Knight Shift and Nuclear Spin Relaxation Rate in a Charge-Ordered State of the One-Dimensional Extended Hubbard Model at Quarter Filling

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We investigate Knight shift and nuclear spin relaxation rate in a charge ordered state of the one-dimensional extended Hubbard model with a quarter filled band by using RPA around the mean-field solution. It is shown that both quantities show splitting below the critical temperature of the charge order, as is experimentally observed. The relationship between the amount of the splitting in the both quantities and the charge disproportionation rate is discussed.

KEYWORDS: extended Hubbard model, quarter filling, charge order, Knight shift, nuclear spin relaxation rate

1. Introduction

Organic conductors have been known to exhibit a variety of states depending on the temperature, pressure and so on. Such a property is indeed due to the low dimensional electronic structure and the mutual interaction. Thus, the materials are one of the most suitable systems for studying exotic states realized by the interaction between electrons in low dimensional systems.1

Recently, the remarkable state with inhomogeneous but regular charge arrangement called as the charge ordering (CO) has been found in the organic materials with 2:1 composition ratio A$_2$B$_2$, such as (DI-DCNQI)$_2$Ag$_{2-4}$ (TMTTF)$_2X$ and (BEDT-TTF)$_2X$ with X being a monovalent anion. Since the molecule B is ionized as $B^+$ or $B^-$ in order to form a closed shell, the average valence of A becomes $-1/2$ or $+1/2$. As a result, the energy band composed of LUMO or HOMO of the molecule A becomes quarter filled in terms of electrons or holes, respectively. Thus, the CO phenomenon is seen in several molecular conductors with the quarter-filled band. Theoretically, the CO phenomenon has been analyzed based on the extended Hubbard model with the parameters reflecting the actual band and crystal structure, and it has been found that the phenomenon is due to the strong mutual interaction between electrons, especially the repulsion between the nearest neighbor molecules.15–17

The nuclear magnetic resonance (NMR) is the powerful experimental technique which can explore the local electronic state. It has been considered to be one of the theoretical experiments to study the charge disproportionation is discussed. The paper is organized as follows. The model we consider and it’s solution by the mean-field treatment are shown in §2. The formulation for calculating the Knight shift and the nuclear spin relaxation rate based on the path integral method is also given in §2. The obtained results are shown in §3. §4 is devoted to Summary.

2. Model and Formulation

We consider the one-dimensional extended Hubbard model at the quarter filling written by the following Hamiltonian, $\mathcal{H} = \mathcal{H}_k + \mathcal{H}_{\text{int}}$,

$$\mathcal{H}_k = -t \sum_{j,s} (c_{j,s}^\dagger c_{j+1,s} + h.c.), \quad (1)$$

$$\mathcal{H}_{\text{int}} = \frac{U}{2} \sum_{j,s} n_{j,s}n_{j,-s} + V \sum_{j,s,s'} n_{j,s}n_{j+1,s'}, \quad (2)$$

where $t$, $U$ and $V$ are the transfer energy between the nearest-neighbor site, the on-site repulsion and the interaction between the nearest-neighbor site, respectively. Here, $c_{j,s}$ denotes the creation operator of the electron at the $j$-th site with spin $s = \pm$, $n_{j,s} = c_{j,s}^\dagger c_{j,s}$ and $n_j = \sum_s n_{j,s}$. The possible ordered states of the model obtained by the standard mean-filed approximation are 4$k_F$-CDW (CO), 2$k_F$-SDW and the coexistence of both...
order realized from 5.5 K to 220 K in (DI-DCNQI) responding to the charge ordered state without magnetic lowing, we assume that the pure 4 \text{k}_F-CDW and the coexistent state of 4\text{k}_F-CDW and 2\text{k}_F-SDW, respectively.

