Effective indirect multi-site spin-spin interactions
in the s-d(f) model

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Using the diagram technique for Matsubara Green’s functions it is shown that the dynamics of the localized spin subsystem in the s-d(f) model can be described in terms of an effective spin model with multi-site spin-spin interactions. An exact representation of the action for the effective purely spin model is derived as an infinite series in powers of s-d(f) exchange interaction $J$. The indirect interactions of the 2-nd, 3-rd and 4-th order are discussed.

1. Introduction

Kondo lattice model or s-d(f) exchange model is widely used to describe the correlation effects in metals (and their compounds) with unfilled $d$- and $f$-shells.

The nature of the ground state in these systems is largely determined by the result of competition between two interactions. On the one hand, s-d(f) exchange coupling between spins of itinerant $s$- and localized $d$($f$)- electrons due to Kondo fluctuations screens the localized spins and tends to form a non-magnetic ground state [1]. In the opposite direction acts indirect exchange (RKKY) interaction between spins of d($f$)-electrons [2], trying to set a long-rang magnetic order which is not necessarily a ferromagnetic or antiferromagnetic. Correlation effects can lead to, for example, helical magnetic structures [3], with period that is determined by the Fermi surface singularities [5, 6].

It is clear that the study of competition between different effective interactions, that occur in the localized spin subsystem and are caused by the same itinerant electrons, must be carried out within the same unified approach.

The purpose of this paper is to derive such an effective Hamiltonian (or rather action), which will allow to study the localized spin subsystem in the s-d(f) model within the model with only spin-spin interactions. At the same time all possible spin-spin interactions that occur in different orders of perturbation theory in $J$, should be determined by the same model parameters: 1) the s-d(f)-exchange coupling constant $J$; 2) the dispersion law of conduction electrons; 3) the filling degree of the conduction band. Such an approach should allow to analyze all types of exchange spin-spin interactions from a unified point of view.

This problem is solved by integrating charge degrees of freedom using the diagram technique for Matsubara Green’s functions. It is shown that in addition to the two-spin indirect exchange interaction, which occurs in the second order in s-d(f)-parameter $J$, in the following orders also appear terms describing, in particular, the ring and biquadratic exchange interactions. The essential point of all these interactions is the account for retardation effects provided by the imaginary time dependence of all the effective multi-site exchange parameters.
2. The Hamiltonian of the s-d(f) exchange model

The Hamiltonian of the Kondo lattice model (or s-d(f) exchange model) can be written as a sum of two terms:

\[ \hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}, \]

where

\[ \hat{H}_0 = \sum_{k \alpha} (\epsilon_k - \mu) c^+_k \alpha c_k \alpha, \quad \hat{H}_{\text{int}} = \frac{J}{2} \sum_f c^+_f \tilde{S}_f c_f. \]

Operator \( \hat{H}_0 \) stands for the energy of noninteracting current carriers (electrons or holes) with dispersion \( \epsilon_k \), \( \mu \) – chemical potential. Operator \( c^+_k \alpha c_k \alpha \) creates (annihilates) a particle in the state with quasimomentum \( k \) and spin projection \( \alpha = \pm 1/2 \).

The second term in (1) describes Kondo exchange interaction between localized spins and itinerant quasiparticles. The intensity of this interaction is defined by the constant \( J \). Operator \( \tilde{S}_f \) is a product of a localized spin vector operator \( \tilde{S}_f = (S^x_f, S^y_f, S^z_f) \) and a vector \( \tilde{\sigma} = (\sigma_x, \sigma_y, \sigma_z) \) which is formed of Pauli matrices. In definition (2) the spinor notations:

\[ c^+_f = (c^+_f \uparrow, c^+_f \downarrow), \quad c_f = (c_f \uparrow, c_f \downarrow), \quad \tilde{S}_f = \begin{pmatrix} S^x_f & S^y_f & S^z_f \\ S^y_f & S^z_f & -S^x_f \end{pmatrix}, \quad S^\pm_f = S^x_f \pm iS^y_f, \]

are used. The operators \( c_f \) and \( c_k \) are related to each other by Fourier transformation: \( c_f = N^{-1/2} \sum_k e^{ikf} c_k \).

