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Magnetic susceptibility of the double-exchange model

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Abstract. Previously a many-body coherent potential approximation (CPA) was used to study the double-exchange (DE) model with quantum local spins S, both for $S = 1/2$ and for general S in the paramagnetic state. This approximation, exact in the atomic limit, was considered to be a many-body extension of Kubo's one-electron dynamical CPA for the DE model. We now extend our CPA treatment to the case of general S and spin polarization. We show that Kubo's one-electron CPA is always recovered in the empty-band limit and that our CPA is equivalent to dynamical mean-field theory in the classical spin limit. We then solve our CPA equations self-consistently to obtain the static magnetic susceptibility χ in the strong-coupling limit. As in the case of the CPA for the Hubbard model, we find unphysical behaviour in χ at half-filling and no magnetic transition for any finite S. We identify the reason for this failure of our approximation and propose a modification which gives the correct Curie-law behaviour of χ at half-filling and a transition to ferromagnetism for all S.

1. Introduction

Recently there has been much interest in the perovskite manganite compounds $T_{1-x}D_xMnO_3$ where T and D are trivalent and divalent cations respectively. These exhibit a rich variety of phases including charge, orbital, ferromagnetic and antiferromagnetic ordering [1, 2]. Of particular interest is $La_{1-x}Ca_xMnO_3$ with $x \sim 0.3$; in this compound ferromagnetic–paramagnetic and metal–insulator transitions occur together, and for temperatures near the critical temperature an applied magnetic field causes a very large reduction in electrical resistance: this is the phenomenon known as colossal magnetoresistance (CMR).

The physically relevant electrons in the manganites are those from the Mn 3d levels, which are split by the approximately cubic crystal field into triply degenerate t_{2g} levels and higher-energy doubly degenerate e_g levels. Occupied e_g levels are further split into two non-degenerate levels by the Jahn–Teller effect. Electrons from the e_g levels are able to hop between Mn sites via the O atoms, forming a narrow conduction band, but those from the t_{2g} levels are localized. There is a strong Hund's-rule coupling on the Mn sites, so the t_{2g} electrons are usually modelled as local $S = 3/2$ spins ferromagnetically coupled to the itinerant e_g electrons. At $x = 0$ there is one e_g electron per site so the system is a Mott insulator. Doping by D atoms produces holes in the e_g band which enables conduction to occur.

The simplest model for the CMR compounds, which neglects the e_g degeneracy and any coupling to phonon modes, is Zener's [3] double-exchange (DE) model

$$H = \sum_{ij\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} - J \sum_i \boldsymbol{S}_i \cdot \boldsymbol{\sigma}_i - h \sum_i L^z_i \tag{1}$$

where i and j are Mn sites, $c_{j\sigma}$ ($c^\dagger_{i\sigma}$) is a σ-spin conduction electron annihilation (creation) operator, $\boldsymbol{S}_i$ is a local spin operator, $\boldsymbol{\sigma}_i$ is a conduction electron spin operator, $L^z_i = S^z_i + \sigma^z_i$

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is the z-component of the total angular momentum on a site, t_{ij} is the hopping integral with discrete Fourier transform t_k, $J > 0$ is the Hund's-rule coupling constant, and $h = g\mu_B B$ is the Zeeman coupling strength, B being the applied magnetic field. The number of conduction electrons per atom n is assumed to be given by $n = 1 - x$. The idea of the DE model is that hopping of e_g electrons between neighbouring sites is easier if the local spins on the sites are parallel, so an effective ferromagnetic coupling between the local spins is induced by the conduction electrons lowering their kinetic energy. Double-exchange coupling differs from conventional Heisenberg coupling by being (for classical local spins) of the form $\cos(\theta_{ij}/2)$ rather than $\cos(\theta_{ij})$, where θ_{ij} is the angle between the i- and j-site local spins.

According to Millis *et al* [4,5] the CMR effect arises from a competition between double-exchange coupling, which produces a tendency towards the conducting ferromagnetic state, and strong coupling to phonon modes, which tends to localize the electrons via self-trapping. In a previous paper [9] we confirmed that the DE model above cannot account for the very high resistivity of the paramagnetic state. Moreover experiments show that coupling to the crystal lattice is important [6–8]. In this paper however we will complete our study of the simple DE model, aiming to understand the purely electronic properties of CMR systems modelled by (1) before tackling more realistic and complicated models. We concentrate particularly on the magnetic properties.

In [9] we derived an approximation for the one-electron Green function which was based on Hubbard's scattering correction approximation for the Hubbard model [10]. In the Hubbard model this approximation is derived by decoupling the Green-function equations of motion according to an alloy analogy in which electrons of one spin are frozen whilst the Green function for those of the opposite spin is calculated. For the finite-S DE model this approach is complicated by the possibility of dynamic spin scattering—conduction electrons exchanging angular momentum with the local electrons. We obtained an approximation which, like Hubbard's, was exact in the atomic limit for all band filling. Since Hubbard's approximation is equivalent to the coherent potential approximation (CPA) in the alloy analogy for the Hubbard model, and our approximation reduces to a one-electron dynamical CPA due to Kubo [11] in the empty-band limit of the DE model, we regard our approximation as a many-body extension of the CPA.

In [9] we concentrated on simple cases in which the CPA system of equations of motion closed easily, and calculated the electronic structure and resistivity of the paramagnetic state. In section 2 we formulate and solve the general CPA equations. In section 3 we then calculate the static magnetic susceptibility self-consistently within the CPA. In section 4 we compare our CPA in the classical spin limit with dynamical mean-field theory, and in section 5 the CPA is modified so as to improve the behaviour of the susceptibility. A summary and outlook are given in section 6.

2. Solution of the CPA equations

In this section we will use equation of motion (EOM) decoupling approximations to derive an expression for the one-electron Green function $G_\sigma^{ij} = \langle\langle c_{i\sigma}; c_{j\sigma}^\dagger \rangle\rangle$ of the DE model. In a previous paper [9] we obtained G_σ^{ij} in the special cases of zero-field paramagnetism and saturated ferromagnetism for all values of S, but only considered the case of arbitrary magnetization for $S = 1/2$. Here we extend this previous treatment to the case of general S and spin polarization. The decoupling approximations used are direct extensions of those used in [9], which were in turn generalizations of Hubbard's scattering correction approximation [10] for the Hubbard model.

We split the Green functions into components $G_\sigma^{ij} = G_\sigma^{ij+} + G_\sigma^{ij-}$ where $G_\sigma^{ij\alpha}$ describes propagation via singly ($\alpha = -$) and doubly ($\alpha = +$) occupied sites: $G_\sigma^{ij\alpha} = \langle\langle n_i^\alpha c_{i\sigma}; c_{j\sigma}^\dagger\rangle\rangle$ where $n_i^+ = n_i$, $n_i^- = 1 - n_i$, $n_{i\sigma}^+ = n_{i\sigma}$, and $n_{i\sigma}^- = 1 - n_{i\sigma}$. Here $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$ and $n_i = n_{i\uparrow} + n_{i\downarrow}$. Now the cases considered in [9] were chosen so that the system of EOMs could be closed using only the Green functions $G_\sigma^{ij\alpha}$, $\langle\langle S_i^z n_i^\alpha c_{i\sigma}; c_{j\sigma}^\dagger\rangle\rangle$, and $\langle\langle S_i^{-\sigma} n_i^\alpha c_{i-\sigma}; c_{j\sigma}^\dagger\rangle\rangle$ where $S_i^{-\sigma} = S_i^-$ and S_i^+ for $\sigma = \uparrow, \downarrow$ respectively; here we will also need the Green functions $\langle\langle (S_i^z)^m n_i^\alpha c_{i\sigma}; c_{j\sigma}^\dagger\rangle\rangle$ and $\langle\langle (S_i^z)^{m-1} S_i^{-\sigma} n_i^\alpha c_{i-\sigma}; c_{j\sigma}^\dagger\rangle\rangle$ for $m > 1$ in order to close the system. It is simplest to work with the Green functions

