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Current status of the Kondo problem

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Contents

Abstract. It is shown that at zero temperature the magnetic field $\mu H \gg T_K$ does not move the system from the strong coupling to the weak coupling regime. As a result, the average of the impurity spin approaches its saturation value as a power of the small parameter $(2T_K/\mu H)^2$. The study of the high-temperature expansion of the free energy shows that the Kondo problem contains at least two energy scales and that these scales are separated by the coupling constant. The Hamiltonian of the Kondo problem is not renormalizable.

1. Introduction

It was observed in 1934 by de Haas et al. [1] that a small concentration of magnetic impurities in a metal leads to a minimum in the temperature dependence of the resistance. Taking into account scattering by nonmagnetic and magnetic impurities in Born approximation calculations gives rise to a temperature-independent residual resistance at low temperatures [2]. In higher-order perturbation theory, introducing scattering by ordinary impurities amounts simply to replacing the Born amplitude by the exact amplitude and so does not lead to any new phenomena.

In 1964 Kondo considered scattering by magnetic impurities in the weak-coupling approximation and showed that taking one step beyond the Born approximation yields a scattering amplitude which varies with energy as $\ln(\varepsilon_F/\varepsilon)$, where ε_F is the Fermi energy and ε is the particle's scattering energy measured from the Fermi surface [3]. Consequently, as the temperature decreases, the contribution to the resistance shows a logarithmic increase, due to the magnetic impurity scattering [3, 4]. Scattering by real excitations in a metal yields an increase in resistance with temperature. For example, scattering by phonons leads to a momentum relaxation with a characteristic time $\tau^{-1}_{\text{ph.mom.}} \sim T^5/\omega_D^4$, with ω_D the Debye temperature. The competition between the two processes produces a resistance minimum at temperatures whose values depend on the impurity concentration and which in practical situations lie very far away from T_K, the characteristic temperature at which the small scattering-amplitude correction becomes large and the weak coupling turns into strong one. The magnetic susceptibility of the electron gas in a metal is low. The effective magnetic moment per particle M_{eff} is [5]

$$M_{\text{eff}} = \frac{3}{8}\,\frac{\mu^2 H}{\varepsilon_F}\,, \tag{1}$$

where μ is the Bohr magneton and H is the magnetic field. This circumstance makes measuring impurity magnetism possible at low impurity concentrations.

In the absence of interaction, a localized electronic state is doubly degenerate in spin. A magnetic field removes the degeneracy and produces a level splitting of magnitude $\pm\mu H/2$. Experimental data indicate, however, that a localized electronic state is completely depolarized at $T = 0$ and that the magnetic moment is proportional to the field provided the field is weak enough [6]. This means that at $T = 0$ the interaction is strong and that it lifts degeneracy even in zero magnetic field. It is more or less obvious that at the $H = 0$, $T = 0$, the splitting of the level is on the order of the Kondo temperature.

The Kondo effect results from the exchange interaction between an electron localized on an impurity and electrons in the conduction band. It is usually assumed that an impurity-localized state contains only one electron. Because of the repulsion effect, placing a second electron in a localized state causes a large increase in energy, and such states may therefore be neglected. The Hamiltonian $\hat{H}$ of the system can then be taken in the form

$$\hat{H} = \hat{H}_0 + \int \mathrm{d}^3 r_1\, \mathrm{d}^3 r_2 V(r_1 - r_2)\chi^+_\alpha(r_1)\varphi^+_\beta(r_2)\chi_\beta(r_2)\varphi_\alpha(r_1)$$

$$- \frac{\mu H}{2}\int \mathrm{d}^3 r_1 \left(\varphi^+_\uparrow(r_1)\varphi_\uparrow(r_1) - \varphi^+_\downarrow(r_1)\varphi_\downarrow(r_1)\right), \tag{2}$$

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where φ_β^+ and χ_α^+ are the electron creation operators for an impurity-localized state and for the continuous spectrum, respectively. The set of eigenfunctions $\{\chi, \varphi\}$ for the potential of the impurity is a complete one. The Hamiltonian $\hat{H}_0$ is that for noninteracting particles in the impurity potential.