3. Spin Green’s functions and effective action

The thermodynamic properties of localized spin subsystem conveniently studied on the basis of the diagram technique for Matsubara Green’s functions [7]:

\[ G^{jj'}(f - f', \tau - \tau') = -\left\langle T\tau \tilde{S}_j(\tau)\tilde{S}_{j'}(\tau') \right\rangle, \quad j, j' = \{x, y, z\}. \]

In this expression the spin operators are written down in the Heisenberg representation: \( \tilde{S}_j(\tau) = e^{\tau \hat{H}} S^j_0 e^{-\tau \hat{H}} \), where \( \hat{H} \) is the s-d(f) exchange Hamiltonian [11], and \( \tau \) – imaginary time varying within interval \((0, 1/T)\) \( (T \) is the temperature). The imaginary time ordering operator \( T\tau \) arranges all operators on the right side of it in the descending order of the imaginary time \( \tau \) from left to right. Angle brackets in (4) denote a thermodynamic average over the grand canonical ensemble described by the Hamiltonian \( \hat{H} \).

As is known [8], in the interaction representation: \( S^j_0(\tau) = e^{\tau \hat{H}_0} S^j_0 e^{-\tau \hat{H}_0} \), the expression (4) transforms into:

\[ G^{jj'}(f - f', \tau - \tau') = -\left\langle T\tau S^j_0(\tau)S^j_0(\tau') \mathcal{G}(\beta) \right\rangle_0. \]

Here the scattering matrix \( \mathcal{G}(\beta) \) is defined by the formula:

\[ \mathcal{G}(\beta) = T\tau \exp \left\{ -\int_0^\beta d\tau \hat{H}_{\text{int}}(\tau) \right\}, \]

(6)
where in the exponent under the integral stands the interaction operator (2) in the interaction representation. Lower index "0", on the right of the angle brackets in (5) indicates that the thermodynamic averaging is over the ensemble of non-interacting spin and fermion systems. Besides, when expanding the thermodynamic averages using Wick’s theorem only connected diagrams should be considered.

Note that for calculating the Green’s function (5) along with the usual diagram technique (8) it is necessary to use the diagram technique for spin operators (9) as well. The expanding of a \( T_\tau \)-ordered average of a product of spin \( S \)- and fermion \( c \)-operators can be divided into two stages: first, only \( c \)-operators are paired, and then the Wick’s theorem is applied to the remaining spin operators. Formally this can be written down as follows:

\[
\langle T_\tau S_l \ldots S_1 c_1 \ldots c_m \rangle_0 = \left\langle T_\tau S_l \ldots S_1 \langle T_\tau c_1 \ldots c_m \rangle_{c0} \right\rangle_{S0}. \tag{7}
\]

Internal thermodynamic average on the right side of the equation (7) with index "0" denotes averaging only over an ensemble of non-interacting fermions. External \( T_\tau \)-ordered averaging, indicated by the index "S0", should be done using the ensemble of non-interacting spin subsystems. Applying equation (7) to the definition of the Green’s function (5) we can write:

\[
G_{jj'}(f - f', \tau - \tau') = -\left\langle T_\tau S_j^\dagger(\tau) S_{j'}(\tau') \mathcal{G}(\beta) \right\rangle_{S0}, \tag{8}
\]

where the effective scattering matrix \( \mathcal{G}(\beta) \) is defined by the expression:

\[
\mathcal{G}(\beta) = \langle \mathcal{G}(\beta) \rangle_{c0}. \tag{9}
\]

To obtain the effective purely spin model first we should pair all the \( c \)-operators according to the Wick’s theorem in each order of the \( \mathcal{G} \)-matrix expansion in powers of the coupling constants \( J \). After that the result must be rewritten as a \( T_\tau \)-ordered exponent. The argument of the exponent will determine the required effective interaction in the subsystem of localized spins. Note that the described here scheme of integrating over the charge degrees of freedom in the s-d(f) model in some sense is similar to the proof of equivalence between diagrammatic expansion for the Green’s function of localized \( f \)-electrons in the periodic Anderson model and diagrammatic expansion of the fermion Green’s function in the Hubbard model [10, 11].

Let us consider for the average \( \langle \mathcal{G}(\beta) \rangle_{c0} \) the diagrams of \( n \)-th order with respect to the constant \( J \). Since each term in \( \hat{H}_{int} \) consists of one spin operator and two second-quantization operators (\( c^+ \) and \( c \)), the structure of diagrams emerging after all possible pairings of \( c \)-operators is characterized by a some number of fermionic loops (see figure 1). The order of each loop is determined by the number of fermionic lines, corresponding to the fermion Green’s functions \( G^{(0)}_k(\tau - \tau') = -\langle T_\tau c_k(\tau)c_k^+(\tau') \rangle_0 \), and the same number of vertices. In contrast to the conventional diagram technique in this case each vertex is related not to a function (or a number) but to the spin operator. Therefore the \( n \)-th order diagram corresponds to a product of \( n \) spin operators entering to the \( T_\tau \)-ordered thermodynamic average \( \langle T_\tau \ldots \rangle_{S0} \).