$$S_\sigma^{ij\alpha}(\lambda) = \left\langle\!\!\left\langle \mathrm{e}^{\lambda S_i^z} n_i^\alpha c_{i\sigma}; c_{j\sigma}^\dagger \right\rangle\!\!\right\rangle \tag{2a}$$

$$T_\sigma^{ij\alpha}(\lambda) = \left\langle\!\!\left\langle \mathrm{e}^{\lambda S_i^z} S_i^{-\sigma} n_i^\alpha c_{i-\sigma}; c_{j\sigma}^\dagger \right\rangle\!\!\right\rangle \tag{2b}$$

which are in generating function form so that

$$\partial^m S_\sigma^{ij\alpha}/\partial\lambda^m|_{\lambda=0} = \langle\langle (S_i^z)^m n_i^\alpha c_{i\sigma}; c_{j\sigma}^\dagger\rangle\rangle$$

and

$$\partial^{m-1} T_\sigma^{ij\alpha}/\partial\lambda^{m-1}|_{\lambda=0} = \langle\langle (S_i^z)^{m-1} S_i^- n_i^\alpha c_{i\downarrow}; c_{j\sigma}^\dagger\rangle\rangle.$$

For notational simplicity we will work for $\sigma = \uparrow$; the $\sigma = \downarrow$ EOM can be obtained by making the replacements $c_{i\sigma} \mapsto c_{i-\sigma}$, $c_{i\sigma}^\dagger \mapsto c_{i-\sigma}^\dagger$, $S_i^z \mapsto -S_i^z$, $S_i^\pm \mapsto S_i^\mp$, $h \mapsto -h$, and $\lambda \mapsto -\lambda$. Recall that with the fermionic definition of Green functions,

$$\langle\langle A; C\rangle\rangle_\epsilon = -\mathrm{i}\int_0^\infty \mathrm{d}t\ \exp(\mathrm{i}\epsilon t)\langle\{A(t), C\}\rangle$$

the EOM is

$$\epsilon\,\langle\langle A; C\rangle\rangle_\epsilon = \langle\{A, C\}\rangle + \langle\langle [A, H]; C\rangle\rangle_\epsilon\,. \tag{3}$$

We use the fact that $S_\sigma^{ij\alpha}(\lambda)$ and $T_\sigma^{ij\alpha}(\lambda)$ are in generating function form to write their exact EOMs in the form

$$\begin{aligned}\left(\epsilon + \frac{h}{2} + \frac{J}{2}\frac{\partial}{\partial\lambda}\right) S_\uparrow^{ij\alpha}(\lambda,\epsilon) + \frac{J}{2}\mathrm{e}^{\lambda\delta_{\alpha+}} T_\uparrow^{ij\alpha}(\lambda,\epsilon) \\ = \delta_{ij}\left\langle \mathrm{e}^{\lambda S^z} n_\downarrow^\alpha\right\rangle + \sum_k t_{ik}\left\langle\!\!\left\langle \mathrm{e}^{\lambda S_i^z} n_{i\downarrow}^\alpha c_{k\uparrow}; c_{j\uparrow}^\dagger\right\rangle\!\!\right\rangle_\epsilon + \left\langle\!\!\left\langle \mathrm{e}^{\lambda S_i^z}\left[n_{i\downarrow}^\alpha, H_0\right] c_{i\uparrow}; c_{j\uparrow}^\dagger\right\rangle\!\!\right\rangle_\epsilon\end{aligned} \tag{4}$$

for $S_\uparrow^{ij\alpha}(\lambda,\epsilon)$ and

$$\begin{aligned}\left(\epsilon + \frac{h}{2} - \frac{J}{2}\left(\delta_{\alpha-} + \frac{\partial}{\partial\lambda}\right)\right) T_\uparrow^{ij\alpha}(\lambda,\epsilon) + \frac{J}{2}\mathrm{e}^{-\lambda\delta_{\alpha+}}\left(S(S+1) + \alpha\,\frac{\partial}{\partial\lambda} - \frac{\partial^2}{\partial\lambda^2}\right) S_\uparrow^{ij\alpha}(\lambda,\epsilon) \\ = -\alpha\delta_{ij}\left\langle \mathrm{e}^{\lambda S^z} S^- \sigma^+\right\rangle + \sum_k t_{ik}\left\langle\!\!\left\langle \mathrm{e}^{\lambda S_i^z} S_i^- n_{i\uparrow}^\alpha c_{k\downarrow}; c_{j\uparrow}^\dagger\right\rangle\!\!\right\rangle_\epsilon \\ + \left\langle\!\!\left\langle \mathrm{e}^{\lambda S_i^z} S_i^-\left[n_{i\uparrow}^\alpha, H_0\right] c_{i\downarrow}; c_{j\uparrow}^\dagger\right\rangle\!\!\right\rangle_\epsilon\end{aligned} \tag{5}$$

for $T_\uparrow^{ij\alpha}(\lambda,\epsilon)$. Here

$$H_0 = \sum_{ij\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma}$$

is the kinetic part of the Hamiltonian and we have dropped the site indices in the expectations, assuming the system to be in a homogeneous phase (i.e. we will not consider antiferromagnetism). If one works directly with EOMs for the Green functions

$\langle\langle (S_i^z)^m n_i^\alpha c_{i\sigma}; c_{j\sigma}^\dagger \rangle\rangle$ and $\langle\langle (S_i^z)^{m-1} S_i^- n_i^\alpha c_{i\downarrow}; c_{j\sigma}^\dagger \rangle\rangle$, as in [9], one is faced with $4S+1$ algebraic equations for general S. Here, using the generating functions, these algebraic equations are reduced to just two differential equations where differentiation with respect to λ corresponds to the coupling between the different algebraic equations.

These EOMs form a closed system apart from the undetermined Green functions on the right-hand sides which correspond to the effects of hopping. The decoupling procedure entails making approximations for these kinetic terms which close the system of equations; since these terms are proportional to t this procedure is exact in the atomic limit $t_{ij} \to 0$. As mentioned in the introduction the idea of the alloy analogy is to neglect the effects of the kinetic part H_0 of the Hamiltonian on electrons of one spin whilst considering the propagation of an electron of the opposite spin. Accordingly we neglect the final terms of (4) and (5) since the occupation number operators in these Green functions are considered to be frozen in time. It remains to find closed approximations for

$$\sum_k t_{ik} \langle\langle \exp(\lambda S_i^z) n_{i\downarrow}^\alpha c_{k\uparrow}; c_{j\uparrow}^\dagger \rangle\rangle_\epsilon$$

and

$$\sum_k t_{ik} \langle\langle \exp(\lambda S_i^z) S_i^- n_{i\uparrow}^\alpha c_{k\downarrow}; c_{j\uparrow}^\dagger \rangle\rangle_\epsilon.$$