The Hamiltonian $\hat{H}_0$ conserves the total spin of the system. The number of particles in the state φ is also conserved. The wave function $|\psi\rangle$ is defined over a Fock space. At zero temperature, the corresponding equation for the ground-state wave function $|\psi\rangle$ is

$$|\hat{H}\psi\rangle = E\,|\psi\rangle\,. \tag{3}$$

It is important for our discussion that the electrons polarized parallel and antiparallel to the field have the same chemical potential, meaning that there is no gap on the Fermi surface. Equation (1) is a consequence of this statement. In the Appendix, an expansion of the ground-state function $|\psi\rangle$ in terms of the basis states of noninteracting particles is given. The subspace in which the function $|\psi\rangle$ is looked for is invariant with respect to the Hamiltonian (2), and hence Eqn (3) does have a solution of the form (A.1).

2. The average spin $\langle S_z\rangle$ as a function of magnetic field at zero temperature

Studies using perturbation theory [4, 7–9] show that the Kondo problem involves a characteristic energy (temperature T_K), which is determined from the equation

$$Z = |g|\ln\left(\frac{\varepsilon_\mathrm{F}}{T_\mathrm{K}}\right), \qquad 1 - 2Z = 0\,, \tag{4}$$

where $|g|$ is the dimensionless coupling constant (see the Appendix). Actually, the value of the parameter Z is the solution of the equation

$$f(Z) = 0 \tag{5}$$

and the second of equations (4) corresponds to the first two terms in the Taylor series expansion of the function $f(Z)$. In Ref. [10], the third term is also found:

$$f(Z) = 1 - 2Z + \frac{Z^2}{3}\,. \tag{6}$$

The solution of Eqn (5) with $f(Z)$ from Eqn (6) is

$$Z = 3 - \sqrt{6} = 0.5505\ldots \tag{7}$$

It is assumed that Eqn (5) has a solution such that $Z > 0$.

Subsequent efforts at studying this problem concentrated on impurity states under tight-binding conditions [7–9]. In Refs [8, 9], the Bethe ansatz was used to calculate the average spin of the impurity $\langle S_z\rangle$ as a function of magnetic field at zero temperature. It was found that in strong fields such that $\mu H \ll T_\mathrm{K}$, the quantity $\langle S_z\rangle$ scales with the field, whereas in strong fields $\mu H \gg T_\mathrm{K}$, the average spin is given by

$$\langle S_z\rangle = \frac{1}{2}\left[1 - \frac{1}{2\ln(\mu H/T_\mathrm{K})}\right]. \tag{8}$$

From Eqn (8) it follows that in a strong field the average spin approaches its saturation value very slowly — as $\ln^{-1}(\mu H/T_\mathrm{K})$. This behaviour is in clear contradiction with the experimental data [6]. To achieve the spin saturation level obtained in Ref. [6] with 6 T, a magnetic field on the order of 100 T is required according to formula (8).

At a temperature $T = 0$, the average value of the spin is expressed in terms of the ground-state energy by

$$\langle S_z\rangle = -\frac{\partial E}{\partial \mu H}\,. \tag{9}$$

From Eqns (8) and (9), we find the ground-state energy in strong fields $\mu H \gg T_\mathrm{K}$ to be

$$\delta E = \frac{\mu H}{4\ln(\mu H/T_\mathrm{K})}\,. \tag{10}$$

The characteristic energy scale defined by this formula for the case of strong fields is $|g|\varepsilon_\mathrm{F}$, where g is the dimensionless coupling constant in the Kondo model. This energy scale is too large to be correct.

In applying the Bethe ansatz, two hypotheses were used:

1. In changing from the momentum representation to the coordinate representation, the replacement $\sin x/x \to \pi\delta(x)$ was made.

2. Boundary conditions in a magnetic field were derived by matching the bulk susceptibility (1) to the corresponding Bethe-ansatz result.

Note, however, that expression (1) depends on the system's spectrum over the entire energy range up to ε_F and that it is obtained from the chemical potential equality for particles with spins parallel and antiparallel to the field, respectively. This condition acts to redistribute electrons in these two spin states while at the same time conserving their total number.

