



Spectral density approach to the double exchange model

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Abstract

In rare-earth manganites the itinerant e_g -electrons interact via exchange coupling with localized t_{2g} -electrons which constitute the spin background. This fact is taken into account in the double exchange model *DEM*. The limit realistic to manganites is the strong coupling limit where the Hund's rule coupling constant between e_g - and t_{2g} -electrons is large compared to the e_g -hopping integral. The spectral density approach, a reliable scheme in the strong coupling limit, has been adopted to obtain the density of states *DOS* for the e_g -band. In this limit, the e_g -band splits into two bands with different weights for spin-up and spin-down states. In the ferromagnetic state, the effective band width of the split band depends on the spin state of the e_g -electron. Within the framework of the model and the approximation, it is observed that the temperature drives the half-metal to metal transition. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Rare-earth manganites have shot into prominence due to the discovery of the phenomenon of colossal magnetoresistance *CMR* [1] and its potential technological applications. In addition to *CMR*, these materials, both doped and undoped, display a fascinating variety of magnetic, charge and orbital ordered phases and also unusual transport properties [2,3]. The chemical formula of doped rare-earth manganites is, $\text{Re}_{1-x}\text{D}_x\text{MnO}_3$ where *Re* is the rare-earth $\text{Re} = \text{La}, \text{Pr}$ or Nd , and *D* is the divalent ion $D = \text{Ca}, \text{Sr}$ or Ba . In the undoped situation, the *Mn* is in the Mn^{3+} state and has four electrons in the d-shell. When doped, there are $1-x$ *Mn* ions in Mn^{3+} state with four d-electrons and x ions in the Mn^{4+} state with three d-electrons. The unusual properties of these systems are ascribed to the electrons in the d-shell of the *Mn* ion. It is well known that strong Hund's rule coupling is operative for the d-states which are five-fold degenerate. Due to crystal field splitting, the states are split into a lower-lying localized three-fold degenerate t_{2g} -level and a higher lying extended doubly degenerate e_g -state. In undoped systems, the t_{2g} -state is occupied by

three electrons with their spins aligned parallel due to the strong Hund's rule coupling and this provides the spin background with $S = 3/2$ in which the e_g -band electrons move. The electrons in the band states, which are $1-x$ per *Mn* where x is the concentration of the divalent dopant, are also strongly Hund's rule coupled to the localized spins with a coupling constant j . Since in the manganites, all the interesting phenomena track with either the dopant concentration or temperature, it is conceivable that the band occupation and temperature have a profound influence on the electronic structure of the e_g -band. It is the purpose of the present work to study this particular aspect.

The magnetic ordering in undoped systems, a representative example of which is LaMnO_3 , was studied long ago [4] by using what is known as the double exchange model *DEM*. It seems reasonable that the same model would be a good starting point for studying the resistivity anomalies which are the focus of attention in the *CMR* materials. The same model, in infinite dimension $D = \infty$ and in the limit of large j was used [5,6] to study the transport properties. It is, however, becoming clear that, in order to understand *CMR*, just the *DEM* is not sufficient but has to be supplemented with other interactions such as the electron-phonon interaction, [7] Hubbard correlation etc. Still, the minimal *DEM* is not yet completely understood even though

