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CPA density of states and conductivity in a double-exchange system containing impurities

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Abstract. We study density of states and conductivity of the doped double-exchange system, treating interaction of charge carriers both with the localized spins and with the impurities in the coherent potential approximation. It is shown that under appropriate conditions there is a gap between the conduction band and the impurity band in paramagnetic phase, while the density of states is gapless in ferromagnetic phase. This can explain metal-insulator transition frequently observed in manganites and magnetic semiconductors. Activated conductivity in the insulator phase is numerically calculated.

PACS. 75.50.Pp Magnetic semiconductors – 75.30.Vn Colossal magnetoresistance – 72.10.-d Theory of electronic transport; scattering mechanisms

1 Introduction

The recent rediscovery of colossal magnetoresistance (CMR) in doped Mn oxides with perovskite structure $R_{1-x}D_xMnO_3$ (R is a rare-earth metal and D is a divalent metal, typically Ba, Sr or Ca) [1] has generated substantial interest in these materials [2]. The doping of parent material $RMnO_3$ by a divalent metal is the source of the holes responsible for the transport properties of these materials. In addition, each divalent atom introduced, is the center of an impurity potential. Many papers analyzed the influence of strong magnetic disorder, inherent in the the CMR materials at finite temperature, upon the single-particle states and transport properties. However, the interplay between the magnetic disorder and the doping-induced disorder was studied less. The impurity potential plays double role. First, the potential fluctuations determine the transport at temperatures well below the ferromagnet (FM) - paramagnet (PM) transition point T_c. Second, strong potential may pin the Fermi level either in the conduction band tail (in the Anderson model of disorder [3]), or in the emerging impurity band. The analysis of experimental data reveals strong relevance of the latter effect to metal-insulator transition (MIT) near T_c both in magnetic semiconductors [4] and manganites [5]. However, to the best of our knowledge the impurity-band scenario in the double-exchange (DE) model was not discussed yet.

The present paper is devoted to the consideration of single-particle states and conductivity in impure DE system. Interaction of charge carriers both with the localized spins and with the impurities is strong, so it is definitely not enough to limit ourselves with the finite number of terms of perturbation expansion. A simple but physically meaningful approximation, allowing to sum up infinite number of perturbation expansion terms is the coherent potential approximation (CPA). Initially CPA was proposed to treat potential disorder [6], but soon after it's appearance the generalization to random spin system was developed [7]. The CPA was also used to describe diluted magnetic semiconductors [8].

In the present paper we for the first time treat on equal footing the interactions of electrons with the core Mn spins and with the doping impurities using the matrix generalization of the CPA. The concurrent action of potential disorder and temperature dependent spin disorder leads to a number of interesting phenomena, in particular to the possibility of the opening of the gap at the Fermi level with the increase of temperature and, hence, to MIT transition.

2 Hamiltonian and theoretical formulation

We consider the DE model with the inclusion of the single-site impurity potential. In addition, as it is widely accepted, we apply the quasiclassical adiabatic approximation and consider each Mn spin as a static vector with a fixed length S ($\mathbf{S}_i = S\mathbf{n}_i$, where $\mathbf{n}_i$ is a randomly oriented unit vector). The Hamiltonian of the model in site representation is

$$\hat{H}_{ij} = t_{i-j} + \delta_{ij}\left(\epsilon_i - J\mathbf{n}_i \cdot \hat{\sigma}\right) = H_{\rm kin} + V_{\rm imp} + \hat{V}_{\rm sd}, \quad (1)$$

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where t_{i-j} is the electron hopping, ϵ_i is the random on-site energy, J is the effective exchange coupling between a Mn core spin and a conduction electron and $\hat{\sigma}$ is the vector of the Pauli matrices. The hat above the operator reminds that in one-particle representation it is a 2×2 matrix in the spin space (we discard the hat when the operator is a scalar matrix in the spin space).