Let us denote by \( p_j \) the number of loops of the same order \( m_j \), where the index \( j \) enumerates all loops of different order. Then obviously \( n = p_1 m_1 + p_2 m_2 + \ldots \). Until the spin operators are not paired the analytical expression corresponding to each \( n \)-th order diagram can be represented as a product of some multipliers. Each multiplier is related to a single fermionic loop. Note that in the case of further application of the spin diagram
technique [9] there should be taken into account only such pairings of spin operators when all fermionic loops are connected graphically with elements of the spin diagram technique.

The fermion loop of the $m_j$-th order is formed due to $m_j$ operators $\hat{H}_{int}$. Let’s use the index ”$L$” in angle brackets $\langle \ldots \rangle_L$ to indicate the fact that when pairing all the $c$-operators in the $T_\tau$-ordered average of $m_j$ operators $\hat{H}_{int}$ there should be taken into account only one-loop diagrams of $m_j$-th order. Then the analytical contribution to the one loop of $m_j$-th order will be determined by the average: $\langle T_\tau \hat{H}_{int}(\tau_1) \ldots \hat{H}_{int}(\tau_m) \rangle_{L_0}$. Strictly speaking this average gives rise to $(m_j - 1)!$ equivalent fermionic loops of $m_j$-th order. Using the definition of the average $\langle T_\tau \ldots \rangle_{L_0}$ the expansion of $\langle \mathfrak{S}(\beta) \rangle_{c_0}$ can be written as:

$$\langle \mathfrak{S}(\beta) \rangle_{c_0} = T_\tau \sum_n \frac{(-1)^n}{n!} \sum_{p_1, p_2, \ldots \atop (p_1 m_1 + \ldots = n)} A(p_1 m_1; p_2 m_2; \ldots) \times$$

$$\times \prod_j \int d\tau^{(j)}_1 \ldots d\tau^{(j)}_{m_j} \langle T_\tau \hat{H}_{int}(\tau^{(j)}_1) \ldots \hat{H}_{int}(\tau^{(j)}_{m_j}) \rangle_{L_0}.$$  

In derivation the formula (10) in each $n$-th order of $\hat{H}_{int}$ the thermodynamic average $\langle T_\tau \ldots \rangle_{L_0}$ was represented as a sum of products of averages $\langle T_\tau \ldots \rangle_{L_0}$. The second sum in (10) takes into account all possible partitions of $n$ operators $\hat{H}_{int}$ into $p_1 + p_2 + \ldots$ groups. Each group corresponds to a single fermionic loop and the equality: $p_1 m_1 + p_2 m_2 + \ldots = n$ must be satisfied. The index $j$ in the product $\prod_j$ runs all groups (loops) at a fixed partition. Because the averaging in the $\langle \mathfrak{S}(\beta) \rangle_{c_0}$ is carried out only over the charge degrees of freedom, the remaining spin operators must be still ordered by the imaginary time. This justifies the presence of operator $T_\tau$ in the right side of (10). Combinatorial factor $A(p_1 m_1; p_2 m_2; \ldots)$ takes into account the equivalent contributions arising at each possible partitioning of $n$ operators $\hat{H}_{int}$ in groups.

It can be shown (cf. [8]) that:

$$A(p_1 m_1; p_2 m_2; \ldots) = \frac{n!}{p_1! (m_1)! p_2! (m_2)! p_3! \ldots}.$$  

Introducing notations:

$$\Xi_m = \frac{(-1)^{m+1}}{m!} \int_0^\beta d\tau_1 \ldots d\tau_m \langle T_\tau \hat{H}_{int}(\tau_1) \ldots \hat{H}_{int}(\tau_m) \rangle_{L_0},$$  

Figure 1: The $n$-th order diagrams, arising due to pairing of all $c$-operators in the expansion of $\langle \mathfrak{S}(\beta) \rangle_{c_0}$, can be represented as a set of fermionic loops. The order of each loop $m_j$ is determined by the number of vertices represented by points and the same number of lines with arrows denoting the fermion propagator. Each vertex corresponds to the spin operator. If the number of vertices of the same order $m_j$ is denoted by $p_j$, then: $p_1 m_1 + p_2 m_2 + \cdots = n.$
Figure 2: The effective action $\Xi$ can be expressed as an infinite series of terms $\Xi_n$. Each term $\Xi_n$ in the diagrammatic representation corresponds to a single loop of $n$-th order in the exchange coupling constant $J$ and describes the effective $n$-site spin-spin interaction. The lines with arrows represent bare fermion Green’s functions and each vertex, shown with a bold circle, corresponds to a spin operator $\tilde{S}$.