![Fig. 1. The phase diagram on the plane of V/t and T/t at U/t = 4.0. Here, (I), (II) and (III) denote the normal state, 4\text{k}_F-CDW and the coexistent state of 4\text{k}_F-CDW and 2\text{k}_F-SDW, respectively.](image)

Fig. 1. The phase diagram on the plane of V/t and T/t at U/t = 4.0. Here, (I), (II) and (III) denote the normal state, 4\text{k}_F-CDW and the coexistent state of 4\text{k}_F-CDW and 2\text{k}_F-SDW, respectively. Note that the pure 2\text{k}_F-SDW appears for the small V region, which is not shown in Fig.1. With decreasing temperature, the transition from the normal state to the coexistent state occurs for V < V_c, whereas the pure 4\text{k}_F-CDW is realized between them for V > V_c. Figure 2 shows the charge disproportionation \rho at U/t = 4.0 as a function of T/t for the several choices of V, where the quantity \rho is defined as \langle n_j \rangle = 1/2 - (-1)^j \rho. At the transition temperature of the coexistent state, the charge disproportionation shows sudden increase. The transition from the normal state to the coexistent state is continuous, whereas the transition become the first order for V ∼ V_c. In the following, we assume that the pure 4\text{k}_F-CDW state (II) corresponds to the charge ordered state without magnetic order realized from 5.5 K to 220 K in (DI-DCNQI)\textsubscript{2}Ag and investigate Knight shift and the nuclear spin relaxation rate in the state (II) by using RPA around the mean-field solution.

The fluctuation around the mean-filed solution can be taken into account by the path integral method. The effective action up to the second order of the charge and spin fluctuation, which correspond to the RPA approximation where the coupling between the different modes is neglected, is divided into the spin and the charge part (see Appendix). The spin part, S_σ is written as follows,

\[ S_σ = \sum_{0 \leq q < Q_0} \sum_{\omega_n} \{ \langle - \delta σ(q, -iω_n) - δσ(q - Q_0, -iω_n) \rangle \]

\[ \times \begin{pmatrix} A_σ(q, iω_n) & B_σ(q, iω_n) \\ B_σ(q, iω_n) & A_σ(q - Q_0, iω_n) \end{pmatrix} \]

\[ \times \begin{pmatrix} \delta σ(q, iω_n) \\ \deltaσ(q - Q_0, iω_n) \end{pmatrix}, \]  

(3)

where \( Q_0 = π/a \) with \( a \) being the lattice spacing and

\[ A_σ(q, iω_n) = -\frac{v_s(q)}{N_L} \{ 1 + 4v_s(q)K(q, iω_n) \}, \]  

(4)

\[ B_σ(q, iω_n) = -\frac{4v_s(q)v_s(q - Q_0)}{N_L}H(q, iω_n), \]  

(5)

with \( v_s(q) = -U/4 \) and \( N_L \) is the total number of the lattice. Here, the quantities \( K(q, iω_n) \) and \( H(q, iω_n) \) are defined as follows,

\[ K(q, iω_n) = -\frac{1}{N_L} \sum_k \left\{ \frac{1}{4} \left( \frac{1}{E_k + E_{k+q}} + \frac{1}{E_k - E_{k+q}} \right) \right. \]

\[ \times \left( \frac{f(E_k - μ) - f(E_{k+q} - μ)}{iω_n + E_k - E_{k+q}} \right. \]

\[ \left. + \frac{f(-E_k - μ) - f(-E_{k+q} - μ)}{iω_n - E_k + E_{k+q}} + \frac{1}{4} \left( \frac{1}{E_k - E_{k+q}} + \frac{Δ}{E_k + E_{k+q}} \right) \right\} \]

\[ \times \left( \frac{f(E_k - μ) - f(E_{k+q} - μ)}{iω_n + E_k - E_{k+q}} \right. \]

\[ \left. + \frac{f(-E_k - μ) - f(-E_{k+q} - μ)}{iω_n - E_k + E_{k+q}} \right\}, \]  