We present Hamiltonian as

$$\hat{H} = H_{\rm kin} + \hat{\Sigma} + V_{\rm imp} + \hat{V}_{\rm sd} - \hat{\Sigma} = \hat{H}_0 + \hat{V} \qquad (2)$$

(the site independent self-energy $\hat{\Sigma}(E)$ is to be determined later), and construct a perturbation theory with respect to random potential $\hat{V} = V_{\rm imp} + \hat{V}_{\rm sd} - \hat{\Sigma}$. To do this let us introduce the T-matrix as the solution of the equation

$$\hat{T} = \hat{V} + \hat{V}\hat{G}_0\hat{T}, \qquad (3)$$

where

$$\hat{G}_0 = \frac{1}{E - \hat{H}_0} \cdot \qquad (4)$$

For the exact Green function we get

$$\hat{G} = \hat{G}_0 + \hat{G}_0\hat{T}\hat{G}_0. \qquad (5)$$

The coherent potential approximation (CPA) is expressed by the equation

$$\left\langle \hat{G} \right\rangle = \hat{G}_0. \qquad (6)$$

This equation can also be presented as

$$\left\langle \hat{T}_i \right\rangle = 0, \qquad (7)$$

where $\hat{T}_i$ is the solution of the equation

$$\hat{T}_i = \hat{V}_i + \hat{V}_i\hat{g}(E - \hat{\Sigma})\hat{T}_i, \qquad (8)$$

and

$$g(E) = (G_0(E))_{ii} = \int \frac{N_0(\varepsilon)}{E - \varepsilon} {\rm d}\varepsilon, \qquad (9)$$

where $N_0(\varepsilon)$ is the bare density of states. The averaging in equations (6, 7) should be performed both with respect to random orientations of core spins and with respect to random on-site energies. We obtained, in fact, the algebraic equation for the 2×2 matrix $\hat{\Sigma}$

$$\left\langle \left[1 - \hat{V}_i\hat{g}(E - \hat{\Sigma})\right]^{-1} \hat{V}_i \right\rangle = 0. \qquad (10)$$

This equation takes into account scattering both due to randomness of the core spins, and due to the impurities. If the impurity potential is negligible ($V = 0$) this equation coincides with the equation (20) of reference [9] obtained in the dynamical mean field approximation (and also with those obtained for the Falikov-Kimball model [10,11]).

In the reference frame where the z-axis is directed along the magnetization, $\hat{\Sigma}$ is diagonal, and equations (10) reduces to the system of two equations for its diagonal matrix elements $\Sigma_\sigma(E)$ ($\sigma = \uparrow, \downarrow$). The equations acquire especially simple form at two extreme particular cases, which we will analyze:

(a) $T = 0$. The magnetic state is coherent FM with $n_i^z = 1$, and equation (10) takes the form

$$\left\langle \frac{\epsilon_i \mp J - \Sigma_{\uparrow,\downarrow}}{1 - (\epsilon_i \mp J - \Sigma_{\uparrow,\downarrow})\, g(E - \Sigma_{\uparrow,\downarrow})} \right\rangle = 0. \qquad (11)$$

(b) $T \geq T_{\rm c}$ and zero magnetic field. The magnetic state is isotropic PM with $\langle \mathbf{n_i} \rangle = 0$, which leads to $\Sigma_\downarrow = \Sigma_\uparrow = \Sigma$, and equation (10) takes the form

$$\left\langle \frac{\epsilon_i + J - \Sigma}{1 - (\epsilon_i + J - \Sigma)\, g(E - \Sigma)} \right\rangle + \left\langle \frac{\epsilon_i - J - \Sigma}{1 - (\epsilon_i - J - \Sigma)\, g(E - \Sigma)} \right\rangle = 0. \qquad (12)$$

We will solve the equations (11) and (12) in the strong Hund coupling limit ($J \to \infty$). In this limit we obtain two decoupled spin sub-bands. The equation for the upper sub-band, after shifting the energy by $-J$, for both cases (a) and (b) can be written down in unified form

$$\left\langle \frac{1}{1 - (\epsilon_i - \Sigma)\, g(E - \Sigma)} \right\rangle = \alpha, \qquad (13)$$

where $\alpha = 1$ for $T = 0$, $\alpha = 2$ for $T \geq T_{\rm c}$. In the model of substitutional disorder ($\epsilon_i = 0$ with probability x, and $\epsilon_i = V$ with probability $1 - x$), equation (13) takes the form

$$\frac{1 - x}{1 + \Sigma g\,(E - \Sigma)} + \frac{x}{1 + (\Sigma - V)\, g\,(E - \Sigma)} = \alpha. \qquad (14)$$

