 38, No. 1, pp. 10–20, 1975.

[14] Jun Takeuchi and Yoshika Masuda. Low temperature specific heat of itinerant electron ferromagnet sc3in. *Journal of the Physical Society of Japan*, Vol. 46, No. 2, pp. 468–474, 1979.