(6)

and

\[ H(q, iω_n) = -\frac{1}{N_L} \sum_k \left\{ \frac{1}{4} \left( \frac{1}{E_k + E_{k+q}} + \frac{Δ}{E_k + E_{k+q}} \right) \right. \]

\[ \times \left( \frac{f(E_k - μ) - f(E_{k+q} - μ)}{iω_n + E_k - E_{k+q}} \right. \]

\[ \left. - \frac{f(-E_k - μ) - f(-E_{k+q} - μ)}{iω_n - E_k + E_{k+q}} + \frac{1}{4} \left( \frac{Δ}{E_k + E_{k+q}} \right) \right\} \]

\[ \times \left( \frac{f(E_k - μ) - f(-E_{k+q} - μ)}{iω_n + E_k - E_{k+q}} \right. \]

\[ - \frac{f(-E_k - μ) - f(E_{k+q} - μ)}{iω_n - E_k + E_{k+q}} \right\}, \]  

(7)
correlations of the spin fluctuation are obtained as fol-

to the local magnetic moment at the

The Knight shift

Here, \( \mu \) is the chemical, \( c_k = -2t \cos k a \) and \( E_k = \sqrt{c_k^2 + \Delta^2} \). The energy gap \( \Delta \geq 0 \) which opens at \( \pm \pi/(2a) \) is related to the charge disproportionation \( \rho \) as

\[
\Delta = -2 v_c(q_0) \rho. \tag{8}
\]

where \( v_c(q) = U/4 + V \cos qa \). By using the action, the correlations of the spin fluctuation are obtained as follows,

\[
\langle \delta \sigma(q, i \omega_n) \delta \sigma(-q, -i \omega_n) \rangle = \frac{1}{2} A_\sigma(q - Q_0, i \omega_n) A_\sigma(q - Q_0, -i \omega_n) - B_\sigma^2(q, i \omega_n), \tag{9}
\]

\[
\langle \delta \sigma(q, i \omega_n) \delta \sigma(-q + Q_0, -i \omega_n) \rangle = \frac{1}{2} B_\sigma(q, i \omega_n) A_\sigma(q - Q_0, i \omega_n) - B_\sigma^2(q, i \omega_n), \tag{10}
\]

for \(-\pi/a \leq q < \pi/a \) and the others are zero. Note that the CO leads to the coupling between \( q \)-component and \(-q + Q_0 \)-component in the fluctuation because the spacing between the equivalent site becomes twice. In the case of the half filling, \( i.e., \mu = 0 \), such a coupling diminishes because the quantity \( H(q, i \omega_n) \) in eq. (7) vanishes.

The spin susceptibility in the site representation, \( \chi_\sigma(x_i, x_j; i \omega_n) \) is obtained as follows,

\[
\chi_\sigma(x_i, x_j; i \omega_n) = \frac{1}{N_L} \sum_{q, q'} e^{-i(q \cdot x_i + q \cdot x_j)} \int_0^\beta d\tau e^{i \omega_n \tau} \langle T_\tau (m(q', \tau) m(q, 0)) \rangle
\]

\[
eq \frac{1}{N_L} \sum_{q, q'} e^{-i(q \cdot x_i + q \cdot x_j)}
\]

\[
\times \left\{ \langle \delta \sigma(q, i \omega_n) \delta \sigma(q', -i \omega_n) \rangle + \delta_{q, -q'} - \frac{1}{2} v_c(q) \right\}
\]

\[
= \frac{1}{N_L} \sum_q e^{i q (x_i - x_j)}
\]

\[
\times \left\{ \chi_\sigma(q, -q; i \omega_n) + (-1)^l \chi_\sigma(q, -q + Q_0; i \omega_n) \right\}, \tag{11}
\]

where

\[
\chi_\sigma(q, -q; i \omega_n) = \frac{\langle \delta \sigma(q, i \omega_n) \delta \sigma(-q, -i \omega_n) \rangle}{N_L} + \frac{1}{2} v_c(q), \tag{12}
\]

\[
\chi_\sigma(q, -q + Q_0; i \omega_n) = \frac{\langle \delta \sigma(q, i \omega_n) \delta \sigma(-q + Q_0, -i \omega_n) \rangle}{N_L}. \tag{13}
\]

The Knight shift \( S_i \) at the location \( x_i \) is proportional to the local magnetic moment at the \( i \)-th site under the uniform and static magnetic magnetic field. Therefore, \( S_i \) is written by using eqs.(12) and (13) as follows,

\[
S_i \propto \sum_j \chi_\sigma(x_i, x_j; 0)
\]

\[
\propto \chi_\sigma(0, 0; 0) + (-1)^l \chi_\sigma(0, Q_0; 0). \tag{14}
\]