3 The CPA equations for semi-circular bare density of states

We consider semi-circular (SC) bare DOS

$$N_0(\varepsilon) = \frac{4}{\pi W}\sqrt{1 - \left(\frac{2\varepsilon}{W}\right)^2}, \qquad (15)$$

at $|\varepsilon| \leq W/2$ and $N_0(\varepsilon) = 0$ otherwise, for which

$$g(E) = \frac{4}{W}\left[\frac{2E}{W} - \sqrt{\left(\frac{2E}{W}\right)^2 - 1}\right]. \qquad (16)$$

Let us introduce the following normalized quantities

$$\lambda = \frac{\Sigma}{W},\ \omega = \frac{E}{W},\ v = \frac{V}{W} \qquad (17)$$

After simple algebra we obtain from equation (14) the cubic equation with respect to

$$\gamma \equiv W g\,(E - \Sigma) = 8\left[\omega - \lambda - \sqrt{(\omega - \lambda)^2 - 1/4}\right], \qquad (18)$$

in the form

$$\gamma^3 + 16\left(v - 2\omega\right)\gamma^2 + 16\left[\frac{1}{\alpha} - 16\omega\left(v - \omega\right)\right]\gamma - 256\frac{\omega}{\alpha} + 256\left(1 - x\right)\frac{v}{\alpha} = 0. \quad (19)$$

The number of electrons per cite n is given by

$$n = \int_{-\infty}^{\infty} f(E)N(E)\mathrm{d}E, \quad (20)$$

where $f(E)$ is the Fermi distribution function, and

$$N(E) = \frac{\alpha}{W\pi}\mathrm{Im}\,\gamma \quad (21)$$

is the actual density of states. To define the position of μ, the Fermi level, we must impose the relation between n and x; the simplest assumption appropriate for manganites is the equation $n = 1 - x$.

4 Conductivity in CPA

For a disordered one-electron system the static conductivity is given by

$$\rho^{-1} = \frac{e^2\pi\hbar}{\mathrm{V}}\int\left(-\frac{\partial f}{\partial E}\right) \times \left\langle \mathrm{Tr}\left[\hat{v}_\alpha \delta\left(E - \hat{H}\right)\hat{v}_\alpha \delta\left(E - \hat{H}\right)\right]\right\rangle \mathrm{d}E, \quad (22)$$

where V is the volume and $\hat{v}_a$ is a Cartesian component of the velocity operator. To obtain the conductivity in CPA let us express operator delta-function as follows

$$\delta\left(E - \hat{H}\right) = \frac{1}{2\pi\mathrm{i}}\left[\hat{G}(E_-) - \hat{G}(E_+)\right]. \quad (23)$$

Using equations (5) and (4) in Bloch representation

$$\left\langle \mathbf{k}\sigma \left| \hat{G}_0(E) \right| \mathbf{k}'\sigma' \right\rangle = \frac{\delta_{\mathbf{k},\mathbf{k}'}\delta_{\sigma,\sigma'}}{E - \varepsilon_{\mathbf{k}} - \Sigma_\sigma\left(E\right)}, \quad (24)$$

we get

$$\left\langle \mathrm{Tr}\left[\hat{v}_a \delta\left(E - \hat{H}\right)\hat{v}_a \delta\left(E - \hat{H}\right)\right]\right\rangle = \sum_{\mathbf{k},\sigma} v_{\mathbf{k}\alpha}^2 \left[A_\sigma\left(\varepsilon_{\mathbf{k}}, E\right)\right]^2 + O\left(\left\langle \hat{T}\hat{T}\right\rangle\right), \quad (25)$$