the expression (10) can be rewritten as:

$$\left\langle \mathcal{G}(\beta) \right\rangle_{c_0} \equiv \mathcal{G}_S(\beta) = T_{\tau} \sum_{p_1, p_2, \ldots} \frac{1}{p_1} (-\Xi_1)^{p_1} \frac{1}{p_2} (-\Xi_2)^{p_2} \ldots = T_{\tau} \exp \{-\Xi_1 - \Xi_2 - \ldots\} . \quad (13)$$

Using the explicit form of $\hat{H}_{int}$ carry out in (12) pairings of all the $c$-operators. Then introducing the variable $x_j = (\vec{R}_f, \tau_j)$ we obtain expression for the operator $\Xi_m$ in the Wannier representation:

$$\Xi_m = \frac{1}{m} \left( \frac{J}{2} \right)^m \int dx_1 \ldots dx_m \; G^{(0)}(x_1 - x_2) \ldots G^{(0)}(x_m - x_1) \; \text{Sp}\left\{ \tilde{S}(x_1) \ldots \tilde{S}(x_m) \right\}, \quad (14)$$

where the integral over $dx$ denotes the operation $\int_0^\beta d\tau \sum_{\sigma_f}$ and the trace is calculated over the Pauli matrix indices.

From the formula (13) it follows that the effective interactions in the localized spin subsystem are determined by the structure of the series:

$$\Xi = \sum_{m=1}^\infty \Xi_m . \quad (15)$$

The effective action $\Xi$ describes all possible multi-site spin-spin interactions in the localized spin subsystem in the arbitrarily order of the coupling constant $J$. The partial action $\Xi_m$ determines the $m$-th order interactions and diagrammatically can be represented as a loop with $m$ lines, corresponding to propagators $G^{(0)}$, and $m$ vertices related to spin operators $\tilde{S}$ (see figure 2). It can be seen that all effective interactions in $\Xi$ take into account retardation effects. This is due to imaginary time dependence of all the effective interactions in each term (14) of the series for $\Xi$.

4. Effective interactions of the 2nd, 3rd and 4th order in $J$

Let us consider the first several terms of the series for $\Xi$. The first term with $n = 1$ is zero, because $\text{Sp}\{\tilde{S}(x_1)\} = \sum_j \tilde{S}^j(x_1)\text{Sp}\{\sigma^j\}$, and $\text{Sp}\{\sigma^j\} = 0$ at any $j = x, y, z$. 

To calculate the operator $\Xi_2$ one should to derive the trace $\text{Sp} \left\{ \tilde{S}(x_1)\tilde{S}(x_2) \right\}$. Using the identity: $\sigma^i\sigma^j = \delta_{ij} + i\varepsilon_{ijl}\sigma^l$, where $\varepsilon_{ijl}$ – Levi-Civita antisymmetric tensor we find:

$$\text{Sp} \left\{ \tilde{S}(x_1)\tilde{S}(x_2) \right\} = 2 \left( \tilde{S}(x_1)\tilde{S}(x_2) \right). \quad (16)$$

Then the operator $\Xi_2$ takes the form:

$$\Xi_2 = \int \! dx_1 dx_2 \, V_2(x_1 - x_2) \, \tilde{S}(x_1)\tilde{S}(x_2), \quad (17)$$

where the effective interaction between spins is defined via a polarization loop (see figure 3):

$$V_2(x_1 - x_2) = \left( \frac{J}{2} \right)^2 G^{(0)}(x_1 - x_2)G^{(0)}(x_2 - x_1). \quad (18)$$

Figure 3: The action $\Xi_2$ can be represented by a loop of the second order in the exchange coupling constant $J$.

From equation (17) it follows, that at $f_1 \neq f_2$ the second order effective action $\Xi_2$ describes indirect exchange interaction of two localized spins through the subsystem of itinerant electrons. However, in contrast to the usual RKKY-interaction here the retardation effects, caused by $\tau$-dependence of $V_2$, are taken into account. Note that in the $\Xi_2$ there is also a term with $f_1 = f_2$. The Fourier transform of the indirect exchange interaction (18) has the form:

$$V_2(k, i\omega_m) = \left( \frac{J}{2} \right)^2 \chi_0(k, i\omega_m), \quad (19)$$

where

$$\chi_0(k, i\omega_m) = \frac{1}{N} \sum_q \frac{f_q - f_{q+k}}{i\omega_m + \varepsilon_q - \varepsilon_{q+k}} \quad (20)$$

is the Lindhard susceptibility in the Matsubara representation, $\omega_m = 2m\pi T$ with $m \in \mathbb{Z}$, and $f_q = (\exp \{ (\varepsilon_q - \mu) / T \} + 1)^{-1}$ is the Fermi-Dirac distribution function. The intensity of the exchange interaction is largely determined by the properties of the itinerant subsystem. The well known RKKY-interaction follows from (20) at $\omega_m \equiv 0$.