Thus, the average between the Knight shift at the charge rich site and that at the poor site is proportional to the magnetic susceptibility. \( \chi_\sigma(0, 0; 0) \) is obtained as follows,

\[
R_i \propto \lim_{\omega \to 0} \frac{\text{Im} \chi_\sigma(x_i, x_i; \omega + i\delta)}{\omega}
\]

\[
eq \frac{1}{N_L} \sum_q \lim_{\omega \to 0} \left\{ \frac{\text{Im} \chi_\sigma(q, -q; \omega + i\delta)}{\omega} + (-1)^l \frac{\text{Im} \chi_\sigma(q, -q + Q_0; \omega + i\delta)}{\omega} \right\}, \tag{15}
\]

where \( \delta = +0 \). Note that the transverse susceptibility is the same as the longitudinal one because the order of the spin does not appear in the region (II). The charge rich and poor sites are the odd site (\( i=\text{odd} \)) and the even site (\( i=\text{even} \)) respectively. In the case of the half-filling, \( i.e., \mu = 0 \), the quantity \( H(q, i \omega_n) \) in eq. (7) vanishes, as is already noted. Therefore, the Knight shift and the relaxation rate do not show splitting. However, in the present quarter-filling case, the splitting of both quantities appear, as will be shown in the next section concretely.

3. Results

In this section, we discuss the Knight shift and the nuclear spin relaxation rate in the pure CO state (II) in the phase diagram of Fig. 1.

Figure 3 shows the Knight shift at the charge rich site \( S_+ \) and that at the poor site \( S_- \) as a function of temperature for \( U/t = 4.0 \) and \( V/t = 3.0 \). These quantities are normalized by the shift in the non-interacting case at \( T = 0, S_0 \). The shift splits into the two components below \( T_{CO} \) and the quantity at the rich site is larger than that at the poor site. The fact can be easily understood by considering the case where the charge order is complete, \( i.e., \rho = 1/2 \). In the charge poor site where there are no carriers, the spin excitation is impossible. Therefore the shift is equal to zero. On the other hand, in the charge rich site with one electron per site, the spin excitation is possible and the Knight shift is finite. The dashed curve and dotted one express the average of \( S_+ \) and \( S_- \), \( i.e., \chi_\sigma(0, 0; 0) \) and \( \chi_\sigma(0, Q_0; 0) \) which has been observed experimentally.

The nuclear spin relaxation rate for \( U/t = 4.0 \) and \( V/t = 3.0 \) normalized by the quantum in the absence of
the interaction at \( T = 0 \), \( R_0 \) is shown in Fig. 4, where \( R_+ \) and \( R_- \) are \((T_1T)^{-1}\) at the charge rich site and that of poor site, respectively. Here the dashed curve expresses the average of \( R_+ \) and \( R_- \) and the rate in the absence of \( \text{CO} \) is shown by the dotted curve. As is seen in the Knight shift, the rate shows splitting and the quantity \( R_+ \) becomes larger \( R_- \) because the spin fluctuation at the charge rich site is larger than that at the poor site. Note that the average seems to have a singularity at \( T = T_{\text{CO}} \), which is different from the magnetic susceptibility.

Next, the relationship between the mount of the splitting in the these quantities and the charge disproportionation \( \rho \) is discussed. The experiments have been analyzed by assuming that the relative shift, \( S_+/(S_++S_-) \) or \( S_-/(S_++S_-) \) measures the local charge\(^2\) and that the relaxation rate is proportional to the local charge \((1/2 \pm \rho)^2\)\(^7\) respectively. If the assumptions are valid, the following relationship should hold,

\[
\frac{S_+ - S_-}{S_+ + S_-} = 2\rho, \tag{16}
\]

\[
\frac{R_+ - R_-}{R_+ + R_-} = \frac{2\rho}{1/2 + 2\rho^2}. \tag{17}
\]

