Dynamical coherent potential approximation and quantum effects in diluted magnetic semiconductors

Masao Takahashi
Kanagawa Institute of Technology
E-mail:taka@gen.kanagawa-it.ac.jp

Abstract

A procedure for the calculation of the dynamical coherent approximation (dynamical CPA) is discussed. In the case of classical localized spins the density of states is symmetrical about the energy center $\omega = 0$. In the case of a finite magnitude of localized spins, it is not symmetrical due to the effect of quantum.

Key Word: diluted magnetic semiconductor (DMS) coherent potential approximation (CPA)

1 Introduction

In the series of our works applying the dynamical coherent approximation (dynamical CPA) to a simple model, we have studied the carrier state in diluted magnetic semiconductors (DMS). In these works classical spins are assumed for localized spins for simplicity [1–7]. The localized spins, however, have a magnitude of $S = 5/2$ for Mn ions. The effect of the finite magnitude of localized spins, therefore, needs to be investigated. Hence, in this report, we discuss a procedure for applying the dynamical CPA to the simple model with finite magnitude of localized spins.

2 Model Hamiltonian

In this report we consider a model that a carrier moves in a randomly distributed potential. We assume E_A for the potential on a non-magnetic site A, while we assume $E_M - I\boldsymbol{\sigma}\cdot\mathbf{S}_n$ for the potential on a magnetic site M. Here, $\mathbf{S}_n$ is the operator of the localized spin of a magnitude of S at a magnetic ion. The symbol $\boldsymbol{\sigma}$ is the Pauli matrix for the carrier, and I is the exchange interaction coupling constant between the carrier and the localized spin. The other notations used here have the usual meaning. We assume that the potential is randomly distributed. Thus, the Hamiltonian for the model is represented by

$$H = H_0 + \sum_n u_n = H_0 + U \; . \tag{1}$$

H_0 is a suitably chosen periodic Hamiltonian which is assumed to be known, and U is the total single particle potential, expressed as a sum of potentials u_n contributed by each site n. u_n takes u_n^A or u_n^M if the site n is occupied, respectively, by an A or M atom. U, and hence H, are configuration dependent.

$$H_0 = \sum_{k\mu} \varepsilon_k a_{k\mu}^\dagger a_{k\mu} = \frac{1}{N}\sum_{mn\mu} \varepsilon_{mn} a_{m\mu}^\dagger a_{n\mu} \tag{2}$$

$$u_n^A = \sum_{\mu} E_A a_{n\mu}^\dagger a_{n\mu} \tag{3}$$

$$u_n^M = \sum_{\mu} E_M a_{n\mu}^\dagger a_{n\mu} - I\sum_{\mu\nu} a_{n\mu}^\dagger \boldsymbol{\sigma}\cdot\mathbf{S}_n a_{n\nu}. \tag{4}$$

Hereafter we set $E_A = 0$.

3 Model density of states

Throughout this work we assume a model density of states with a semicircular profile whose half-bandwidth is Δ,

$$\rho(\varepsilon) = \frac{2}{\pi\Delta}\sqrt{1-\left(\frac{\varepsilon}{\Delta}\right)^2} \tag{5}$$

as the unperturbed density of states. Then, the diagonal component of the Green function is calculated as

$$F_\mu(\omega) \equiv \int_{-\Delta}^{\Delta} d\varepsilon \frac{\rho(\varepsilon)}{\omega - \varepsilon - \Sigma_\mu(\omega)} \quad (6)$$

$$= \frac{2}{\Delta}\left\{\left(\frac{\omega - \Sigma_\mu}{\Delta}\right) - \sqrt{\left(\frac{\omega - \Sigma_\mu}{\Delta}\right)^2 - 1}\right\}. \quad (7)$$