The derivation of scattering correction approximations for these terms is identical to that of [9] apart from the occurrence of the factor $\exp(\lambda S_i^z)$ and will not be repeated here. In fact we use

$$\sum_k t_{ik} \left\langle\!\!\left\langle e^{\lambda S_i^z} n_{i\downarrow}^\alpha c_{k\uparrow}; c_{j\uparrow}^\dagger \right\rangle\!\!\right\rangle_\epsilon$$

$$\approx \langle e^{\lambda S^z} n_\downarrow^\alpha \rangle \sum_k t_{ik} \left\langle\!\!\left\langle c_{k\uparrow}; c_{j\uparrow}^\dagger \right\rangle\!\!\right\rangle_\epsilon + J_\uparrow(\epsilon) \left\langle\!\!\left\langle (e^{\lambda S_i^z} n_{i\downarrow}^\alpha - \langle e^{\lambda S^z} n_\downarrow^\alpha \rangle) c_{i\uparrow}; c_{j\uparrow}^\dagger \right\rangle\!\!\right\rangle_\epsilon \tag{6a}$$

$$= \langle e^{\lambda S^z} n_\downarrow^\alpha \rangle \left(\sum_k t_{ik} G_\uparrow^{kj}(\epsilon) - J_\uparrow(\epsilon) G_\uparrow^{ij}(\epsilon) \right) + J_\uparrow(\epsilon) S_\uparrow^{ij\alpha}(\lambda, \epsilon) \tag{6b}$$

$$\sum_k t_{ik} \left\langle\!\!\left\langle e^{\lambda S_i^z} S_i^- n_{i\uparrow}^\alpha c_{k\downarrow}; c_{j\uparrow}^\dagger \right\rangle\!\!\right\rangle_\epsilon$$

$$\approx -\alpha \langle e^{\lambda S^z} S^- \sigma^+ \rangle \sum_k t_{ik} G_\uparrow^{kj}(\epsilon) + J_\downarrow(\epsilon + h) \left\langle\!\!\left\langle e^{\lambda S_i^z} S_i^- n_{i\uparrow}^\alpha c_{i\downarrow}; c_{j\uparrow}^\dagger \right\rangle\!\!\right\rangle_\epsilon$$

$$+ J_\uparrow(\epsilon) \left\langle\!\!\left\langle \alpha \langle e^{\lambda S^z} S^- \sigma^+ \rangle c_{i\uparrow}; c_{j\uparrow}^\dagger \right\rangle\!\!\right\rangle_\epsilon \tag{6c}$$

$$= -\alpha \langle e^{\lambda S^z} S^- \sigma^+ \rangle \left(\sum_k t_{ik} G_\uparrow^{kj}(\epsilon) - J_\uparrow(\epsilon) G_\uparrow^{ij}(\epsilon) \right) + J_\downarrow(\epsilon + h) T_\uparrow^{ij\alpha}(\lambda, \epsilon) \tag{6d}$$

where $J_\sigma(\epsilon) = \epsilon - \Sigma_\sigma(\epsilon) - G_\sigma(\epsilon)^{-1}$. Here $\Sigma_\sigma(\epsilon)$ is the self-energy, local within this approximation, and $G_\sigma(\epsilon) = G_\sigma^{ii}(\epsilon)$ is the local component of the Green function. $J_\sigma(\epsilon)$ contains the effects of coherent propagation of the electron as a σ-spin from site i back to site i via paths avoiding the site at intermediate stages [12]. Note that if an $\uparrow$-spin electron of energy ϵ becomes a $\downarrow$-spin by exchanging angular momentum with a local spin it must then propagate at energy $\epsilon + h$—hence the occurrence of $J_\downarrow(\epsilon + h)$ in (6*c*) and (6*d*) above. It may be seen that approximations (6*a*) and (6*c*) close the system of equations (4) and (5). No further approximations are made.

For convenience we now define $E_\sigma(\epsilon) = \epsilon - J_\sigma(\epsilon)$, which will later be related to the Weiss function of dynamical mean-field theory [13], $E_\sigma^h(\epsilon) = E_\sigma(\epsilon + h\delta_{\sigma\downarrow}) + \sigma h/2$, which

puts the energy shift effects of the magnetic field into E_σ, and

$$\lambda_\sigma^{ij}(\epsilon) = \delta_{ij} + \sum_k t_{ik} G_\sigma^{kj}(\epsilon) - J_\sigma(\epsilon) G_\sigma^{ij}(\epsilon). \tag{7}$$

We make the above approximations so that (4) and (5) become

$$\begin{bmatrix} E_\uparrow^h(\epsilon) + (J/2)\dfrac{\partial}{\partial\lambda} & (J/2)\mathrm{e}^{\lambda\delta_{\alpha+}} \\ (J/2)\mathrm{e}^{-\lambda\delta_{\alpha+}}\left[S(S+1) + \alpha\dfrac{\partial}{\partial\lambda} - \dfrac{\partial^2}{\partial\lambda^2}\right] & E_\downarrow^h(\epsilon) - (J/2)\left(\delta_{\alpha-} + \dfrac{\partial}{\partial\lambda}\right) \end{bmatrix} \begin{pmatrix} S_\uparrow^{ij\alpha}(\lambda,\epsilon) \\ T_\uparrow^{ij\alpha}(\lambda,\epsilon) \end{pmatrix}$$

$$\approx \lambda_\uparrow^{ij}(\epsilon) \begin{pmatrix} \langle \mathrm{e}^{\lambda S^z} n_\downarrow^\alpha \rangle \\ -\alpha \langle \mathrm{e}^{\lambda S^z} S^- \sigma^+ \rangle \end{pmatrix}. \tag{8}$$

This is a coupled pair of linear differential equations (with respect to λ) of first and second order respectively. The first equation is used to eliminate $T_\uparrow^{ij\alpha}(\lambda,\epsilon)$ in terms of $S_\uparrow^{ij\alpha}(\lambda,\epsilon)$:

$$T_\uparrow^{ij\alpha}(\lambda,\epsilon) = \frac{2}{J}\mathrm{e}^{-\lambda\delta_{\alpha+}}(\lambda_\uparrow^{ij}(\epsilon)\langle \mathrm{e}^{\lambda S^z} n_\downarrow^\alpha \rangle - E_\uparrow^h(\epsilon) S_\uparrow^{ij\alpha}(\lambda,\epsilon) - \frac{J}{2}\frac{\partial}{\partial\lambda} S_\uparrow^{ij\alpha}(\lambda,\epsilon)). \tag{9}$$

Substituting into the second equation we obtain a first-order equation for $S_\uparrow^{ij\alpha}(\lambda,\epsilon)$:

$$\frac{\partial}{\partial\lambda} S_\uparrow^{ij\alpha}(\lambda,\epsilon) + \left(\frac{(J/2)S(S+1) - E_\uparrow^h(\epsilon)((2/J)E_\downarrow^h(\epsilon) + \alpha)}{E_\uparrow^h(\epsilon) - E_\downarrow^h(\epsilon)}\right) S_\uparrow^{ij\alpha}(\lambda,\epsilon)$$

$$= -\left\langle \frac{((2/J)E_\downarrow^h(\epsilon) + \alpha - S^z)\mathrm{e}^{\lambda S^z} n_\downarrow^\alpha + \alpha \mathrm{e}^{\lambda(\delta_{\alpha+} + S^z)} S^- \sigma^+}{E_\uparrow^h(\epsilon) - E_\downarrow^h(\epsilon)} \right\rangle \lambda_\uparrow^{ij}(\epsilon). \tag{10}$$