Figures 5 and 6 show the l.h.s. of eq.(16) and that of eq.(17) as a function of the r.h.s. of the respective equations. In Fig. 5, the relative shift is well proportional to \( 2\rho \), but the coefficient becomes larger than unity. The coefficient approaches to unity when \( V \) increases. Similarly, the quantity \((R_+ - R_-)/(R_+ + R_-)\) approaches to \( 2\rho/(1/2 + 2\rho^2) \) for large \( V \). Thus, the assumptions, eqs. (16) and (17), seems to hold approximately well for the

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**Fig. 3.** The normalized Knight shift at the charge-rich site \( S_+ / S_0 \) and that at the poor site \( S_- / S_0 \) as a function of the temperature for the pure charge ordered state in the case of \( U/t = 4.0 \) and \( V/t = 3.0 \). Here \( S_0 \) is the shift of the non-interaction case at \( T = 0 \). The dashed curve expresses the the average of \( S_+ \) and \( S_- \), i.e., magnetic susceptibility. The shift without taking account of the charge order is denoted by the dotted curve.

**Fig. 4.** The normalized nuclear spin relaxation rate at the charge-rich site \( R_+ / R_0 \) and that at the poor site \( R_- / R_0 \) as a function of the temperature for the pure charge ordered state in the case of \( U/t = 4.0 \) and \( V/t = 3.0 \). Here \( R_0 \) is the rate of the non-interaction case at \( T = 0 \). The dashed curve expresses the the average of \( R_+ \) and \( R_- \). The rate without taking account of the charge order is denoted by the dotted curve.

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**Fig. 5.** The mount of splitting of the Knight shift \( S_+ - S_- \) normalized by \( S_+ + S_- \) as a function of \( 2\rho \) for several choices of \( V/t \) at \( U/t = 4.0 \).

**Fig. 6.** The mount of splitting of the nuclear spin relaxation rate \( R_+ - R_- \) normalized by \( R_+ + R_- \) as a function of \( 2\rho/(1/2 + 2\rho^2) \) for several choices of \( V/t \) at \( U/t = 4.0 \).
large $V$ in which the CO state becomes robust, but with decreasing $V$ such relationship become worse. It should be noted that eqs. (16) and (17) gives the smaller value of the charge disproportionation that the actual one.\(^7\)

### 4. Summary

In the present paper, we investigated the Knight shift and the nuclear spin relaxation rate in the state with charge ordering based on the one-dimensional extended Hubbard model with a quarter filled band with taking account of the fluctuation around the mean-field solution by RPA. It is shown that both quantities shows splitting below the critical temperature of the charge order. The result is different from the case of the half-filling where the splitting does not occur even in the CO state due to the particle-hole symmetry. The quantities at the charge rich site are larger than those at the poor site due to the larger spin fluctuation. Thus, in the materials with the quarter filling, the splitting of these quantities can be considered to be the experimental evidence of appearance of the charge order as has been already experimentally insisted. The relationship between the amount of the splitting normalized by the average is discussed. It is shown that the assumptions used in the analysis of the experiments hold well when $V$ becomes larger, i.e., the CO is robust, though the estimated mount of the charge disproportionation becomes generally smaller compared with the actual value.

In the present study, we used the method in which we apply the mean-field treatment to the present model and take into account of the fluctuation around the solution by using RPA. Though this method can succeed to show the splitting of the Knight shift and the relaxation rate, the transport properties of the actual materials such as (DL-DNCNQI)$_2$Ag cannot be described. The materials show the insulating behavior even at the room temperature, i.e., $T > T_{\text{CO}}$.\(^2\) In addition, the derivative of the resistivity in terms of the temperature has a peak at $T = T_{\text{CO}}$.\(^21\) where the Metal-Insulator transition occurs. On the other hand, the present mean-field treatment leads to the metallic state both $T > T_{\text{CO}}$ and $T < T_{\text{CO}}$. That is, the metal insulator transition do not happen even at $T = T_{\text{CO}}$ because the energy gap opens at not $\pm k_F$ but $\pm 2k_F$. Therefore the further studies are necessary to describe the CO state consistently in both the transport and magnetic properties.