The equation leads to

$$\frac{\Sigma_\mu}{\Delta} = \frac{\omega}{\Delta} - \frac{1}{F_\mu \Delta} - \frac{F_\mu \Delta}{4} \quad (8)$$

where we set $F_\mu \equiv F_\mu(\omega)$ and $\Sigma_\mu \equiv \Sigma_\mu(\omega)$ for simplicity. Here, Σ_μ is the coherent potential with the spin of μ ($\mu =\uparrow$ or $\downarrow$). The density of states with μ spin $D_\mu(\omega)$ is calculated as

$$D_\mu(\omega) = -\frac{1}{\pi}\mathrm{Im}F_\mu(\omega) \quad (9)$$

4 Calculation for completely ferromagnetic case

4.1 Carrier with up spin

In the completely ferromagnetic case, $S_z = S$. Thus, a carrier with up-spin moving in the effective medium feels the potential of $E_M - IS - \Sigma_\uparrow$ at a magnetic site. No spin flip occurs. Therefore, the t-matrix element for the magnetic site embedded in the effective medium is given by

$$t^M_{\uparrow\uparrow} = \frac{E_B - \Sigma_\uparrow}{1 - (E_B - \Sigma_\uparrow)F_\uparrow}, \quad (10)$$

where $E_B = E_M - IS$. Thus, the CPA condition is given by

$$(1-x)t^A_{\uparrow\uparrow} + x t^M_{\uparrow\uparrow} = 0 \quad (11)$$

or

$$(1-x)\frac{E_A - \Sigma_\uparrow}{1 - (E_A - \Sigma_\uparrow)F_\uparrow} + x\frac{E_B - \Sigma_\uparrow}{1 - (E_B - \Sigma_\uparrow)F_\uparrow} = 0. \quad (12)$$

Inserting Eq. (8) to (12), we obtain a cubic equation for $F_\uparrow\Delta[\equiv F_\uparrow(\omega)\Delta]$ as

$$(F_\uparrow\Delta)^3 + a(F_\uparrow\Delta)^2 + b(F_\uparrow\Delta) + c = 0 \quad (13)$$

where

$$a = 4\left(\frac{E_B + E_A - 2\omega}{\Delta}\right) \quad (14)$$

$$b = 4\left\{1 + \frac{4(\omega - E_A)(\omega - E_B)}{\Delta^2}\right\} \quad (15)$$

$$c = 16\left\{\frac{(1-x)(E_B - E_A) - \omega + E_A}{\Delta}\right\} \quad (16)$$

4.2 Carrier with down spin

In the case of classical localized spins, the situation is very simple. We can use Eq. (13) with the coefficients of (14)-(16) only after changing $E_B = E_M - IS$ to $E_B = E_M + IS$ because a carrier with down-spin moves in the effective medium without spin flip. When we consider the effect of a finite value of S (localized spin), however, the situation becomes cumbersome. The spin of the carrier can flip with the conservation of the total spin. This is the quantum effect due to a finite value of S. The t-matrix element for the scattering of carrier with down spin by the magnetic site embedded in the effective medium is given by (see Appendix A in Ref.[7])

$$t^M_{\downarrow\downarrow} = \frac{V_\downarrow + F_\uparrow(W_\downarrow - V_\downarrow U_\uparrow)}{1 - F_\uparrow U_\uparrow - F_\downarrow[V_\downarrow + F_\uparrow(W_\downarrow - V_\downarrow U_\uparrow)]}. \quad (17)$$

In the completely ferromagnetic case, after setting $S_z = S$, we have

$$V_\uparrow \equiv E_M - IS_z - \Sigma_\uparrow = E_M - IS - \Sigma_\uparrow \quad (18)$$

$$V_\downarrow \equiv E_M + IS_z - \Sigma_\downarrow = E_M + IS - \Sigma_\downarrow \quad (19)$$

$$U_\uparrow \equiv E_M - I(S_z - 1) - \Sigma_\uparrow = E_M - I(S-1) - \Sigma_\uparrow \quad (20)$$

$$U_\downarrow \equiv E_M + I(S_z + 1) - \Sigma_\downarrow = E_M + I(S+1) - \Sigma_\downarrow \quad (21)$$