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there are some exactly known limiting cases. The DEM is nothing but the ferromagnetic Kondo lattice model in the strong coupling limit. This model has two important non-trivial limiting cases, namely, the zero band width limit [8] and the ferromagnetically saturated semiconductors [9,10], where it is exactly solvable. For finite band width and varying band occupation and for finite temperature, it is still interesting and necessary to develop reliable approximation schemes. Since the motivation is to study the manganites where the Hund's rule coupling is the dominant interaction, the DEM has to be treated in the strong coupling limit. In the case of the Hubbard model, a reliable approximation scheme known as the spectral density approach *SDA* [11,12] was developed for the strong coupling limit. We propose to employ the same approach to solve the DEM. In this approach a physically motivated ansatz is made for the one-electron spectral density which involves some unknown parameters. These parameters are evaluated self-consistently by using a moment method. Once the one-electron spectral density is known, it is but a short step to determine the quasi-particle density of states *QDOS*. The so obtained QDOS when studied for different temperatures and band occupations, yield interesting features of the model. In the present work, as in most theoretical works on CMR (e.g., [13]), the local spin of the t_{2g} -electrons is treated as a classical object. The justification for this is that the most important energy scale is JS and we are working in the limit of large JS . However, recently, there have been some efforts to treat the t_{2g} -spin quantum mechanically [14]. Further, the magnetic ordering of the localized spins is assumed to originate outside the model and is introduced into the calculation by hand by using a Langevin type of function for the magnetization. Obtaining magnetization as a self-consistent result of the calculation will be attempted in future. Thus, the temperature dependence of the QDOS of the e_g -band stems from the temperature dependence of the magnetization. The concentration of the dopant fixes the e_g -band occupation, which in turn fixes the chemical potential.

2. Model and approximation

The Hamiltonian of the double exchange model is given by [4]

$$H = \sum_{i,j,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} - \frac{J}{2} \sum_i [S_i^z (c_{i\uparrow}^\dagger c_{i\downarrow} - c_{i\downarrow}^\dagger c_{i\uparrow}) + S_i^+ c_{i\downarrow}^\dagger c_{i\uparrow} + S_i^- c_{i\uparrow}^\dagger c_{i\downarrow}] \quad (1)$$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) is the creation (annihilation) operator for an e_g -electron with spin σ , S_i is the total spin of the localized t_{2g} -electrons, j is the Hund's rule coupling constant for the intra-site interaction between the e_g - and t_{2g} -electrons, t_{ij} is the hopping integral for the e_g -electrons from the lattice site

i to j and is related to Bloch energy $\epsilon_{\mathbf{k}}$ by

$$t_{ij} = \frac{1}{N} \sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \quad (2)$$

The Greens functions and the spectral function to be calculated respectively are

$$G_{\mathbf{k}\sigma}(E) = \langle\langle c_{\mathbf{k}\sigma}^\dagger; c_{\mathbf{k}\sigma} \rangle\rangle_E \quad ; \quad S_{\mathbf{k}\sigma}(E) = -\frac{1}{\pi} \Im G_{\mathbf{k}\sigma} \quad (3)$$

In the atomic limit and for classical spin, this Greens function has two poles at $\pm JS/2$. Therefore it is realistic to make a two pole ansatz in the finite band width case also:

$$S_{\mathbf{k}\sigma}(E) = \sum_{j=1}^2 \alpha_{j\sigma}(\mathbf{k}) \delta(E - E_{j\sigma}(\mathbf{k})) \quad (4)$$

Here the spectral weights $\alpha_{j\sigma}(\mathbf{k})$ and the poles $E_{j\sigma}(\mathbf{k})$ are unknown and are to be determined. For this, we use two definitions of spectral moments:

$$P_{\mathbf{k}\sigma}^{(m)} = \int dE E^m S_{\mathbf{k}\sigma}(E) \quad (5)$$

$$P_{\mathbf{k}\sigma}^{(m)} = \langle \underbrace{[\dots [c_{\mathbf{k}\sigma}, H]_-, \dots, H]_-, c_{\mathbf{k}\sigma}^\dagger}_m \rangle_+ \quad (6)$$