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### Appendix: Effective action of the fluctuation around the pure $4k_F$-CDW state

The partition function corresponding to the Hamiltonian, (1) and (2), is written by utilizing the Stratonovich-Hubbard transformation as follows,

$$Z = \int \mathcal{D}(c^*,c)\mathcal{D}\rho \mathcal{D}\sigma e^{-(S_k+S_{\text{int}})},$$  \hspace{1cm} (A-1)

$$S_k = \int^\beta_0 d\tau \sum_{k,s} c^*_k c_s(\tau)(\partial_\tau - \mu + \epsilon_k)c_k(\tau),$$  \hspace{1cm} (A-2)

$$S_{\text{int}} = \int^\beta_0 d\tau \sum_q \left\{ \frac{v_s(q)}{N_L} \left[ -\rho(q,\tau)\rho(-q,\tau) + 2\rho(q,\tau)n(-q,\tau) \right] + \frac{v_s(q)}{N_L} \left[ -\sigma(q,\tau)\sigma(-q,\tau) + 2\sigma(q,\tau)m(-q,\tau) \right] \right\},$$  \hspace{1cm} (A-3)

where $\mu$ is the chemical potential, $N_L$ is the total number of sites, $\beta = 1/T$, $v_s(q) = U/4 + V \cos qa$, $v_c(q) = -U/4$, $n(q,\tau) = \sum_{k,s} \epsilon_k c^*_k c_s(\tau)c_k s(\tau)$ and $m(q,\tau) = \sum_{k,s} \sigma c^*_k c_s(\tau)c_k s(\tau)$. Here we assume that the pure $4k_F$-CDW is realized. Then the Stratonovich-Hubbard fields, $\rho(q,\tau)$ and $\sigma(q,\tau)$, are rewritten as follows,

$$\rho(q,\tau) = \begin{cases} \rho(0) + \delta\rho(0,\tau), & \text{for } q = 0 \\ \rho(Q_0) + \delta\rho(Q_0,\tau), & \text{for } q = Q_0 \\ \delta\rho(q,\tau), & \text{for the others} \end{cases}$$  \hspace{1cm} (A-4)

$$\sigma(q,\tau) = \delta\sigma(q,\tau).$$  \hspace{1cm} (A-5)

When eqs. (A-4) and (A-5) are substituted into (A-3), the action of the interaction part is divided as $S_{\text{int}} = S_{\text{int,0}} + S_{\text{int,1}} + S_{\text{int,2}}$, where

$$S_{\text{int,0}} = -\beta\frac{v_c(0)}{N_L} \rho(0)^2 - \beta\frac{v_s(Q_0)}{N_L} \rho(Q_0)^2,$$  \hspace{1cm} (A-6)

$$S_{\text{int,0}} = \int^\beta_0 d\tau \left\{ \sum_{q\neq0,Q_0} 2\frac{v_c(q)}{N_L} \delta\rho(q,\tau)n(-q,\tau) \\ + 2\frac{v_s(Q_0)}{N_L} (n(0,\tau) - \rho(0))\delta\rho(0,\tau) \\ + 2\frac{v_s(Q_0)}{N_L} (n(Q_0,\tau) - \rho(Q_0))\delta\rho(Q_0,\tau) \\ + \sum_q 2\frac{v_s(q)}{N_L} \delta\sigma(q,\tau)m(-q,\tau) \right\},$$  \hspace{1cm} (A-7)

$$S_{\text{int,1}} = \int^\beta_0 d\tau \left\{ \sum_{q\neq0,Q_0} 2\frac{v_s(q)}{N_L} \delta\rho(q,\tau)n(-q,\tau) \\ + 2\frac{v_s(Q_0)}{N_L} (n(0,\tau) - \rho(0))\delta\rho(0,\tau) \\ + 2\frac{v_s(Q_0)}{N_L} (n(Q_0,\tau) - \rho(Q_0))\delta\rho(Q_0,\tau) \\ + \sum_q 2\frac{v_s(q)}{N_L} \delta\sigma(q,\tau)m(-q,\tau) \right\},$$  \hspace{1cm} (A-8)