$$W_\uparrow \equiv I^2[S(S+1) - S_z^2 - S_z] = 0 \quad (22)$$

$$W_\downarrow \equiv I^2[S(S+1) - S_z^2 + S_z] = 2I^2 S \quad (23)$$

The CPA condition is given by

$$(1-x)t^A_{\downarrow\downarrow} + x t^M_{\downarrow\downarrow} = 0 \quad (24)$$

or

$$(1-x)\frac{E_A - \Sigma_\downarrow}{1 - (E_A - \Sigma_\downarrow)F_\downarrow} + x\frac{V_\downarrow + F_\uparrow(W_\downarrow - V_\downarrow U_\uparrow)}{1 - F_\uparrow U_\uparrow - F_\downarrow[V_\downarrow + F_\uparrow(W_\downarrow - V_\downarrow U_\uparrow)]} = 0. \quad (25)$$

Inserting Eq. (8) to (25), we obtain a cubic equation for $F_\downarrow\Delta[\equiv F_\downarrow(\omega)\Delta]$ as

$$(F_\downarrow\Delta)^3 + a(F_\downarrow\Delta)^2 + b(F_\downarrow\Delta) + c = 0 \quad (26)$$

where

$$a = 4\left(\frac{E_C + E_A - 2\omega}{\Delta}\right) \quad (27)$$

$$b = 4\left\{1 + \frac{4(\omega - E_A)(\omega - E_C)}{\Delta^2}\right\} \quad (28)$$

$$c = 16\left\{\frac{(1-x)(E_C - E_A) - \omega + E_A}{\Delta}\right\}. \quad (29)$$

Here, $E_C \equiv E_M + IS + Q$; Q denotes the effect of the finite value of S, and given by

$$Q = \frac{F_\uparrow W_\downarrow}{1 - F_\uparrow U_\uparrow} = \frac{W_\downarrow}{F_\uparrow^{-1} - U_\uparrow} \quad (30)$$

$$= \frac{2I^2 S}{F_\uparrow^{-1} + \Sigma_\uparrow - E_M + I(S-1)} \quad (31)$$

$$= \frac{2\left(\frac{IS}{\Delta}\right)^2 \frac{1}{S}}{\frac{\omega}{\Delta} - \frac{E_M}{\Delta} + \frac{IS}{\Delta}\left(1 - \frac{1}{S}\right) - \frac{1}{4}(F_\uparrow \Delta)}. \quad (32)$$

Therefore, the carrier state in the completely ferromagnetic state ($\langle S_z \rangle = S$) is calculated as follows. First, the carrier state with up-spin is calculated by solving the cubic equation on $F_\uparrow \Delta$ (i.e., Eq. (13) with (14) ~ (16)) for a given value of ω and for assumed values of E_M/Δ and IS/Δ. Next, using $F_\uparrow \Delta$ determined above, Q is calculated by Eq. (32). Then, a, b and c are calculated by Eqs. (27) ~ (29). The carrier state with down-spin is calculated by solving Eq. (22). Therefore, $F_\uparrow \Delta$ affects the result of $F_\downarrow \Delta$ through c (of Eq. (29)) where Q defined by Eq. (32) is employed.

5 Calculations for paramagnetic case

For paramagnetic states of $\langle S_z \rangle = 0$, $F_\uparrow = F_\downarrow = F$, and $\Sigma_\uparrow = \Sigma_\downarrow = \Sigma$. Thus, the t matrix is given by

$$t^M = \langle t^M_{\uparrow\uparrow} \rangle = \langle t^M_{\downarrow\downarrow} \rangle$$

$$= p\left\{\frac{E_M - IS - \Sigma}{1 - F(E_M - IS - \Sigma)}\right\}$$

$$+ a\left\{\frac{E_M + I(S+1) - \Sigma}{1 - F(E_M + I(S+1) - \Sigma)}\right\} \quad (33)$$

where $p = \left(\frac{S+1}{2S+1}\right)$ and $a = \left(\frac{S}{2S+1}\right)$. Equation (33) is a consequence that the exchange term $-I\boldsymbol{\sigma} \cdot \mathbf{S}$ has two energy eigenvalues, $-IS$ (with $2S+2$-fold degeneracy) and $I(S+1)$ (with $2S$-fold degeneracy). Therefore, the CPA condition