In the Kondo problem, only those electrons at a distance $\pm D$ from the Fermi surface are important. Here, D is the characteristic energy scale within which an impurity interacts effectively with the conduction electrons, and it is no restriction to assume in the Kondo model that D is independent of ε_F and, in particular, that $D \ll \varepsilon_\mathrm{D}$. Besides, the fact that free electrons with spins $\pm 1/2$ differ in density is not used in the model Hamiltonian and does not manifest itself in any order of perturbation theory, nor in the set of an exact equations for the ground-state energy [10]. This implies that the quantity (1) does not appear in the Kondo problem, and that the arguments of Refs [8, 9] about the magnetic-field dependence of the Bethe ansatz interval, though plausible, are incorrect.

In a previous paper by the present authors [10], an exact set of equations is derived for the ground state of a Kondo problem whose Hamiltonian (denoted there by $\hat{H}$) includes a magnetic impurity, noninteracting conduction electrons, and exchange interaction between the impurity and the electrons (see the Appendix).

The wave function $|\psi\rangle$ may be written as a sum of two terms

$$|\psi\rangle = |\psi\rangle_\uparrow + |\psi\rangle_\downarrow\,. \tag{11}$$

the first and second term representing the states with a localized electron polarized parallel and antiparallel to the field, respectively.

According to the general rules of quantum mechanics, the expectation value of the spin of an 'impurity' electron can be

represented in the form

$$\langle S_z\rangle = \frac{1}{2}\frac{\big||\psi\rangle_\uparrow\big|^2 - \big||\psi\rangle_\downarrow\big|^2}{\big||\psi\rangle_\uparrow\big|^2 + \big||\psi\rangle_\downarrow\big|^2}\,, \tag{12}$$

the symbol $\big||\psi\rangle\big|$ denoting the norm of the wave function. It is convenient to separate large terms irrelevant to the Kondo problem from the energy E by writing

$$E = E_0 - \frac{\mu H}{2} + \delta E^{(1)} + \delta E\,, \tag{13}$$

where E_0 is the energy of the noninteracting system, and $\delta E^{(1)}$ is the trivial ground-state energy shift, proportional to the interaction potential averaged over conduction electron states. One of the basic assumptions of Refs [8, 9] is that a sufficiently strong magnetic field $\mu H \gg T_K$ changes the system from a strong-coupling state to the weak-coupling — perturbative — region. Using the system of equations proposed in Ref. [10], the ground-state energy can be evaluated by perturbation theory. There are two types of terms which arise in this approach. In type 1 terms, the magnetic field leads to the logarithmic singularity being cut off at the magnetic field value at low energies. In type 2 terms, the magnetic field fails to remove the singularity (see the Appendix). In Refs [8, 9], terms of this type are considered irrelevant for the calculation of the average spin $\langle S_z\rangle$ and are omitted. These terms are significant, however: in the fourth order of perturbation theory they lead to a difference between the $\langle S_z\rangle$ values calculated from Eqns (9) and (12). In expression (12) for the average spin, logarithmic cross terms absent from Eqn (9) appear in fourth order calculations, meaning that, in its simplest form, perturbation theory is not even applicable at strong magnetic fields — indeed a magnetic field $\mu H \ll \varepsilon_F$ fails to take the system out of its strong-coupling state.

A method developed in Ref. [10] allows a nonperturbative solution to Eqn (3). The average spin is found to be

$$\langle S_z\rangle = \frac{\mu\tilde{H}}{4(T_K^2 + (\mu\tilde{H}/2)^2)^{1/2}}\,, \tag{14}$$

where the effective field $\tilde{H}$ and the external field H are related by the expression

$$\mu\tilde{H} = \mu H\left[1 + Z\left(-\frac{1}{2} + \langle S_z\rangle\right)\right]. \tag{15}$$

The quantities Z and T_K in formula (15) are defined by Eqns (4) and (5).