where

$$A_\sigma\left(\varepsilon, E\right) = \frac{1}{\pi}\frac{\mathrm{Im}\Sigma_\sigma\left(E\right)}{\left[E - \varepsilon - \mathrm{Re}\Sigma_\sigma\left(E\right)\right]^2 + \left[\mathrm{Im}\Sigma_\sigma\left(E\right)\right]^2} \quad (26)$$

is the one-particle spectral weight function. On account of the locality of T-matrix the second term in the trace is equal to

$$\sum_{s,s'=\pm;\,\mathbf{k},\mathbf{k}',\sigma,\sigma'} ss' v_{\mathbf{k}\alpha} v_{\mathbf{k}'\alpha} G_\sigma\left(\varepsilon_{\mathbf{k}}, E_s\right) G_{\sigma'}\left(\varepsilon_{\mathbf{k}'}, E_s\right) \times G_{\sigma'}\left(\varepsilon_{\mathbf{k}'}, E_{s'}\right) G_\sigma\left(\varepsilon_{\mathbf{k}}, E_{s'}\right) \times \left\langle T_{\sigma\sigma'}(\mathbf{k} - \mathbf{k}', E_s) T_{\sigma'\sigma}(\mathbf{k}' - \mathbf{k}, E_{s'})\right\rangle. \quad (27)$$

Since in CPA $\left\langle T_{\sigma\sigma'}(\mathbf{k} - \mathbf{k}', E_s) T_{\sigma'\sigma}(\mathbf{k}' - \mathbf{k}, E_{s'})\right\rangle$ does not depend on $\mathbf{k}$ and $\mathbf{k}'$ and $v_{-\mathbf{k}\alpha} = -v_{\mathbf{k}\alpha}$ the above expression is identically zero [12,13]. Thus, finally

$$\rho^{-1} = \frac{e^2\pi\hbar}{v}\int\int\left(-\frac{\partial f}{\partial E}\right) \times v_\alpha^2\left(\varepsilon\right) N_0\left(\varepsilon\right)\sum_\sigma \left[A_\sigma\left(\varepsilon, E\right)\right]^2 \mathrm{d}E\mathrm{d}\varepsilon, \quad (28)$$

where v is the unit cell volume and by definition

$$v_\alpha^2\left(\varepsilon\right) N_0\left(\varepsilon\right) = \frac{1}{N\hbar^2}\sum_{\mathbf{k}}\left(\frac{\partial\varepsilon_{\mathbf{k}}}{\partial k_\alpha}\right)^2 \delta\left(\varepsilon - \varepsilon_{\mathbf{k}}\right). \quad (29)$$

Let us assume nearest-neighbor tight binding spectrum on simple d-hypercubic lattice ($v = a^d$)

$$\varepsilon_{\mathbf{k}} = -t\sum_{\alpha=1}^{d}\cos ak_\alpha,$$
$$v_\alpha^2\left(\varepsilon\right) N_0\left(\varepsilon\right) = -\frac{a^2}{d\hbar^2}\int_{-\infty}^{\varepsilon}\epsilon N_0\left(\epsilon\right)\mathrm{d}\epsilon. \quad (30)$$

In SC DOS model

$$v_\alpha^2\left(\varepsilon\right) N_0\left(\varepsilon\right) = -\frac{4}{\pi W}\frac{a^2}{d\hbar^2}\int_{-W/2}^{\varepsilon} z\sqrt{1 - \left(\frac{2z}{W}\right)^2}\mathrm{d}z = \frac{1}{3}\frac{W}{\pi}\frac{a^2}{d\hbar^2}\left(1 - \frac{4\varepsilon^2}{W^2}\right)^{3/2}. \quad (31)$$

Substituting this result into equation (28) we obtain

$$\rho^{-1} = \sigma_0\int\left(-\frac{\partial f}{\partial E}\right)\Lambda\left(E\right)\mathrm{d}E, \quad (32)$$

with

$$\sigma_0 = \frac{e^2}{2\pi d a^{d-2}\hbar} \quad (33)$$

being the Mott minimal metallic conductivity, and

$$\Lambda\left(E\right) = \frac{2W}{3\pi}\int_{-W/2}^{W/2}\left(1 - \frac{4\varepsilon^2}{W^2}\right)^{3/2} \sum_\sigma\left\{\mathrm{Im}\left[\frac{1}{E - \varepsilon - \Sigma_\sigma\left(E\right)}\right]\right\}^2 \mathrm{d}\varepsilon. \quad (34)$$