$$(1-x)t^A + xt^M = 0 \quad (34)$$

results in the CPA condition for a ternary alloy in which three species of atom having the energy level of $E_p = E_M - IS$, $E_a = E_M + I(S+1)$ and E_A are distributed at random with the fraction of xp, xa, and $1-x$;

$$(1-x)\frac{E_A - \Sigma}{1 - (E_A - \Sigma)F} + xp\frac{E_p - \Sigma}{1 - (E_p - \Sigma)F} + xa\frac{E_a - \Sigma}{1 - (E_a - \Sigma)F} = 0 \quad (35)$$

In order to solve Equation (35) we set

$$\xi = \frac{1}{F} + \Sigma. \quad (36)$$

Then, the CPA condition (35) results in a quartic equation for ξ

$$\xi^4 + A\xi^3 + B\xi^2 + C\xi + D = 0. \quad (37)$$

Here,

$$A = -(\omega + E_A + E_p + E_a) \quad (38)$$

$$B = \frac{\Delta^2}{4} + \omega(E_A + E_p + E_a) + E_p E_a \quad (39)$$

$$C = -\{\omega[(E_p E_a + E_A E_p + E_A E_a)] + E_A E_p E_a + \frac{\Delta^2}{4}[(1-x)(E_p + E_a) + x(E_A + pE_a + aE_p)]\} \quad (40)$$

$$D = \omega E_p E_a E_A + \frac{\Delta^2}{4}[(1-x)E_p E_a + x(pE_a + aE_p)] \quad (41)$$

If ξ is obtained for ω, F is calculated by

$$F = \frac{4}{\Delta^2}(\omega - \xi). \quad (42)$$

The density of states is calculated by Eq. (9).

6 Calculation for ferromagnetic states

6.1 Thermal average operation

If the magnetization $m = \langle S_z \rangle / S$ $(0 \leq m \leq 1)$is given, the parameter λ is determined so as to satisfy the equation

$$\frac{1}{\sum_{S^z=-S}^{S} e^{\lambda S^z}} \sum_{S^z=-S}^{S} e^{\lambda S^z} S^z = mS. \quad (43)$$

Employing the parameter λ determined above, the thermal average of the t-matrix over the fluctuating localized spins is taken as

$$\langle t^M_{\uparrow\uparrow}(S_z)\rangle = \frac{1}{\sum_{S^z=-S}^{S} e^{\lambda S^z}} \sum_{S^z=-S}^{S} e^{\lambda S^z} t^M_{\uparrow\uparrow}(S^z). \quad (44)$$

6.2 Initial values for coherent potentials

For a finite magnetization m, the CPA condition can not be solved analytically. Thus, we have to solve the CPA equations numerically. In the numerical procedure it is strongly desirable to take suitable initial value for the coherent potential Σ_μ. In the completely ferromagnetic case ($m=1$), we can obtain the coherent potential with $\uparrow$ spin, $\Sigma^1_\uparrow(=\omega - F^{-1}_\uparrow - F_\uparrow\Delta^2/4)$, by solving the cubic equation (13) with (14) $\sim$ (16). Then we can obtain the coherent potential with $\downarrow$ spin $\Sigma^1_\downarrow$ by solving the cubic equation (26) with (27) $\sim$ (29). In the case of paramagnetic ($m=0$) case, on the other hand, we can obtain the coherent potential $\Sigma^0(=\Sigma^0_\uparrow = \Sigma^0_\downarrow)$ by solving the quartic equation for $\xi(=\Sigma + F^{-1})$ or Eq. (37) with (38) $\sim$ (41).

When the magnetization is m, thus, we take initial values for the coherent potential Σ as

$$\Sigma^m_\uparrow = (1-m)\Sigma^0 + m\Sigma^1_\uparrow \quad (45)$$

$$\Sigma^m_\downarrow = (1-m)\Sigma^0 + m\Sigma^1_\downarrow. \quad (46)$$

6.3 Iteration approach

The iteration method for the s-f model is presented in Appendix A in Ref. [8].