The solution of Ref. [10] can be used as a trial function for obtaining an upper bound variationally for the ground-state energy. The result is

$$\delta E = -\Delta\,, \tag{16}$$

where Δ satisfies the equation

$$\Delta(\mu\tilde{H} + \Delta) = T_K^2\,. \tag{17}$$

Eqn (17) has two solutions. One of them,

$$\Delta = -\frac{\mu\tilde{H}}{2} + \sqrt{T_K^2 + \left(\frac{\mu\tilde{H}}{2}\right)^2}\,, \tag{18}$$

determines the ground-state energy, whereas the second,

$$\Delta = -\frac{\mu\tilde{H}}{2} - \sqrt{T_K^2 + \left(\frac{\mu\tilde{H}}{2}\right)^2}\,, \tag{19}$$

gives the splitting energy δE_g:

$$\delta E_g = 2\sqrt{T_K^2 + \left(\frac{\mu\tilde{H}}{2}\right)^2}\,. \tag{20}$$

The energy determined by Eqns (16), (18) lies below the value (10) obtained in Refs [8, 9], suggesting that at least one of the hypotheses used in these papers is incorrect. In our opinion, at least two hypotheses are incorrect:

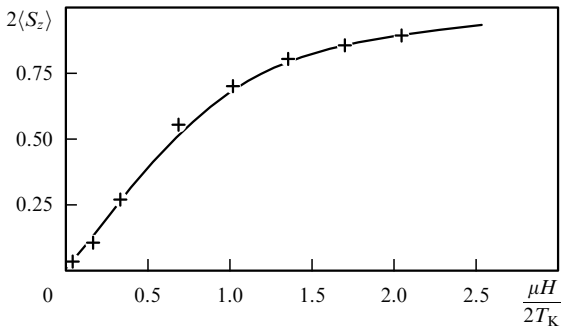
(1) the weak binding assumption in the region $\mu H \gg T_K$; and, more importantly,

(2) the way in which the length of the interval 'B' is taken to vary with the magnetic field [8, 9].

The dependence of $\langle S_z\rangle$ on the parameter $\mu H/2T_K$ is shown in the accompanying figure. From Eqns (14) and (15), it follows that the point where $\mu H/2T_K = 0.24$ is an inflection point for the function $\langle S_z\rangle$. In the region $\mu H \gg T_K$, we find

$$\langle S_z\rangle = \frac{1}{2}\left[1 - \frac{1}{2}\left(\frac{2T_K}{\mu H}\right)^2\right]. \tag{21}$$

The graph of the experimental data of Ref. [6] clearly reveals the presence of an inflection point and is in excellent agreement with the dependence given by formulas (14) and (15).



Magnetic field dependence of the average spin $\langle S_z\rangle$. Experimental data points are from Ref. [6].

3. Two energy scales and the violation of renormalizability in the Kondo problem

Two assumptions usually adopted in treating the Kondo problem are that the problem has only one energy scale and that it is amenable to renormalization. At zero temperature, these assumptions imply that the free energy associated with the presence of a magnetic impurity can be written in the form

$$\mathcal{F} = D\sum_n g^n C_n + T_K f\left(\frac{\mu H}{T_K}, \frac{T}{T_K}\right), \tag{22}$$

where the C_n are numerical coefficients.

The first term in this formula is not universal and is considered as a shift common to all the energy levels of the system. Consequently, at $T = 0$ the ground-state energy may

be represented by

$$\delta E = D\sum_n g^n \tilde{C}_n + T_K \tilde{f}\left(\frac{\mu H}{T_K}\right). \tag{23}$$

The renormalizability and single-scale assumptions on the Kondo problem suggest that the equalities

$$C_n = \tilde{C}_n. \tag{24}$$

should hold. Also, in calculating the function f, in no order of perturbation theory should there appear terms proportional to ε_F. The verification of these hypotheses is complicated by the fact that no simple perturbation theory scheme is currently available for calculating the free energy directly. To obtain the free energy, it is necessary to calculate the partition function Z. This function can be represented as a perturbation series in the interaction potential $V(r)$ between the impurity electron and the conduction electrons [2]:

$$\begin{aligned} Z &= \mathrm{Tr}\,\exp\left(-\frac{\hat{H}}{T}\right) \\ &= \mathrm{Tr}\left\{\exp\left(-\frac{\hat{H}_0}{T}\right)\left[1-\int_0^{1/T} \mathrm{d}\tau_1 \hat{V}(\tau_1)+\right.\right. \\ &\left.\left.+\int_0^{1/T}\mathrm{d}\tau_1\int_0^{\tau_1}\mathrm{d}\tau_2\hat{V}(\tau_1)\hat{V}(\tau_2)-\ldots\right]\right\}, \end{aligned} \tag{25}$$

where the operator $\hat{V}(\tau)$ is defined by the expression

$$\hat{V}(\tau) = \exp(\hat{H}_0\tau)V_{\mathrm{int}}(r)\exp(-\hat{H}_0\tau). \tag{26}$$