This equation is of the form

$$\frac{\partial S(\lambda)}{\partial\lambda} + PS(\lambda) = R(\lambda) \tag{11}$$

where P is independent of λ, which has the solution

$$S(\lambda) = C\mathrm{e}^{-P\lambda} + \mathrm{e}^{-P\lambda}\int^\lambda \mathrm{d}\lambda\, \mathrm{e}^{P\lambda} R(\lambda) \tag{12}$$

where C is a constant of integration. Furthermore, by inserting local spin projection operators into definition (2*a*) of $S_\uparrow^{ij\alpha}(\lambda,\epsilon)$ it may be seen that

$$S_\uparrow^{ij\alpha}(\lambda,\epsilon) = \sum_{m=-S}^{S} a_{m\uparrow}^{ij\alpha}(\epsilon)\exp(m\lambda)$$

where $a_{m\uparrow}^{ij\alpha}(\epsilon)$ is independent of λ. Since in general P is not an integer we must have $C = 0$. Hence we find

$$S_\uparrow^{ij\alpha}(\lambda,\epsilon) = \left\langle \frac{(E_\downarrow^h(\epsilon) + (J/2)(\alpha - S^z))\mathrm{e}^{\lambda S^z} n_\downarrow^\alpha - ((J/2)\delta_{\alpha-})\mathrm{e}^{\lambda S^z} S^- \sigma^+}{(E_\uparrow^h(\epsilon) + JS^z/2)(E_\downarrow^h(\epsilon) + (J/2)(\alpha - S^z)) - (J^2/4)(S + \alpha S^z)(S + 1 - \alpha S^z)} \right.$$

$$\left. + \frac{((J/2)\delta_{\alpha+})\mathrm{e}^{\lambda(S^z+1)} S^- \sigma^+}{(E_\uparrow^h(\epsilon) + (J/2)(S^z+1))(E_\downarrow^h(\epsilon) - JS^z/2) - (J^2/4)(S - S^z)(S + 1 + S^z)} \right\rangle$$

$$\times \lambda_\uparrow^{ij}(\epsilon) \tag{13}$$

where we adopt the convention that a quotient of operators, B divided by A, means $A^{-1}B$. Substituting into (9) we also obtain

$$T_{\uparrow}^{ij\alpha}(\lambda,\epsilon) = \left\langle \frac{\mathrm{e}^{\lambda(S^z-\delta_{\alpha+})}((J/2)(S^z+\alpha S)(S^z-\alpha(S+1))n_{\downarrow}^{\alpha}+\delta_{\alpha-}(E_{\uparrow}^{h}(\epsilon)+JS^z/2)S^{-}\sigma^{+})}{(E_{\uparrow}^{h}(\epsilon)+JS^z/2)(E_{\downarrow}^{h}(\epsilon)+(J/2)(\alpha-S^z))-(J^2/4)(S+\alpha S^z)(S+1-\alpha S^z)} \right.$$
$$\left. - \frac{\delta_{\alpha+}(E_{\uparrow}^{h}(\epsilon)+(J/2)(1+S^z))\mathrm{e}^{\lambda S^z}S^{-}\sigma^{+}}{(E_{\uparrow}^{h}(\epsilon)+(J/2)(1+S^z))(E_{\downarrow}^{h}(\epsilon)-JS^z/2)-(J^2/4)(S-S^z)(S+1+S^z)} \right\rangle$$
$$\times \lambda_{\uparrow}^{ij}(\epsilon). \tag{14}$$

It is easy to check that these expressions reduce to those obtained in [9] in the appropriate cases.

We are mostly interested in the local components of the Green functions, $S_{\uparrow}^{\alpha} = S_{\uparrow}^{ii\alpha}$ and $T_{\uparrow}^{\alpha} = T_{\uparrow}^{ii\alpha}$, and in the following we drop site indices. Note that $\lambda_{\sigma}^{ii}(\epsilon) = 1$; this follows from the relation $\lambda_{\sigma}^{ij}(\epsilon) = G_{\sigma}^{ij}(\epsilon)/G_{\sigma}(\epsilon)$ which is easy to obtain from the definition (7) of $\lambda_{\sigma}^{ij}(\epsilon)$ using Fourier transforms and the locality of $\Sigma_{\sigma}(\epsilon)$, as in section 3.1 of [9].

Since E_{σ}^{h} is a functional of G_{σ}, equation (13) with $i = j$ determines G_{σ}, in principle, in terms of the expectations $\langle\exp(\lambda S^z)\rangle$, $\langle\exp(\lambda S^z)n_{\sigma}\rangle$, and $\langle\exp(\lambda S^z)S^{\mp}\sigma^{\pm}\rangle$ as functions of λ. These expectations must be evaluated self-consistently in terms of $S_{\sigma}^{\alpha}(\lambda)$ and $T_{\sigma}^{\alpha}(\lambda)$. The equation for G_{σ} simplifies if we approximate the physical (simple cubic tight-binding) bare density of states (DOS) by an elliptic DOS, $D(\epsilon) = 2\sqrt{W^2-\epsilon^2}/(\pi W)$ where W is the half-bandwidth; for this DOS it may be shown that $E_{\sigma}(\epsilon) = \epsilon - (W^2/4)G_{\sigma}(\epsilon)$, so E_{σ} is an explicit function of G_{σ}. We now define the functional

$$\mathcal{I}[g] = \oint_{\gamma} \frac{\mathrm{d}\epsilon}{2\pi\mathrm{i}}\, f(\epsilon-\mu)g(\epsilon) \tag{15}$$

where f is the Fermi function, μ is the chemical potential, and γ is the anticlockwise contour lying just below and just above the real axis. This functional is useful owing to the usual sum rule

$$\mathcal{I}[\langle\langle A;C\rangle\rangle] = \langle CA\rangle \tag{16}$$

from which it follows (exactly) that

$$\mathcal{I}\left[S_{\sigma}^{\alpha}(\lambda)\right] = \left\langle \mathrm{e}^{\lambda S^z} n_{-\sigma}^{\alpha} n_{\sigma}\right\rangle \tag{17a}$$
$$\mathcal{I}\left[T_{\sigma}^{\alpha}(\lambda)\right] = \left\langle \mathrm{e}^{\lambda S^z} S^{-\sigma}\sigma^{\sigma}\right\rangle \delta_{\alpha-}. \tag{17b}$$

The expectations $\langle\exp(\lambda S^z)n_{\sigma}\rangle$ and $\langle\exp(\lambda S^z)S^{\mp}\sigma^{\pm}\rangle$ can therefore be obtained directly from the sum rule as $\langle\exp(\lambda S^z)n_{\sigma}\rangle = \mathcal{I}[S_{\sigma}(\lambda)]$, where $S_{\sigma}(\lambda) = \sum_{\alpha} S_{\sigma}^{\alpha}(\lambda)$, and $\langle\exp(\lambda S^z)S^{-}\sigma^{+}\rangle = \mathcal{I}[T_{\uparrow}(\lambda)]$. However $\langle\exp(\lambda S^z)\rangle$ must be obtained indirectly, in principle, by solving the system of equations obtained by applying the $\mathcal{I}$-functional to (13) and (14). Knowledge of $\langle\exp(\lambda S^z)\rangle$ is equivalent to knowledge of $P(S^z)$, the probability distribution function for local spins, and it is not clear that this quantity will be accurately obtained from the (approximate) single-electron Green functions that we have considered here. In fact, as will be seen later, the self-consistent determination of this quantity causes problems with our CPA.

We now specialize to three important cases of particular interest: $n = 0$, $J = \infty$, and $S = \infty$, in which (13) and (14) simplify considerably. The results below generalize previous work [9] which was restricted, for general magnetization, to the case $S = 1/2$.

2.1. The empty-band limit

In [11] Kubo used a one-electron dynamical CPA to derive an expression for $G_\uparrow$ valid in the low-density limit $n \to 0$. From (13) with $\lambda = 0$ we calculate $G_\uparrow$ in this limit as

$$G_\uparrow = \left\langle \frac{E^h_\downarrow - (J/2)(1+S^z)}{(E^h_\uparrow + (J/2)S^z)(E^h_\downarrow - (J/2)(1+S^z)) - (J^2/4)(S+1+S^z)(S-S^z)} \right\rangle. \tag{18}$$

This is equivalent to Kubo's equation for $G_\uparrow$ so our decoupling approximation is indeed a many-body extension of the CPA.