In the case of diluted magnetic semiconductors, the CPA condition is written as

$$(1-x)t^A_{\uparrow\uparrow} + x\langle t^M_{\uparrow\uparrow}\rangle = 0 \quad (47)$$

$$(1-x)t^A_{\downarrow\downarrow} + x\langle t^M_{\downarrow\downarrow}\rangle = 0. \quad (48)$$

The t-matrix $t^M_{\uparrow\uparrow}$ includes not only $\uparrow$ components as $F_\uparrow$ and $\Sigma_\uparrow$ but also $\downarrow$ component as $F_\downarrow$ and $\Sigma_\downarrow$. Therefore, we have to solve Eqs. (47) and (48) simultaneously. In order to calculate $\Sigma_\uparrow$ and $\Sigma_\downarrow$ by the iteration method, we rewrite the CPA conditions. For the sake of convenience, we define the following coefficients (complex numbers) (see Appendix A in Ref. [8]).

$$A_\uparrow = \left\langle \frac{S_z/S}{(1-F_\uparrow V_\uparrow)(1-F_\downarrow U_\downarrow) - F_\uparrow F_\downarrow W_\uparrow} \right\rangle \quad (49)$$

$$B_\uparrow = \left\langle \frac{1}{(1-F_\uparrow V_\uparrow)(1-F_\downarrow U_\downarrow) - F_\uparrow F_\downarrow W_\uparrow} \right\rangle \quad (50)$$

$$A_\downarrow = \left\langle \frac{S_z/S}{(1-F_\downarrow V_\downarrow)(1-F_\uparrow U_\uparrow) - F_\downarrow F_\uparrow W_\downarrow} \right\rangle \quad (51)$$

$$B_\downarrow = \left\langle \frac{1}{(1-F_\downarrow V_\downarrow)(1-F_\uparrow U_\uparrow) - F_\downarrow F_\uparrow W_\downarrow} \right\rangle \quad (52)$$

Employing the coefficients defined above we further calculate the following coefficients by

$$D_\uparrow = ISF_\downarrow A_\uparrow - [1 + F_\downarrow(\Sigma_\downarrow - E_M)]B_\uparrow \quad (53)$$

$$D_\downarrow = -ISF_\downarrow A_\downarrow - [1 + F_\uparrow(\Sigma_\uparrow - E_M)]B_\downarrow \quad (54)$$

$$E_\uparrow = (IS)\{(IS)F_\downarrow B_\uparrow - A_\uparrow[1 + F_\downarrow(\Sigma_\downarrow - E_M)\} \quad (55)$$

$$E_\downarrow = (IS)\{(IS)F_\uparrow B_\downarrow + A_\downarrow[1 + F_\uparrow(\Sigma_\uparrow - E_M)\}. \quad (56)$$

Thus, the CPA conditions (47) and (48) are rewritten as

$$\Sigma_\uparrow = \frac{x(E_\uparrow - E_M D_\uparrow)}{1 - x\{1 + D_\uparrow + F_\uparrow[E_\uparrow - D_\uparrow(E_M - \Sigma_\uparrow)]\}} \quad (57)$$

$$\Sigma_\downarrow = \frac{x(E_\downarrow - E_M D_\uparrow)}{1 - x\{1 + D_\downarrow + F_\downarrow[E_\downarrow - D_\downarrow(E_M - \Sigma_\downarrow)]\}} \quad (58)$$

The iteration procedure is explained as follows. We first calculate the initial values $\Sigma^m_\uparrow$ and $\Sigma^m_\downarrow$ by using the exact results for paramagnetic and ferromagnetic cases. Employing these $\Sigma^m_\uparrow$ and $\Sigma^m_\downarrow$, $F_\uparrow$ and $F_\downarrow$ are calculated by Eq. (7). Furthermore, coefficients A_μ, B_μ, C_μ and D_μ are calculated by the definition. Then, new $\Sigma_\uparrow$ and $\Sigma_\downarrow$ are obtained. The procedure is iterated until the calculation converges.