For the strong Hund coupling we obtain

$$\Lambda\left(E\right) = \frac{4}{3\pi}\int_{-1}^{1}\left(1 - x^2\right)^{3/2}\left\{\mathrm{Im}\left[\frac{1}{x - z}\right]\right\}^2 \mathrm{d}x, \quad (35)$$

where $z = 2\left(\omega - \lambda\right)$. equations (32, 35) give the conductivity in the framework of bare SC DOS model for arbitrary hole concentration x and impurity potential strength V.

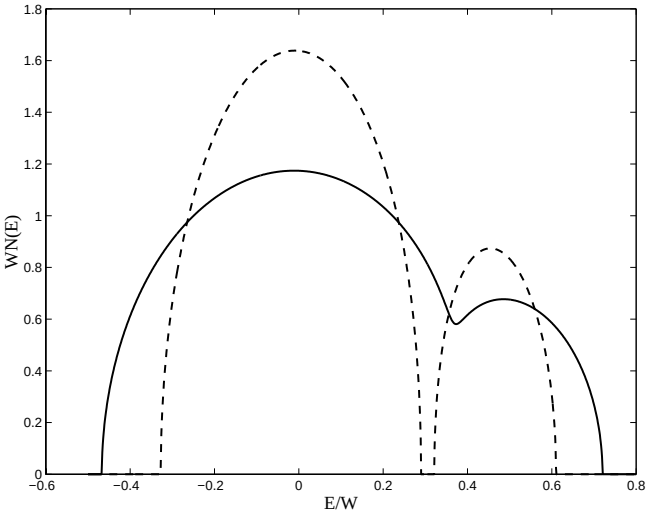

Fig. 1. DOS at $T = 0$ (full line) and high-temperature (dashed line) in the units of W^{-1} for $x = 0.2$ and $V/W = 0.4$

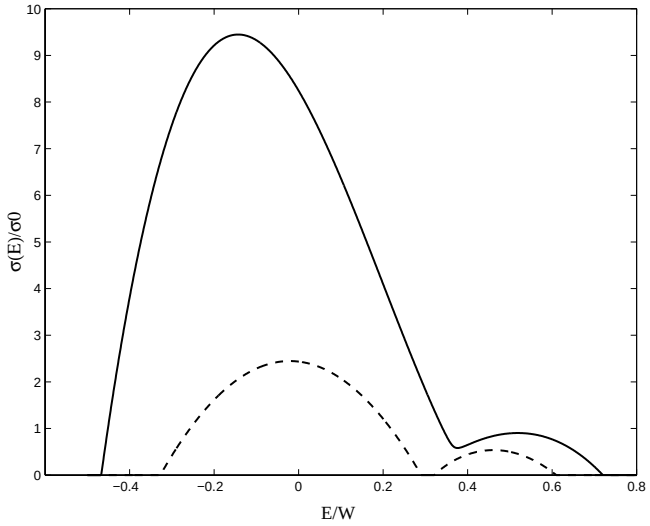

Fig. 2. Energy-dependent conductivity at $T = 0$ (full line) and high-temperature (dashed line) in the units of σ_0 for $x = 0.2$ and $V/W = 0.4$

5 Influence of the impurity potential

First, consider density of states. It is known, that within CPA for every x there exists critical value of potential-to-bandwidth ratio $v_c(x)$ such that at $v > v_c(x)$ the separate impurity band splits off the conduction band (that is a gap opens in $N(E)$). In our approach we get two different curves $v_c(x, T = 0)$ and $v_c(x, T \geq T_c)$, which present boundaries of metal-insulator and metal-semiconductor "phase diagrams", respectively, in (v, x) plane. Due to effect of magnetic disorder it appears that $v_c(x, T = 0) > v_c(x, T \geq T_c)$.