The free energy $\mathcal{F}$ is expressed in terms of the partition function by

$$\mathcal{F} = -T\ln Z. \tag{27}$$

This approach is inconvenient in that higher-order perturbation terms in Eqn (25) contain various powers of the large parameter (ε_F/T), requiring a method by which the exact cancellation of such terms in the free energy expression (27) could be detected. Such a method has been devised and outlined in Refs [11, 12]. What complicates the problem is that there is no Wick's theorem for the mean of the products of the operators φ, φ^+ at various times. To calculate each one of such correlators is not a problem. The problem is that there is no simple rule for the calculation and counting of all possible means in Eqn (25). The existence in the Kondo model of scales other than T_K turns out to be a simpler problem.

In Ref. [10], the ground-state energy correction of fourth order in the coupling constant g is given, thus enabling one to calculate the coefficients $\tilde{C}_2$, $\tilde{C}_3$, $\tilde{C}_4$. The coefficients C_2, C_3, C_4 were obtained in Refs [11, 12]. It turned out that only the first two of them were identical: $C_2 = \tilde{C}_2$, $C_3 = \tilde{C}_3$. The coefficients C_4 and $\tilde{C}_4$ are not equal, suggesting that there exists another energy scale besides T_K. This second scale, T_0, is proportional to the cut-off energy (D, ε_F) and to a certain power of the coupling constant g. Another consequence of this is the appearance, in the high-temperature expansion of the free energy, of terms which depend on temperature and are proportional to a higher-than-first power of the cutoff energy.

Because the problem arises only in fourth-order terms in the interaction, we do not find it possible even to outline in brief — nor to demonstrate — the method for the exact cancellation of large terms in the free-energy expression (12). By using the method developed in Ref [12], it proved possible to show that up to and including the sixth order in g, the free energy contains no anomalous terms and can be represented as a sum of two terms, one of which is proportional to ε_F and is temperature and magnetic-field independent. The second term contains ε_F only as a factor under the logarithm sign. In the eighth order of perturbation theory, however, a term of the form [12]

$$\delta\mathcal{F}_{\mathrm{an}} = \mathrm{const}\ \frac{g^8\varepsilon_F^2}{T} \tag{28}$$

appears [28], indicating that the Hamiltonian of the Kondo problem is nonrenormalizable. The second scale involved in the problem can be established by comparing the fourth-order result for the specific heat with what the anomalous term (28) contributes to this property. This comparison shows that, in order of magnitude, the second scale is given by the expression

$$T_0 \approx g^2\varepsilon_F. \tag{29}$$

The total depolarization of spin at $T = 0$ indicates that the bare term in the expansion of the function $|\psi\rangle$ in the basis set of noninteracting systems contributes little to the norm of a state for $\mu H \ll T_K$. This allows one, in principle, to improve the estimate of T_0.

4. Conclusions

We have shown that at $T = 0$, for the Kondo problem with $g < 0$, a strong coupling exists in magnetic fields $\mu H \ll \varepsilon_F$. Perturbation theory turns out to be inapplicable even in the region of relatively strong fields, $\mu H \gg T_K$. As a result, the spin of a localized electron approaches its saturation value as a power of the parameter $2T_K/\mu H \ll 1$ [see Eqn (21)].

The magnetic field dependence of the average spin has an inflection point at $\mu H \approx 0.24T_K$. This agrees well with the experimental data.