6.4 Minimization method

Though the iteration approach is an effective method, the calculation for the energy difference needs be more accurate. For the purpose, we also perform the minimization method. We define f as a function of $\Sigma_\uparrow$ and $\Sigma_\downarrow$ by

$$f \equiv \left|(1-x)t^A_{\uparrow\uparrow} + x\langle t^M_{\uparrow\uparrow}\rangle\right| + \left|(1-x)t^A_{\downarrow\downarrow} + x\langle t^M_{\downarrow\downarrow}\rangle\right| \quad (59)$$

Once $\Sigma_\uparrow$ and $\Sigma_\downarrow$ are given, the calculation for f is straightforward. Then, we can search for the pairs of $\Sigma_\uparrow$ and $\Sigma_\downarrow$ which satisfy the condition (59) well enough. The actual steps are as follows. When $\Sigma_\uparrow^j$ and $\Sigma_\downarrow^j$ are given, we calculate f for the total $81(= 9\times 9)$ pairs of $\Sigma_\uparrow$ and $\Sigma_\downarrow$; $\Sigma_\uparrow = \Sigma_\uparrow^j, \Sigma_\uparrow^j + (\pm d, \pm di)$ and $\Sigma = \Sigma_\downarrow^j + (\pm d, \pm di)$. Here $d (> 0)$ is a step unit for search and $i = \sqrt{-1}$ is complex unit. If the value of f for $\Sigma_\uparrow^j$ and $\Sigma_\downarrow^j$ is not the minimum among these 81 values, the $\Sigma_\uparrow^j$ and $\Sigma_\downarrow^j$ are replaced by $\Sigma_\uparrow$ and $\Sigma_\downarrow$ which give the minimum for f. If the value of f for $\Sigma_\uparrow^j$ and $\Sigma_\downarrow^j$ is the minimum among these 81 values, the step is taken as $d/2$. The procedure is continued until the value of f diminishes small enough.

7 Results and Discussion

The iteration approach gives satisfactory result in the case of the *s*-*f* model as shown in Ref. [8]. However, it does not converge well in the case of DMS even though the classical spins are assumed for localized spins [9]. This is because of the difficulty due to the random distribution of magnetic ions incorporated in DMS. More precise calculations are needed for the energy difference. Therefore, we will employ the minimization method together with the iteration approach. Here, we note that the minimization method and the iteration approach are used together in our previous works in Ref. [4–7, 10].

We have verified in the classical spin limit that the dynamical CPA gives almost the same result as obtained by the dynamical mean field theory (DMFT) [10]. However, the dynamical CPA is more easily tractable than the DMFT and gives some important limiting cases.

References

[1] M. Takahashi: Physical Review B **60** no. 23 (1999) pp. 15858-15865.

[2] M. Takahashi: J. Phys; Cond. Matt. 13, no. 14, (2001) pp. 3433-3441.

[3] M. Takahashi: J. Phys. Soc. Japan **70** no. 7, (2001) pp. 2224-2225.

[4] M. Takahashi and K. Kubo: Phys. Rev. B **66**, (2002) 153202.

[5] M. Takahashi and K. Kubo: J. Phys. Soc. Jpn **72**, (2003) 2866.

[6] Masao Takahashi: Recent Research Developments in Magnetism & Magnetic Materials Vol.1 -2003 Part I Ed. by S. G. Pandalai (Transworld Research Network, Kerala, India) pp. 195-225 (2003).

[7] M. Takahashi: Phys. Rev. B **70** no. 3 (2004) 035207.

[8] M. Takahashi and K. Mitsui: Phys. Rev. B **54** (1996) 11298.

[9] M. Takahashi: Phase Transition **75** no. 7-8 (2002) pp. 1035-1041.

[10] M. Takahashi, N. Furukawa, and K. Kubo: J. Magn. Magn. Mater. **272-276**, (2004) 2021.