2.2. The strong-coupling limit

In the physical systems for which the double-exchange model was introduced, $J \gg t_{ij}$ and $0 < n < 1$. In this situation the chemical potential lies in the lowest band near $-JS/2$, so we shift the energy origin, $E^h_\sigma \mapsto E^h_\sigma - JS/2$, and let $J \to \infty$. Equations (13) and (14) then become

$$S^\alpha_\uparrow(\lambda) = \left\langle e^{\lambda S^z} \frac{(S+1+S^z)n^-_\downarrow + S^-\sigma^+}{(S+1+S^z)E^h_\uparrow + (S-S^z)E^h_\downarrow} \right\rangle \delta_{\alpha-} \tag{19a}$$

$$T^\alpha_\uparrow(\lambda) = \left\langle e^{\lambda S^z}(S-S^z) \frac{(S+1+S^z)n^-_\downarrow + S^-\sigma^+}{(S+1+S^z)E^h_\uparrow + (S-S^z)E^h_\downarrow} \right\rangle \delta_{\alpha-}. \tag{19b}$$

2.3. The classical spin limit

In dynamical mean-field theory (DMFT), a local approximation exact in the infinite-dimensional limit [13], the double-exchange model can be solved exactly in the classical spin limit $S \to \infty$ in which the local spins can be treated as static [14]. Since our approximation is also local it is interesting to compare our results with DMFT in this limit. We let $S \to \infty$ in (13) and (14), scaling J, λ, h, and $T^\alpha_\sigma(\lambda)$ as $1/S$, and obtain

$$S^\alpha_\uparrow(\lambda) = \left\langle e^{\lambda S^z} \frac{(E_\downarrow - (J/2)S^z)n^\alpha_\downarrow + \alpha(J/2)S^-\sigma^+}{(E_\uparrow + (J/2)S^z)(E_\downarrow - (J/2)S^z) - (J^2/4)(1-(S^z)^2)} \right\rangle \tag{20a}$$

$$T^\alpha_\uparrow(\lambda) = \left\langle e^{\lambda S^z} \frac{(J/2)((S^z)^2-1)n^\alpha_\downarrow - \alpha(E_\uparrow + (J/2)S^z)S^-\sigma^+}{(E_\uparrow + (J/2)S^z)(E_\downarrow - (J/2)S^z) - (J^2/4)(1-(S^z)^2)} \right\rangle \tag{20b}$$

where by S^z and S^- we mean S^z/S and S^-/S respectively. Note that in this limit $E^h_\sigma = E_\sigma$. In section 4 below we will derive these Green functions within DMFT for comparison, and it will be found that our CPA agrees with DMFT for S^α_σ and T_σ, but not for $T^\pm_\sigma$.

3. Self-consistent CPA susceptibility

In this section we calculate the static magnetic susceptibility χ of the zero-field paramagnetic state. For simplicity we specialize to the strong-coupling limit $J = \infty$ most favourable to ferromagnetism and use the elliptic bare DOS mentioned in the previous section. We drop the spin suffices on zero-field paramagnetic state quantities and define δA to be the first-order deviation of any quantity A from its value in this state. Thus δA is proportional to the applied magnetic field B or equivalently to $\delta\langle L^z\rangle = \delta\langle S^z + \sigma^z\rangle$. We proceed by calculating $\delta\langle L^z\rangle$ in terms of the Zeeman energy $h = g\mu_B B$ and using

$$\chi = (g\mu_B)^2 \lim_{h\to 0}(\delta\langle L^z\rangle/h).$$

We first derive a couple of useful identities. Equations (19*a*) and (19*b*) imply that $T_\sigma(\lambda) = (S - \partial/\partial\lambda)S_\sigma(\lambda)$. We apply the sum rule (16)–(17*b*) to this relation and, using the fact that all the self-consistently determined expectations are real, obtain the rather obvious results

$$\left\langle \mathrm{e}^{\lambda S^z} \boldsymbol{S}\cdot\boldsymbol{\sigma}\right\rangle = (S/2)\left\langle \mathrm{e}^{\lambda S^z} n\right\rangle \tag{21a}$$

$$\left\langle S^z n\right\rangle = 2S\left\langle \sigma^z\right\rangle. \tag{21b}$$

These serve as a check on our approximation and will later be used to manipulate expectations.

The Green functions are especially simple in the ($J = \infty$) zero-field paramagnetic state: from (19*a*) with $\lambda = 0$ and $\alpha = -$ we have

$$G(\epsilon) = \frac{(S + 1 - n/2)/(2S + 1)}{E(\epsilon)} \tag{22}$$

which corresponds to a band of weight $(S + 1 - n/2)/(2S + 1)$ per spin. In the elliptic DOS case, for which $E(\epsilon) = \epsilon - (W^2/4)G(\epsilon)$, we can easily solve this equation to obtain

$$G(\epsilon) = \frac{2}{W^2}\left(\epsilon - \sqrt{\epsilon^2 - \bar{W}^2}\right) \tag{23}$$

where

$$\bar{W} = W\sqrt{(S + 1 - n/2)/(2S + 1)}$$

is the halfwidth of the renormalized band, which is also elliptic.

We expand the denominator of (19*a*) in powers of S^z:

$$S_\uparrow(\lambda) = \sum_{r=0}^{\infty} \frac{(E^h_\downarrow - E^h_\uparrow)^r}{((S+1)E^h_\uparrow + SE^h_\downarrow)^{r+1}} \frac{\partial^r}{\partial\lambda^r}\left\langle \mathrm{e}^{\lambda S^z}\left((S + 1 + S^z)n^-_\downarrow + S^-\sigma^+\right)\right\rangle. \tag{24}$$

Owing to the presence of the factor $E^h_\downarrow - E^h_\uparrow$, which is zero in the $h = 0$ paramagnetic state, only the $r = 0$ and $r = 1$ terms contribute to $\delta S_\uparrow(\lambda)$. From (24) we find

$$\begin{aligned}\delta S_\uparrow(\lambda) = {} & \frac{\delta E^h_\downarrow(\partial/\partial\lambda - S) - \delta E^h_\uparrow(\partial/\partial\lambda + S + 1)}{(2S+1)^2E^2}\left\langle \mathrm{e}^{\lambda S^z}((S + 1 + S^z)n^-_\downarrow + S^-\sigma^+)\right\rangle \\ & + \frac{\delta\left\langle \mathrm{e}^{\lambda S^z}((S + 1 + S^z)n^-_\downarrow + S^-\sigma^+)\right\rangle}{(2S+1)E}\end{aligned} \tag{25}$$

where the paramagnetic state value, which may easily be evaluated, is used for the first expectation. Now it may be shown that $\delta\mu = 0$, so $\delta\mathcal{I}[A] = \mathcal{I}[\delta A]$ for any A. We calculate $\delta\mathcal{I}[(\partial/\partial\lambda)(S_\uparrow(\lambda) + S_\downarrow(\lambda))]_{\lambda=0}$ from (25), and using (21*a*) and (21*b*) and the sum rule (17*a*) find

$$\begin{aligned}\delta\left\langle S^z(n_\uparrow + n_\downarrow)\right\rangle = 2S\delta\left\langle \sigma^z\right\rangle = {} & \frac{n}{S + 1 - n/2}\left((S+1)\delta\left\langle L^z\right\rangle - (2S+1)\delta\left\langle \sigma^z\right\rangle\right) \\ & + \frac{2S(S + 1 - n/2)}{3(2S+1)^2}\mathcal{I}\left[\frac{\delta E^h_\downarrow - 2(S+1)\,\delta E^h_\uparrow}{E^2}\right].\end{aligned} \tag{26}$$

Since $E^h_\sigma(\epsilon) = \epsilon + h/2 - (W^2/4)G_\sigma(\epsilon + h\delta_{\sigma\downarrow})$ we have

$$\delta E^h_\sigma = h/2 - (W^2/4)(\delta G_\sigma + h\,\delta_{\sigma\downarrow}G') \tag{27}$$

where $G'(\epsilon) = \mathrm{d}G(\epsilon)/\mathrm{d}\epsilon$. By setting $\lambda = 0$ in (25) and using (21*a*), (21*b*), and (27), we obtain

$$\delta G_\sigma = \sigma\frac{\delta\left\langle L^z\right\rangle E + h(\bar{W}/W)^2(SW^2G'/6 - (S + 1/2))}{(2S+1)E^2 - ((2/3)S+1)\bar{W}^2/4}. \tag{28}$$

Note that since $\delta G_\sigma \propto \sigma$, spectral weight is transferred between the different spin bands at constant energy, so $\delta\mu = 0$ as mentioned above. Equation (28) illustrates how $\delta\langle L^z\rangle$, and hence $\delta\langle S^z\rangle$, is determined indirectly in terms of the Green functions rather than by a direct sum rule of the type of (17*a*) and (17*b*).