The high-temperature expansion of the free energy reveals that the Kondo model involves at least two energy scales, T_K and T_0, of which the former is exponentially small in the inverse coupling constant, whereas the latter is only small as a power. In the region $T \gg T_0$, the free energy is calculable using perturbation theory. The point T_0 apparently marks the crossover from weak coupling to strong coupling. The presence in the free energy of terms of the form

$$\frac{g^k\varepsilon_F^{l+1}}{T^l}$$

indicates that the Hamiltonian of the Kondo problem is nonrenormalizable.

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5. Appendix

The function $|\psi\rangle$ of the ground state of the Hamiltonian (2) is sought in the form [10]

$$
\begin{aligned}
|\psi\rangle = {} & |10;11;11;\ldots\rangle + \sum C_{2K}^{2L-1} \Big| 01; \overset{2K}{10}; \overset{2L-1}{10} \Big\rangle \\
& + \sum C_{2K-1}^{2L-1} \Big| 10; \overset{2K-1}{01}; \overset{2L-1}{10} \Big\rangle + \sum C_{2K}^{2L} \Big| 10; \overset{2K}{10}; \overset{2L}{01} \Big\rangle \\
& + \sum_{K_1<K} C_{2K_1\,2K}^{2L_1\,2L-1} \hat{N} \Big| 01; \overset{2K_1}{10}; \overset{2K}{10}; \overset{2L_1}{01}; \overset{2L-1}{10} \Big\rangle \\
& + \sum C_{2K_1-1;2K}^{2L_1;2L-1} \hat{N} \Big| 10; \overset{2K_1-1}{01}; \overset{2K}{10}; \overset{2L_1}{01}; \overset{2L-1}{10} \Big\rangle \\
& + \sum_{L_1<L} C_{2K_1;2K-1}^{2L_1-1;2L-1} \hat{N} \Big| 01; \overset{2K_1}{10}; \overset{2K-1}{01}; \overset{2L_1-1}{10}; \overset{2L-1}{10} \Big\rangle \\
& + \sum_{K_1<K;\,L_1<L} C_{2K_1-1;2K-1}^{2L_1-1;2L-1} \hat{N} \Big| 10; \overset{2K_1-1}{01}; \overset{2K-1}{01}; \overset{2L_1-1}{10}; \overset{2L-1}{10} \Big\rangle \\
& + \sum_{K_1<K;\,L_1<L} C_{2K_1;2K}^{2L_1;2L} \hat{N} \Big| 10; \overset{2K_1}{10}; \overset{2K}{10}; \overset{2L_1}{01}; \overset{2L}{01} \Big\rangle + \ldots \qquad \text{(A.1)}
\end{aligned}
$$

Each position inside $|;\ldots;\rangle$ indicates a state with a definite energy and a spin parallel (first number) or antiparallel (second number) to the field. The number 1 indicates that the state is occupied, 0 denotes an empty state. The first position is for the impurity-localized electron state. Indices K, L designate states below and above the Fermi surface. $\hat{N}$ is an ordering operator, with every interchange of two neighbouring filled states giving a factor of (-1). The first term in Eqn (A.1) is the bare term; in the absence of interaction, it corresponds to the ground state of the system. The Hamiltonian (2) couples only those states with the number of upper (low) indices differing by no more than unity.

The energy δE in Eqn (13) is expressed in terms of the coefficients C as

$$
\delta E = \sum \left[\left(I_{2K-1}^{2L-1} \right)^* C_{2K-1}^{2L-1} - \left(I_{2K}^{2L-1} \right)^* C_{2K}^{2L-1} \right]. \qquad \text{(A.2)}
$$

The quantities I in Eqn (A.2) are equal to the matrix elements of the potential V. An example is

$$
I_{2K}^{2L-1} = \int \mathrm{d}^3 r_1\, \mathrm{d}^3 r_2 \chi_\uparrow^*(r_1)\, \varphi_\downarrow^*(r_2)\, \varphi_\uparrow(r_1)\, \chi_\downarrow(r_2)\, V(r_1 - r_2)\,. \qquad \text{(A.3)}
$$