For a typical concentration $x = 0.2$, $v_c(0.2, T = 0) \approx 0.49$ and $v_c(0.2, T \geq T_c) \approx 0.35$. So if we choose $v = 0.4$ both $N(E)$ and $\Lambda(E)$ must be gapless at $T = 0$ but do have a gap at $T \geq T_c$. Numerical calculations of the DOS performed at $T = 0$ and $T \geq T_c$ for the above x and v clearly demonstrate FM-PM transition induced band splitting (Fig. 1).

Now address the question of conductivity. Consider first the position of μ and conductivity at $T = 0$. We get from equation (20) $\mu(T = 0) = 0.3662W$. Note that $\mu(T = 0)$ lies on the neck connecting conduction band and impurity states derived parts of the band. As a result, the residual conductivity (equation (32)) $\rho^{-1}(T = 0) = 0.6163\sigma_0$ is less then the Mott limit.

At $T \geq T_c$ DOS and $\Lambda(E)$ have the same gap $\Delta = 0.031W$ (see Figs. 1, 2), so $\mu(T \geq T_c)$ must lie in the gap. Thus, the model describes a bad metal at $T = 0$, and a semiconductor at $T \geq T_c$. The transition between two types of conduction (FM-PM transition induced MIT) should occur at some temperature below T_c. Such a picture agrees with the recent photoemission experiments showing drastic decrease of DOS at the Fermi level [14] as temperature increases towards T_c.

It is checked numerically that DOS displays square-root like behavior near the top of the conduction band E_c

$$N(E) \approx n_c \sqrt{E_c - E}, \tag{36}$$

and the bottom of the impurity band E_i

$$N(E) \approx n_i \sqrt{E - E_i}. \tag{37}$$

Unlike DOS $\Lambda(E)$ behaves *linearly* near the band edges

$$\begin{aligned} \Lambda(E) &\approx W^{-1}\lambda_c(E_c - E), \quad \text{for } E < E_c; \\ \Lambda(E) &\approx W^{-1}\lambda_i(E - E_i), \quad \text{for } E > E_i. \end{aligned} \tag{38}$$

The assumption $T < \Delta$ allows us to explicitly obtain $\mu(T \geq T_c)$. Calculating integrals in equation (20) with exponential accuracy we obtain

$$\mu \approx \frac{1}{2}\left(E_c + E_i + T \ln \frac{n_c}{n_i}\right). \tag{39}$$

The integral in equation (32), calculated with the same accuracy, leads to activation law for conductivity with *linear* temperature pre-exponent

$$\rho^{-1} \approx \sigma_0 \frac{BT}{W} \exp\left(-\frac{E_A}{T}\right), \tag{40}$$

where $E_A = \Delta/2 \approx 0.015W$ and B is the following numerical constant

$$B = \lambda_c \sqrt{\frac{n_i}{n_c}} + \lambda_i \sqrt{\frac{n_c}{n_i}} \approx 22 \tag{41}$$

for the parameters considered.

Low values of conductivity obtained for the case of spin disorder are an indication of the possibility of Anderson localization [4,15,16], which CPA is incapable of accessing. But the present results complement and support the localization based approach. In fact, the results of reference [4] were obtained under the assumption of the Fermi level pinning, which is now explained as being due to strong electron-impurities interaction (and the impurity band formation).

In another aspect, the model considered may also explain low-temperature MIT observed in initially metallic manganites $R_{1-x}D_xMnO_3$ upon substitution of R by isovalent atoms (*e.g.* La by Y [17]). One may speculate that the substitution forms a deep impurity band which can capture holes in $R_{1-x}D_xMnO_3$.

6 Conclusion

To conclude, we derived CPA equations for the one-electron Green function and conductivity of DE system containing impurities. The equations were solved for the SC bare DOS and substitutional disorder model. It was shown that if the electron-impurity interaction is strong enough, there is a gap between the conduction band and the impurity band in PM phase, the density of states being gapless in FM phase. Under appropriate doping conditions the chemical potential is pinned inside the gap. This can explain metal-insulator transition observed in manganites and magnetic semiconductors.

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