The ansatz for the spectral density involves only four unknowns. Therefore it is sufficient to calculate only four moments. Calculating the commutators in Eq. 6 and using Eq. 4 in Eq. 5 we get the set of equations to determine the spectral weights $\alpha_{j\sigma}(\mathbf{k})$ and the poles $E_{j\sigma}(\mathbf{k})$ as follows:

$$\alpha_{1\sigma}(\mathbf{k}) + \alpha_{2\sigma}(\mathbf{k}) = 1$$

$$E_{1\sigma}(\mathbf{k})\alpha_{1\sigma}(\mathbf{k}) + E_{2\sigma}(\mathbf{k})\alpha_{2\sigma}(\mathbf{k}) = \epsilon_{\mathbf{k}} - z_{\sigma} \frac{J}{2} M$$

$$E_{1\sigma}^2(\mathbf{k})\alpha_{1\sigma}(\mathbf{k}) + E_{2\sigma}^2(\mathbf{k})\alpha_{2\sigma}(\mathbf{k}) = \epsilon_{\mathbf{k}}^2 - z_{\sigma} J \epsilon_{\mathbf{k}} M + \frac{J^2}{4} \langle S^2 \rangle \quad (7)$$

$$E_{1\sigma}^3(\mathbf{k})\alpha_{1\sigma}(\mathbf{k}) + E_{2\sigma}^3(\mathbf{k})\alpha_{2\sigma}(\mathbf{k}) = \epsilon_{\mathbf{k}}^3 - z_{\sigma} \frac{3}{2} J \epsilon_{\mathbf{k}}^2 M$$

$$+ \frac{J^2}{2} \epsilon_{\mathbf{k}} \left[\langle S^2 \rangle + \frac{M^2}{2} \right] - z_{\sigma} \frac{J^3}{8} M \langle S^2 \rangle$$

Where $M = \langle S^z \rangle$. Solving the above set of equations we obtain:

$$E_{j\sigma}(\mathbf{k}) = \frac{1}{2} \left[\epsilon_{\mathbf{k}} + (-1)^j \sqrt{\epsilon_{\mathbf{k}}^2 - 2z_{\sigma} J \epsilon_{\mathbf{k}} M + J^2 \langle S^2 \rangle} \right] \quad (8)$$

$$\alpha_{j\sigma}(\mathbf{k}) = \frac{1}{2} \left[1 + (-1)^j \frac{(\epsilon_{\mathbf{k}} - z_{\sigma} J M)}{\sqrt{\epsilon_{\mathbf{k}}^2 - 2z_{\sigma} J \epsilon_{\mathbf{k}} M + J^2 \langle S^2 \rangle}} \right] \quad (9)$$

Here $z_{\uparrow, \downarrow} = \pm 1$. This way of determining the spectral function and therefore the Green's function is known as the spectral density approach *SDA* [8]. In this method the

only approximation is making an ansatz for $S_{\mathbf{k}\sigma}(E)$. The rest of the calculation is exact. Therefore, the method is non-perturbative and is known to be more reliable in the strong coupling limit for the Hubbard model and Anderson model. In manganites, it is the strong coupling limit since j is larger than the bandwidth. Having determined $S_{\mathbf{k}\sigma}(E)$, the self-energy is obtained from the equation for the the Green's function $G_{\mathbf{k}\sigma}(E)$:

$$G_{\mathbf{k}\sigma}(E) = \frac{\alpha_{1\sigma}(\mathbf{k})}{E - E_{1\sigma}(\mathbf{k})} + \frac{\alpha_{2\sigma}(\mathbf{k})}{E - E_{2\sigma}(\mathbf{k})} = \frac{1}{E - \epsilon_{\mathbf{k}} - \Sigma_{\sigma}(E)} \quad (10)$$

Then we get the self-energy $\Sigma_{\sigma}(E)$ as

$$\Sigma_{\sigma}(E) = -z_{\sigma} \frac{JM}{2} \frac{E - z_{\sigma} \frac{J\langle S^2 \rangle}{2M}}{E - z_{\sigma} \frac{JM}{2}} \quad (11)$$

In the present calculation, $M = \langle S^z \rangle$ does not come as a consequence of the interaction included. Therefore it has been treated as a parameter. From the knowledge of $G_{\mathbf{k}\sigma}(E)$ it is straightforward to obtain the spin dependent density of states $\rho_{\sigma}(E)$:

$$\rho_{\sigma}(E) = -\frac{1}{\pi N} \sum_{\mathbf{k}} \Im G_{\mathbf{k}\sigma}(E + i0^+) \quad (12)$$

The spin dependent average occupation number is given by

$$n_{\sigma} = \int_{-\infty}^{\infty} dE f_{-}(E - \mu) \rho_{\sigma}(E) \quad (13)$$

where $f_{-}(E - \mu)$ is the Fermi function.