The dimensionless coupling constant g is defined by the relation

$$
\begin{aligned}
& \sum_K I_{2K}(\ldots) \to g \int_0^{\varepsilon_\mathrm{F}} \mathrm{d}x(\ldots)\,, \\
& \sum_L I^{2L}(\ldots) \to g \int_0^{\varepsilon_\mathrm{F}} \mathrm{d}y(\ldots)\,, \qquad \text{(A.4)}
\end{aligned}
$$

where

$$
\varepsilon_L - \varepsilon_\mathrm{F} = y\,; \qquad \varepsilon_\mathrm{F} - \varepsilon_K = x\,.
$$

Equation (3) reduces to an infinite set of equations for the coefficients C:. Eqn (A.2) is the first of them. The next three equations are as follows [10]:

$$
\begin{aligned}
& -I_{2K}^{2L-1} + \sum C_{2K_1}^{2L-1} I_{2K}^{2K_1} - \sum C_{2K_1-1}^{2L-1} I_{2K}^{2K_1-1} \\
& \quad - \sum C_{2K}^{2L_1} I_{2L_1}^{2L-1} + (\mu H + \varepsilon_L - \varepsilon_K - \delta E) C_{2K}^{2L-1} \\
& \quad + \sum_{K_1<K} C_{2K_1;2K}^{2L_1;2L-1} I_{2L_1}^{2K_1} - \sum_{K<K_1} C_{2K;2K_1}^{2L_1;2L-1} I_{2L_1}^{2K_1} \\
& \quad - \sum C_{2K_1-1;2K}^{2L_1;2L-1} I_{2L_1}^{2K_1-1} = 0\,; \qquad \text{(A.5)} \\
& I_{2K-1}^{2L-1} - \sum I_{2K-1}^{2K_1} C_{2K_1}^{2L-1} + \sum C_{2K_1-1}^{2L-1} I_{2K-1}^{2K_1-1} \\
& \quad - \sum I_{2L_1-1}^{2L-1} C_{2K-1}^{2L_1-1} + (\varepsilon_L - \varepsilon_K - \delta E) C_{2K-1}^{2L-1} \\
& \quad + \sum_{L_1<L} C_{2K_1;2K-1}^{2L_1-1;2L-1} I_{2L_1-1}^{2K_1} - \sum_{L<L_1} C_{2K_1;2K-1}^{2L-1;2L_1-1} I_{2L_1-1}^{2K_1} \\
& \quad + \sum_{K<K_1;\,L_1<L} C_{2K-1;2K_1-1}^{2L_1-1;2L-1} I_{2L_1-1}^{2K_1-1} \\
& \quad - \sum_{K_1<K;\,L_1<L} C_{2K_1-1;2K-1}^{2L_1-1;2L-1} I_{2L_1-1}^{2K_1-1} \\
& \quad - \sum_{L<L_1;\,K<K_1} C_{2K-1;2K_1-1}^{2L-1;2L_1-1} I_{2L_1-1}^{2K_1-1} \\
& \quad + \sum_{K_1<K;\,L<L_1} C_{2K_1-1;2K-1}^{2L-1;2L_1-1} I_{2L_1-1}^{2K_1-1} = 0\,; \\
& - \sum I_{2L_1-1}^{2L} C_{2K}^{2L_1-1} + (\varepsilon_L - \varepsilon_K - \delta E) C_{2K}^{2L} \\
& \quad + \sum_{K<K_1} C_{2K;2K_1}^{2L;2L_1-1} I_{2L_1-1}^{2K_1} - \sum_{K_1<K} C_{2K_1;2K}^{2L;2L_1-1} I_{2L_1-1}^{2K_1} \\
& \quad + \sum C_{2K_1-1;2K}^{2L;2L_1-1} I_{2L_1-1}^{2K_1-1} = 0\,.
\end{aligned}
$$

From the system (A.5) we find that to the first order of perturbation theory the coefficients C: are

$$
C_{2K}^{2L-1} = \frac{I_{2K}^{2L-1}}{\varepsilon_L - \varepsilon_K + \mu H}\,; \qquad C_{2K-1}^{2L-1} = -\frac{I_{2K-1}^{2L-1}}{\varepsilon_L - \varepsilon_K}\,. \qquad \text{(A.6)}
$$

From Eqns (A.2), (9), (12), and (A.6) it follows that terms of two types are present both in expression (A.2) for the ground-state energy and in expression (12) for the average spin of an impurity electron.

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