Calculating the 3rd order effective action $\Xi_3$ we obtain (see also figure 4):

$$\Xi_3 = \int \! dx_1 dx_2 dx_3 \, V_3(x_1, x_2, x_3) \, \tilde{S}(x_1) \cdot (\tilde{S}(x_2) \times \tilde{S}(x_3)), \quad (21)$$

where
The formula (21) describes the three-spin interactions in the form of a mixed product of three spin operators. This type of interaction was first considered in [12] but without taking into account the retardation effects.

It is evident that the interaction (21) favors the chiral order in the magnetic subsystem. In this regard we note that in the paper [13] the third order corrections to the Hall conductivity due to s-d(f) exchange interaction were shown to give rise the anomalous Hall effect provided that non trivial spin configuration (chirality) is formed in the spin subsystem. Interestingly the structure of the expression for the Hall conductivity obtained in [13] is similar to that of (21).

The operator $\Xi^4$, which determines the effective spin-spin interactions in the fourth order in the coupling constant $J$, after calculating the trace $\text{Sp}\{\tilde{S}(x_1)\tilde{S}(x_2)\tilde{S}(x_3)\tilde{S}(x_4)\}$ takes the form (see also figure 5):

$$\Xi_4 = \int dx_1 dx_2 dx_3 dx_4 \ V_4(x_1, x_2, x_3, x_4) \ \left(\tilde{S}(x_1)\tilde{S}(x_2)\right) \cdot \left(\tilde{S}(x_3)\tilde{S}(x_4)\right),$$  

(23)

where

$$V_4(x_1, x_2, x_3, x_4) = \frac{1}{2} \left(\frac{J}{2}\right)^4 \left[ G^{(0)}(x_1 - x_2)G^{(0)}(x_2 - x_3)G^{(0)}(x_3 - x_4)G^{(0)}(x_4 - x_1) - \
- G^{(0)}(x_1 - x_3)G^{(0)}(x_3 - x_2)G^{(0)}(x_2 - x_4)G^{(0)}(x_4 - x_1) + \
+ G^{(0)}(x_1 - x_4)G^{(0)}(x_4 - x_3)G^{(0)}(x_3 - x_2)G^{(0)}(x_2 - x_1) \right].$$  

(24)

The terms of the expression (23) with unequal indices of sites $f_{j}$ ($j = 1, \ldots, 4$) correspond to the four-spin exchange interactions. Among them there are, in particular, interactions describing ring exchange of four spins located, for example, at the square plaquette vertices. For the first time the ring exchange (without retardation effects) was obtained from the
Hubbard model at half filling in the fourth order of the perturbation theory in the parameter $t/U$, where $t$ is the tunneling integral, and $U$ is the Coulomb repulsion energy of two electrons at the same site [14]. The four-spin ring exchange interaction was involved to explain the magnetic ordering features in the quantum crystal $^3\text{He}$ [15]. In the paper [16] it was argued that ring exchange is important to describe magnetic properties of cuprate high-temperature superconductors. Effect of ring exchange interaction on the superconductivity in cuprates was investigated in [17].

At pairwise coincident site indices: $f_3 = f_1$ and $f_4 = f_2$, in the sum [28] there are terms that are responsible for biquadratic exchange interaction. These interactions were first used in [18] for explaining the paramagnetic resonance on Mn ions in the compound MnO. Besides, the biquadratic exchange interaction is essential in multilayer magnetic systems [19].

5. Conclusion

The paper presents a method of deriving all possible kinds of effective indirect interactions between the localized spins due to s-d(f) exchange coupling of these spins with the subsystem of itinerant electrons. After integrating over charge degrees of freedom in the s-d(f) exchange model an exact representation for an action of a purely spin model is obtained. Using this action allows to study the spin subsystem in the s-d(f) model in the framework of effective purely spin model. The important point of this model is that all effective interactions take into account retardation effects. Although explicit expressions are written only for two-, three- and four-spin interactions, the formula (14) allows to generate multi-site spin-spin interactions in the arbitrary order in the s-d(f) exchange coupling constant $J$.

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