3. Results and discussion

Before presenting the numerical results, it is instructive to examine the limiting case of large J . As is to be expected, the e_g -band splits into two sub-bands for each spin direction centred at $\pm JS/2$. In the limit of large J , for the two sub-bands we get

$$E_{j\sigma}(\mathbf{k}) = (-1)^j \frac{JS}{2} + \frac{\epsilon_{\mathbf{k}}}{2} \left[1 - (-1)^j z_{\sigma} \frac{M}{S} \right] \quad (14)$$

$$\alpha_{j\sigma}(\mathbf{k}) = \frac{1}{2} \left[1 - (-1)^j z_{\sigma} \frac{M}{S} \right] \times \left[1 + (-1)^j \frac{\epsilon_{\mathbf{k}}}{JS} \left(1 + (-1)^j z_{\sigma} \frac{M}{S} \right) \right] \quad (15)$$

For $\sigma = \uparrow$, $\epsilon(\mathbf{k})^{eff} = \epsilon(\mathbf{k}) \frac{1}{2} (1 \pm \frac{M}{S})$ or $t_{ij}^{eff} = t_{ij} \frac{1}{2} (1 \pm \frac{M}{S})$ which means that the width of the lower sub-band increases with magnetization (either with the decrease of temperature or increase of magnetic field) and the width of the upper sub-band decreases. Similarly as the magnetization increases, the spectral weight is transferred from the upper sub-band to lower sub-band. For down spin, on the other hand, exactly the opposite takes place. This result is similar to the one that follows from the $D = \infty$ model [5,6].

The magnetization M is to be obtained from a selfconsistent equation which can be derived from the free energy in the SDA. It is known from the $D = \infty$ limit of DEM that the thermal behaviour of M is mean field like with an effective exchange $\sim W^2/J$ (W is the band width of the free Bloch band) between the neighbouring spins [15]. In the present work, we study the QDOS with M as a parameter and

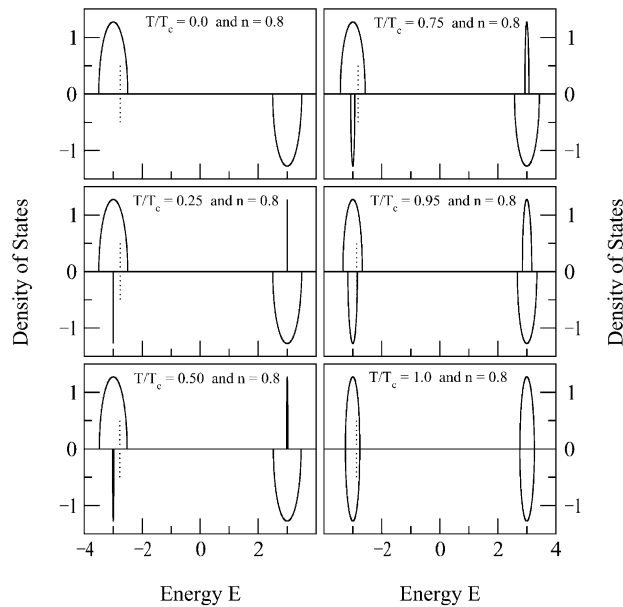


Fig. 1. Quasi particle density of states for various temperatures and band filling $n = (n_{\uparrow} + n_{\downarrow}) = 0.8$. In each picture, the upper half is ρ_{\uparrow} and the lower half is ρ_{\downarrow} . Thin dotted vertical lines show the position of chemical potential μ .

assume that its T -dependence is governed by classical Langevin function with a finite T_c .