$$S_{\text{int,2}} = -\int^\beta_0 d\tau \sum_q \left\{ \frac{v_s(q)}{N_L} \delta\rho(q,\tau)\delta\rho(-q,\tau) \\ + \frac{v_s(q)}{N_L} \delta\sigma(q,\tau)\delta\sigma(-q,\tau) \right\}.$$  \hspace{1cm} (A-9)
By integrating out the electron degree of freedom, the partition function is calculated as,
\[ Z = e^{-S_{\text{Int,0}}} Z_e^0 \int \mathcal{D}c \mathcal{D}c' e^{-\langle S_{\text{Int}} \rangle} \langle \exp(-S_{\text{Int,1}}) \rangle, \] (A-10)
where \( Z_e^0 = \int \mathcal{D}c \mathcal{D}c' e^{-\langle S_e + S_{\text{Int,0}} \rangle} \) and \( \langle \cdots \rangle \) denotes the average in terms of \( S_k + S_{\text{Int,0}} \) given as follows,
\[
S_k + S_{\text{Int,0}} = \int_0^\beta d\tau \sum_{0 < k \leq Q_0} (c_k^+, \sigma) (c_k, \sigma) \hspace{1cm} \times \left( \frac{\partial \tau - \mu + \epsilon_k}{\Delta} \right) \left( \frac{\partial \tau - \mu - \epsilon_k}{\Delta} \right) \langle c_k, \sigma \rangle (c_k, \sigma) \right),
\] (A-11)
with \( \Delta = 2v_c(Q_0)\rho(Q_0)/N_L = -2v_c(Q_0)\rho \). Here
\[
\langle A \rangle = \frac{1}{\beta} \sum_{\epsilon_n} e^{-\epsilon_n \tau} \left( \frac{G(k, \tau)}{F(k - Q_0, \tau)} \right) \left( \frac{F(k, \tau)}{G(k - Q_0, \tau)} \right),
\] (A-12)
\[
G(k, \tau) = \frac{u_k^2}{\epsilon_n + \mu - E_k} + \frac{v_k^2}{\epsilon_n + \mu + E_k},
\] (A-13)
\[
F(k, \tau) = \frac{u_k v_k}{\epsilon_n + \mu - E_k} - \frac{u_k v_k}{\epsilon_n + \mu + E_k},
\] (A-14)
for \(-Q_0 < k \leq Q_0\) with \( E_k = \sqrt{\epsilon_k^2 + \Delta^2} \) (\( \Delta > 0 \)) and
\[
u = \frac{1}{2} \left( 1 + \frac{\epsilon_k}{E_k} \right),
\] (A-15)
\[
u = \frac{1}{2} \left( 1 - \frac{\epsilon_k}{E_k} \right).
\] (A-16)
We expand \( e^{-S_{\text{Int,1}}} \) up to the second order. The condition \( \langle S_{\text{Int,1}} \rangle = 0 \) gives rise to the following equations,
\[
\rho(0) = \frac{N_L}{2} = 2 \sum_{0 < k \leq Q_0} \{ f(-E_k - \mu) + f(E_k - \mu) \},
\] (A-17)
\[
\Delta = 4v_c(Q_0) \frac{1}{N_L} \sum_{0 < k \leq Q_0} \frac{\Delta}{E_k} \{ f(E_k - \mu) - f(-E_k - \mu) \}.
\] (A-18)
Note that the chemical potential \( \mu \) is determined by eq.(A-17) and eq.(A-18) is the self-consistent equation of \( \Delta \). On the other hand, the quantity \( \langle S_{\text{Int,1}}^2 \rangle/2 \) is calculated as follows,
\[
\frac{\langle S_{\text{Int,1}}^2 \rangle}{2} = \frac{1}{2} \int_0^\beta d\tau' \sum_{q, q'} \left\{ \frac{2v_c(q) 2v_c(q')}{N_L} \delta(q, q') \delta(q', \tau') \langle n(-q, \tau) \cdots n(-q', \tau') \rangle + \frac{2v_c(q) 2v_c(q')}{N_L} \delta(q, q') \delta(q', \tau') \langle m(-q, \tau) \cdots m(-q', \tau') \rangle \right\}
\] (A-19)
where \( : A := A - \langle A \rangle \) and
\[
K(q, \tau) = -\frac{1}{N_L} \sum_k \left\{ G(k - q, -\tau)G(k, \tau) + F(k - q, -\tau)F(k, \tau) \right\},
\] (A-20)
\[
H(q, \tau) = -\frac{1}{N_L} \sum_k \left\{ G(k - q, -\tau)F(k, \tau) + F(k - q, -\tau)G(k, \tau) \right\},
\] (A-21)
The quantities, \( K(q, \omega_n) \) and \( H(q, \omega_n) \) are given as follows,
\[
K(q, \omega_n) = -\frac{1}{\beta N_L} \sum_{k, \epsilon_n} \left\{ G(k - q, \omega_n)G(k, \epsilon_n + \omega_n) + F(k - q, \epsilon_n)F(k, \epsilon_n + \omega_n) \right\},
\] (A-22)
\[
H(q, \omega_n) = -\frac{1}{\beta N_L} \sum_{k, \epsilon_n} \left\{ G(k - q, \omega_n)F(k, \epsilon_n + \omega_n) + F(k - q, \epsilon_n)G(k, \epsilon_n + \omega_n) \right\},
\] (A-23)
and obtained in eq.(6) and (7), respectively. When the Fourier transformation, \( \delta \nu(q, \omega_n) = 1/\sqrt{\beta} \int_0^\beta d\tau \exp(-\imath \omega_n \tau) \delta \nu(q, \tau) \) (\( \nu = \rho, \sigma \)) is introduced, eq.(A-19) is written as follows,
\[
\frac{\langle S_{\text{Int,1}}^2 \rangle}{2} = \sum_{0 < q \leq Q_0} \sum_{\omega_n} \left\{ \delta \nu(-q, -\omega_n), \delta \rho(-q + Q_0, -\omega_n) \right\} \times \left( \frac{4v_c^2(q)}{N_L} K(q, \omega_n) \right),
\] (A-24)
On the other hand, $S_{\text{int}2}$ is written as follows,