We use (27) and (28) to eliminate the δE_σ^hs from (26) in terms of h and $\delta\langle L^z\rangle$. By applying $\mathcal{I}$ to (28) we obtain an expression for $\delta\langle\sigma^z\rangle$. We then eliminate $\delta\langle\sigma^z\rangle$ between this equation and (26) and solve the resulting equation for $\chi = (g\mu_B)^2\delta\langle L^z\rangle/h$:

$$\chi = -\frac{(g\mu_B)^2}{2n}\Big((4/3)S(S+1) + ((4/3)S+1)n\Big)\frac{\mathcal{I}[G']}{Q-1} \tag{29a}$$

$$Q = \mathcal{I}\left[\frac{E}{E^2 - \nu^2\bar{W}^2/4}\right] \tag{29b}$$

where $\nu^2 = ((2/3)S+1)/(2S+1)$. Note that for $S = 1/2$ this expression for χ does reduce to the one given (for the $S = 1/2$ case) in [15]. We can simplify Q by changing variables to $z = W^2G(\epsilon)/(2\bar{W})$ so that

$$\mathrm{d}\epsilon = (2\bar{W}/W^2)G'(\epsilon)^{-1}\,\mathrm{d}z = (\bar{W}/2)(1-(2\bar{W}/W^2)^2G(\epsilon)^{-2})\,\mathrm{d}z = (\bar{W}/2)(1-z^{-2})\,\mathrm{d}z$$

and from the functional form (23) of G it may be seen that the contour γ for ϵ becomes $-\gamma'$, the clockwise unit circle, for z. Hence

$$Q = \oint_{\gamma'}\frac{\mathrm{d}z}{2\pi\mathrm{i}}\,\bar{f}\left(\frac{z+z^{-1}}{2} - \bar{\mu}\right)\frac{z-z^{-1}}{z^2-\nu^2} \tag{30}$$

where $\bar{\mu} = \mu/\bar{W}$ and $\bar{f}(\epsilon) = f(\bar{W}\epsilon)$. The same change of variables can be used to show that

$$\mathcal{I}[G'] = -\frac{2\bar{W}}{W^2}\oint_{\gamma'}\frac{\mathrm{d}z}{2\pi\mathrm{i}}\,\bar{f}\left(\frac{z+z^{-1}}{2} - \bar{\mu}\right). \tag{31}$$

Now a second-order transition to ferromagnetism corresponds to a divergence in χ, i.e. $Q = 1$. From (28) with $h = 0$ and (29*b*) it may be seen, using the sum rule, that $\delta\langle L^z\rangle Q/(2S{+}1) = \mathcal{I}[\delta G_\uparrow] = \delta\langle\sigma^z\rangle$ (for $h = 0$). The equation $Q = 1$ for a zero-field magnetic transition is therefore equivalent to the consistency condition $2\delta\langle\sigma^z\rangle = \delta\langle S^z + \sigma^z\rangle/(S+1/2)$, which certainly holds at $n = 1$ but not at $n = 0$. Integral (30) can be evaluated analytically in the limits of zero and infinite temperature where the Fermi function is of a simple form and the results are plotted in figure 1. It is clear that within the CPA there is no magnetic transition for $0 < n < 1$, as is the case for the CPA for the Hubbard model [16]. This appears to be a considerable drawback of our approximation given that the ferromagnetic–paramagnetic phase transition is a major reason for interest in the double-exchange model. We will propose a method for circumventing this problem in section 5.

We now consider the behaviour of χ at $n = 0, 1$. In these cases electron hopping does not occur and the system consists of a lattice of free local moments of magnitude $S' = S$ and $S+1/2$ respectively, so we expect χ to take the Curie-law form, $\chi_C = (g\mu_B)^2\beta S'(S'+1)/3$ where $\beta = (k_BT)^{-1}$ is the inverse temperature. We calculate χ in these cases by expanding the integrals in (30) and (31) in powers of n or $1-n$. At $n = 0$ we find $\chi = \chi_C$ with $S' = S$ as expected, but at $n = 1$ we find $\chi = \chi_C\phi$ (with $S' = S+1/2$) where

$$\phi = \left(\oint_{\gamma'}\mathrm{d}z\ \exp(\bar{\beta}(z+z^{-1})/2)\right)\Big/\left(\oint_{\gamma'}\mathrm{d}z\ \exp(\bar{\beta}(z+z^{-1})/2)(z-z^{-1})/(z^2-\nu^2)\right) \tag{32}$$

and $\bar{\beta} = \bar{W}\beta$. Now $\phi \to 1$ as $\beta \to 0$ so the Curie law is obtained at high temperature, but as $\beta \to \infty$ we find $\phi \to 8S^2/(3(2S+1)(4S+3)) \neq 1$ so Curie-law behaviour does not extend over the whole temperature range. Note that $\phi(\beta = \infty) \neq 1$ even at $S = \infty$ where, as will be