The numerical results are obtained for different band occupations with $J/W = 4$, $S = 3/2$. The free Bloch band is taken to be semielliptical and centered at zero. The temperature dependence of the results is given in terms of the reduced temperature T/T_c .

Numerical results have been obtained for different temperatures T and for different band occupations. In Fig. 1, the QDOS of the e_g -band is plotted for different values of T from $T = 0$ ($M/S = 1$) to $T = T_c$ ($M/S = 0$). The chemical potential μ is fixed by the value of n . In Fig. 1, the position of μ is indicated for $n = 0.8$. As is to be expected, QDOS is strongly T -dependent. At $T = 0$, for the lower sub-band, ρ_{\uparrow} has zero width and ρ_{\downarrow} has twice the width of the free Bloch-band. For $n = 0.8$, μ lies in the lower sub-band. Therefore $\rho(\mu)$ has contribution only from spin- \uparrow electrons. This is the situation described in literature as half metal [16–18]. In manganites, the existence of half-metal phase has been established by spin-resolved photoemission studies [19]. As T is increased, ρ_{\downarrow} acquires a finite width in the lower sub-band, but $\rho(\mu)$ still has contribution only from spin- \uparrow electrons. After a certain T , the width of ρ_{\downarrow} becomes sufficiently large so that $\rho(\mu)$ is contributed by spin- \downarrow electrons also. Thus one sees a transition from half-metal to metal at a particular temperature. This temperature should depend on n . Instead of changing the temperature, one can also change the external magnetic field and thereby manipulate the value of M . This also brings about analogous changes in QDOS. Therefore one can also achieve half-metal to metal transition by tuning the magnetic field. Since the change in QDOS and the shift in

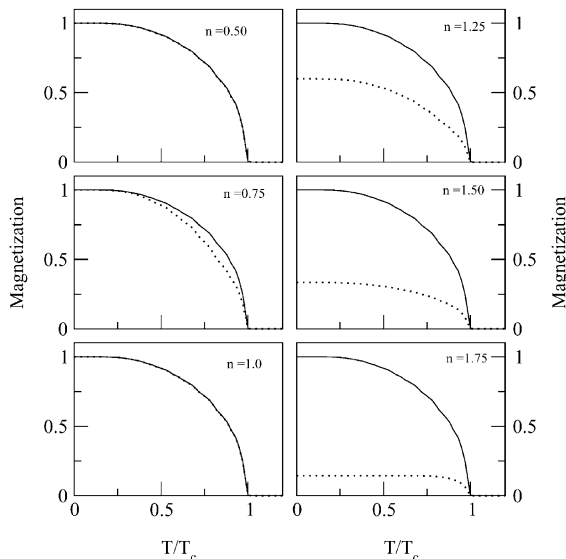


Fig. 2. Local magnetization M/S (solid lines) and band magnetization $m = (n_{\uparrow} - n_{\downarrow})/(n_{\uparrow} + n_{\downarrow})$ (dotted lines) as a function of temperature for various bandfilling $n = (n_{\uparrow} + n_{\downarrow})$.

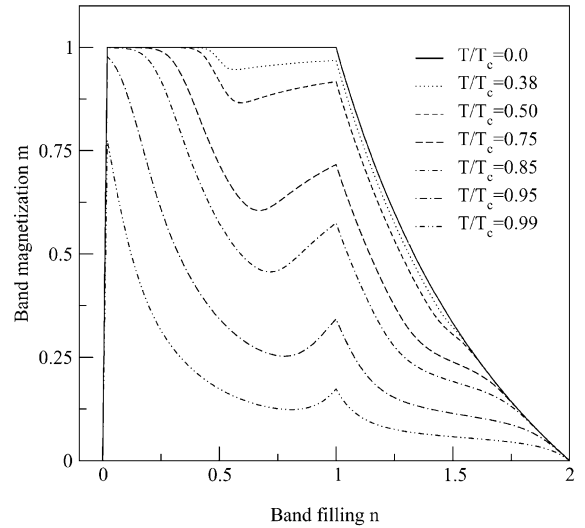


Fig. 3. Band magnetization $m = (n_{\uparrow} - n_{\downarrow})/(n_{\uparrow} + n_{\downarrow})$ as a function of band filling $n = (n_{\uparrow} + n_{\downarrow})$ for different temperatures.

the position of μ are continuous, the transition half-metal to metal is continuous.