\[
S_{\text{int}2} = - \sum_{0 < q \leq Q_0} \sum_{\omega_n} \left( \delta\rho(-q, -i\omega_n), \delta\rho(-q + Q_0, -i\omega_n) \right) \times \left( \frac{\nu_c(q)}{N_L} \right) \frac{\nu_c(q - Q_0)}{N_L} \delta\sigma(q, \omega_n) \delta\sigma(q - Q_0, \omega_n).
\]

Thus the effective action $S_{\text{eff}} = S_{\text{int}2} - \langle S_{\text{int}1}^2 \rangle / 2$ is given by the sum of the charge and spin part, $\sum_{\nu = \rho, \sigma} S_{\nu}$, where

\[
S_{\nu} = \sum_{0 < q \leq Q_0} \sum_{\omega_n} (\delta\nu(-q, -i\omega_n), \delta\nu(-q + Q_0, -i\omega_n)) \times \left( \begin{array}{cc}
A_{\nu}(q, \omega_n) & B_{\nu}(q, \omega_n) \\
B_{\nu}(q, \omega_n) & A_{\nu}(q - Q_0, \omega_n)
\end{array} \right) \times \left( \begin{array}{cc}
\delta\nu(q, \omega_n) \\
\delta\nu(q - Q_0, \omega_n)
\end{array} \right),
\]

with

\[
A_{\nu}(q, \omega_n) = -\frac{\nu_c(q)}{N_L} \left\{ 1 + 4\nu_c(q)K(q, \omega_n) \right\}, \quad B_{\nu}(q, \omega_n) = -\frac{4\nu_c(q)\nu_c(q - Q_0)}{N_L} H(q, \omega_n),
\]

\[
A_{\nu}(q, \omega_n) = -\frac{\nu_s(q)}{N_L} \left\{ 1 + 4\nu_s(q)K(q, \omega_n) \right\}, \quad B_{\nu}(q, \omega_n) = -\frac{4\nu_s(q)\nu_s(q - Q_0)}{N_L} H(q, \omega_n).
\]