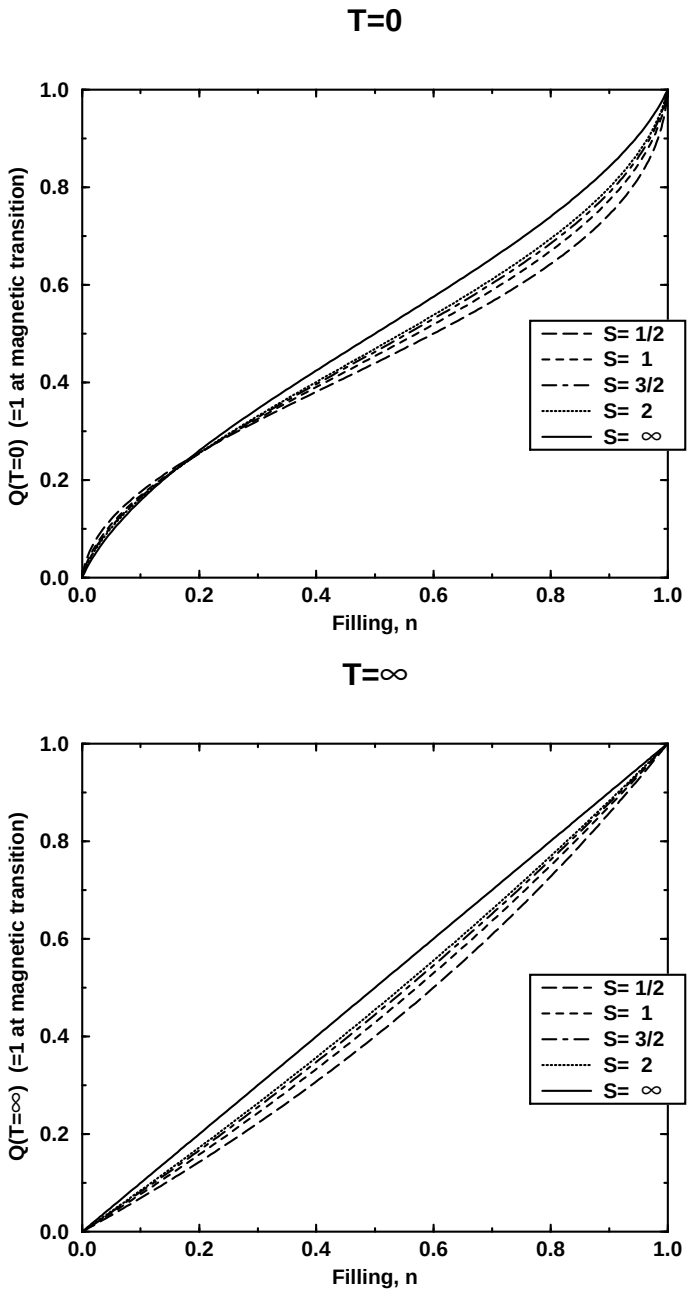

Figure 1. The Q-function versus filling n at temperature $T = 0$ and $T = \infty$.

shown in the next section, our Green-function equations reduce to DMFT. The reasons for this unphysical behaviour, and a way to avoid it, are discussed in section 5. In figure 2 below we compare χ at $n = 1$ and $S = \infty$ with χ_C and the low-temperature asymptote $\chi_C/3$. In figure 3 we plot χ^{-1} at $S = \infty$ and $n = 0.75$, comparing it with the Curie-law and DMFT values, the DMFT plot only being displayed for the paramagnetic phase.

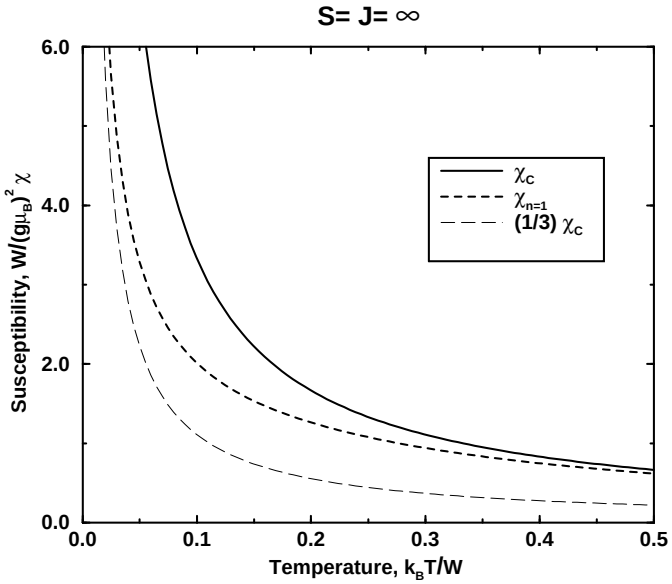

Figure 2. The $S = J = \infty$ magnetic susceptibility χ for $n = 1$ compared with its low-temperature asymptote and the Curie law.

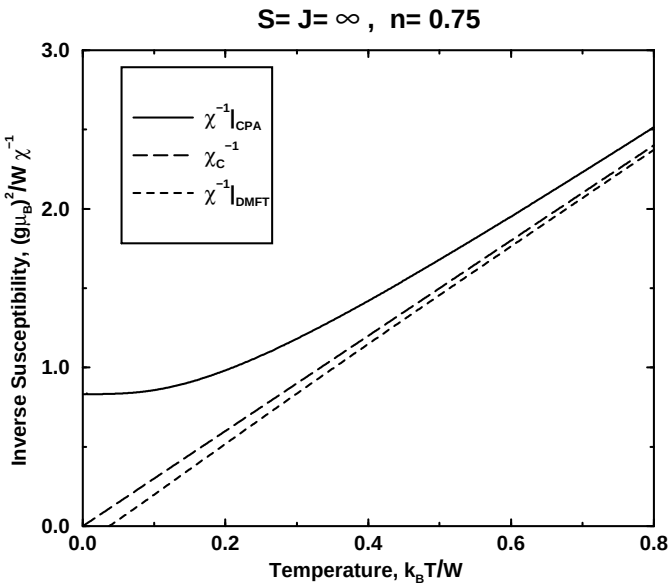

Figure 3. The $S = J = \infty$ inverse magnetic susceptibility χ^{-1} for $n = 0.75$ compared with the Curie-law and DMFT values.

4. Comparison with dynamical mean-field theory at $S = \infty$

In this section we will obtain equations for the S^{α}_{σ} and T^{α}_{σ} Green functions within dynamical mean-field theory (DMFT) at $S = \infty$ and compare them with equations (20*a*) and (20*b*) of

the CPA. The DMFT single-site effective action $\tilde{S}$ of the DE model is [14]

$$\tilde{S} = \int_0^\beta \int_0^\beta \mathrm{d}\tau\ \mathrm{d}\tau'\ \sum_\sigma c_\sigma^\dagger(\tau) E_\sigma(\tau-\tau') c_\sigma(\tau') - \frac{J}{2}\int_0^\beta \mathrm{d}\tau\ \Big[S^z(\tau)\big[n_\uparrow(\tau)-n_\downarrow(\tau)\big] + S^+(\tau)\sigma^-(\tau) + S^-(\tau)\sigma^+(\tau)\Big] - h\int_0^\beta \mathrm{d}\tau\ \Big[S^z(\tau)+\sigma^z(\tau)\Big] + \mathrm{i}\int_0^\beta \mathrm{d}\tau\ S^z\tau\,\partial_\tau \tan^{-1}\Big(S^y(\tau)/S^x(\tau)\Big) \tag{33}$$

where E_σ describes the self-consistently determined coupling with the conduction electron bath. Here $c_\sigma^\dagger(\tau)$ and $c_\sigma(\tau)$ are Grassmann variables [17]. Now DMFT for the DE model is exactly solvable in the classical spin limit $S\to\infty$ where $\boldsymbol{S}(\tau)$ becomes τ-independent (we scale J and h as $1/S$), since for $\boldsymbol{S}$ constant $\tilde{S}$ is diagonal in the Matsubara frequency representation:

$$\tilde{S} = -\sum_n \begin{pmatrix} c_{n\uparrow}^\dagger & c_{n\downarrow}^\dagger \end{pmatrix} \begin{bmatrix} E_{n\uparrow} + (J/2)S^z & (J/2)S^- \\ (J/2)S^+ & E_{n\downarrow} - (J/2)S^z \end{bmatrix} \begin{pmatrix} c_{n\uparrow} \\ c_{n\downarrow} \end{pmatrix} - \beta h S^z \tag{34a}$$

$$= \sum_n \begin{pmatrix} c_{n\uparrow}^\dagger & c_{n\downarrow}^\dagger \end{pmatrix} A_n \begin{pmatrix} c_{n\uparrow} \\ c_{n\downarrow} \end{pmatrix} - \beta h S^z = \sum_n \tilde{S}_n - \beta h S^z \tag{34b}$$

defining the matrix A_n and the components $\tilde{S}_n$ of $\tilde{S}$. Here

$$E_{n\sigma} = -\int_0^\beta \mathrm{d}\tau\ \exp(\mathrm{i}\omega_n\tau) E_\sigma(\tau)$$

and ω_n is a fermionic Matsubara frequency. Note that as shown in [13] the self-consistency condition for $E_{n\sigma}$ can be written as $E_{n\sigma} = \Sigma(\mathrm{i}\omega_n) + G(\mathrm{i}\omega_n)^{-1}$, so $E_{n\sigma}$ is just our quantity $E_\sigma(\mathrm{i}\omega_n)$.