In Fig. 2, the band magnetization $m = (n_{\uparrow} - n_{\downarrow})/(n_{\uparrow} + n_{\downarrow})$ and M/S are plotted as functions of T . As already seen, M has a strong influence on QDOS. On the other hand, m depends on n . Therefore it is quite possible that they have different T -dependence. However, in the present approximation, m follows M as a function of T . The only thing is, since m is normalized, its magnitude could be different for different values of n .

In Fig. 3 m is plotted as a function of n for different temperatures. The dependence is clearly nonmonotonic. In order to understand this behaviour, one has to plot QDOS for a particular T and locate μ for different values of n . This is done in Fig. 4 for $T/T_c = 0.75$. For $0 < n < 0.5$, μ lies in the lower sub-band and only spin- \uparrow electrons contribute to m and so $m = 1$. As n increases, μ shifts higher and the spin- \downarrow electrons also contribute and so m decreases up to a certain value of n . Beyond that particular n , μ lies above the spin-down band and with further increase of n the magnetization increases as the extra electrons populate the spin-up band. As a result m again increases with n . For $n > 1$, μ moves into the upper sub-band and again spin- \downarrow electrons contribute resulting in the decrease of m . Finally for $n = 2$, m vanishes.

4. Conclusion

The DEM has been approximately solved using the SDA which is expected to be reliable in the strong coupling limit. A calculation has been performed that it is applicable to manganites. The QDOS of the band states is studied as a function of temperature and for different band-filling. In the

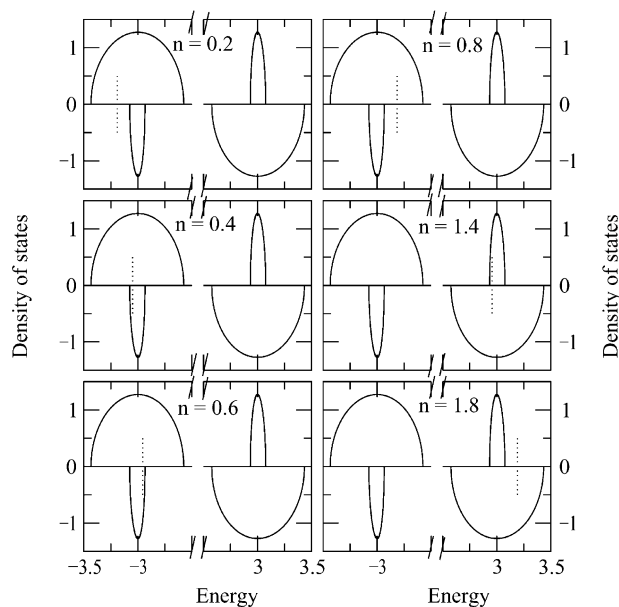


Fig. 4. Quasi particle density of states for various band fillings $n = n_{\uparrow} + n_{\downarrow}$ with $T/T_c = 0.75$. In each picture, the upper half is ρ_{\uparrow} and the lower half is ρ_{\downarrow} . Thin dotted vertical lines show the position of chemical potential μ .

limit of large j , the band splits into two bands with spin-dependent spectral weights. The spontaneous magnetization favours higher spectral weight for the spin-up electrons as compared to that for the spin-down electrons for the lower band. The situation is reversed for the upper band. The model in this approximation shows the possibility of half-metal to metal transition as a function of dopant concentration. The self-energy obtained here is real, which is an artifact of the ansatz made for the spectral density. Therefore, the effect of finite life time of quasiparticles is beyond the scope of the present approximation. The detailed results of the self-consistent solution of magnetization will be reported elsewhere.

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