In terms of $\tilde{S}$ the partition function Z is given by

$$Z = \int \mathrm{d}^2S \int \Big(\prod_{n\sigma} \mathrm{d}c_{n\sigma}^\dagger\ \mathrm{d}c_{n\sigma}\Big) \mathrm{e}^{-\tilde{S}} = \int \mathrm{d}^2S\ \mathrm{e}^{\beta h S^z} \prod_n \int \Big(\prod_\sigma \mathrm{d}c_{n\sigma}^\dagger\ \mathrm{d}c_{n\sigma}\Big) \mathrm{e}^{-\tilde{S}_n} \tag{35}$$

where $\int \mathrm{d}^2S$ is the integral over the local spin direction. All site-diagonal correlation functions can be calculated explicitly in terms of the $E_{n\sigma}$s, for example the one-electron Green function is given by

$$\Big\langle\Big\langle c_{i\uparrow}; c_{i\uparrow}^\dagger \Big\rangle\Big\rangle_{\mathrm{i}\omega_n} = -\Big\langle c_{n\uparrow} c_{n\uparrow}^\dagger \Big\rangle = -\frac{1}{Z}\int \mathrm{d}^2S \Big(\prod_{m\sigma} \mathrm{d}c_{m\sigma}^\dagger\ \mathrm{d}c_{m\sigma}\Big) \mathrm{e}^{-\tilde{S}} c_{n\uparrow} c_{n\uparrow}^\dagger. \tag{36}$$

It is convenient to work with the generating function

$$Z_n = \int \Big(\prod_\sigma \mathrm{d}c_{n\sigma}^\dagger\ \mathrm{d}c_{n\sigma}\Big) \exp(-\tilde{S}_n + \sum_\sigma (\eta_{n\sigma}^\dagger c_{n\sigma} + c_{n\sigma}^\dagger \eta_{n\sigma})) \tag{37a}$$

$$= \det(A_n) \exp\left[\begin{pmatrix} \eta_{n\uparrow}^\dagger & \eta_{n\downarrow}^\dagger \end{pmatrix} A_n^{-1} \begin{pmatrix} \eta_{n\uparrow} \\ \eta_{n\downarrow} \end{pmatrix}\right]. \tag{37b}$$

In terms of Z_n the partition function

$$Z = \int \mathrm{d}^2S\ \exp(\beta h S^z) \Big(\prod_n Z_n\Big)_{\eta'=\eta^\dagger=0} = \int \mathrm{d}^2S\ \exp(\beta h S^z) \prod_n \det(A_n)$$

and the local spin probability distribution function

$$P(\boldsymbol{S}) = Z^{-1} \exp(\beta h S^z) \Big(\prod_n Z_n\Big)_{\eta'=\eta^\dagger=0}.$$

Note that

$$\langle U(\boldsymbol{S})\rangle = \int \mathrm{d}^2 S\ P(\boldsymbol{S})U(\boldsymbol{S})$$

for any U.

Explicitly, our full Green functions are given in $S=\infty$ DMFT by

$$S_\uparrow(\lambda, \mathrm{i}\omega_n) = \frac{1}{Z}\int \mathrm{d}^2 S\ \mathrm{e}^{(\lambda+\beta h)S^z}\left(\frac{\mathrm{d}^2}{\mathrm{d}\eta^\dagger_{n\uparrow}\,\mathrm{d}\eta_{n\uparrow}}\prod_{n'} Z_{n'}\right)_{\eta'=\eta^\dagger=0} \tag{38a}$$

$$T_\uparrow(\lambda, \mathrm{i}\omega_n) = \frac{1}{Z}\int \mathrm{d}^2 S\ \mathrm{e}^{(\lambda+\beta h)S^z} S^-\left(\frac{\mathrm{d}^2}{\mathrm{d}\eta^\dagger_{n\downarrow}\,\mathrm{d}\eta_{n\uparrow}}\prod_{n'} Z_{n'}\right)_{\eta'=\eta^\dagger=0} \tag{38b}$$

and these expressions are easily evaluated to give

$$S_\uparrow(\lambda, \mathrm{i}\omega_n) = \left\langle \mathrm{e}^{\lambda S^z}\frac{E_{n\downarrow}-(J/2)S^z}{(E_{n\uparrow}+(J/2)S^z)(E_{n\downarrow}-(J/2)S^z)-(J^2/4)(1-(S^z)^2)}\right\rangle \tag{39a}$$

$$T_\uparrow(\lambda, \mathrm{i}\omega_n) = -\frac{J}{2}\left\langle \mathrm{e}^{\lambda S^z}\frac{1-(S^z)^2}{(E_{n\uparrow}+(J/2)S^z)(E_{n\downarrow}-(J/2)S^z)-(J^2/4)(1-(S^z)^2)}\right\rangle. \tag{39b}$$

Summing the $S=\infty$ CPA expressions (20*a*) and (20*b*) over α we obtain the analytic continuations (from the Matsubara frequencies) of (39*a*) and (39*b*) respectively, i.e. in the classical spin limit our CPA agrees with DMFT for $S_\sigma(\lambda)$ and $T_\sigma(\lambda)$. This is important since DMFT is known to be exact for dimension $D=\infty$ [13] and is perhaps the most natural local approximation for D finite.

Similarly we can calculate the $\alpha=\pm$ components of these Green functions within DMFT as

$$S^+_\uparrow(\lambda, \mathrm{i}\omega_n) = \frac{1}{\beta}\sum_{n'}\left\langle \mathrm{e}^{\lambda S^z}\frac{(E_{n\downarrow}-(J/2)S^z)(E_{n'\uparrow}+(J/2)S^z)-(J^2/4)(1-(S^z)^2)}{\det(A_n)\det(A_{n'})}\right\rangle \tag{40a}$$

$$T^+_\uparrow(\lambda, \mathrm{i}\omega_n) = \frac{J}{2}\frac{1}{\beta}\sum_{n'}\left\langle \mathrm{e}^{\lambda S^z}(1-(S^z)^2)\frac{E_{n\downarrow}-E_{n'\downarrow}}{\det(A_n)\det(A_{n'})}\right\rangle \tag{40b}$$

and $S^-_\uparrow = S_\uparrow - S^+_\uparrow$, etc. Now for a function $g(z)$ analytic off the real axis

$$\beta^{-1}\sum_n g(\mathrm{i}\omega_n) = \mathcal{I}[g]$$

so applying this to the n'-summations in (40*a*) we find

$$S^+_\uparrow(\lambda, \mathrm{i}\omega_n) = \left\langle \mathrm{e}^{\lambda S^z}\frac{(E_{n\downarrow}-JS^z/2)(I_{1\uparrow}(S^z)+JS^zI_2(S^z)/2)-(J^2/4)(1-(S^z)^2)I_2(S^z)}{(E_{n\uparrow}+(J/2)S^z)(E_{n\downarrow}-(J/2)S^z)-(J^2/4)(1-(S^z)^2)}\right\rangle \tag{41}$$

where

$$I_{1\uparrow}(S^z) = \mathcal{I}\left[\frac{E_\uparrow}{(E_\uparrow+(J/2)S^z)(E_\downarrow-(J/2)S^z)-(J^2/4)(1-(S^z)^2)}\right] \tag{42a}$$

$$I_2(S^z) = \mathcal{I}\left[\frac{1}{(E_\uparrow+(J/2)S^z)(E_\downarrow-(J/2)S^z)-(J^2/4)(1-(S^z)^2)}\right]. \tag{42b}$$

It may be shown using the sum rules (17*a*) and (17*b*) that

$$\langle (S^z)^m(I_{1\uparrow}(S^z)+JS^zI_2(S^z)/2)\rangle = \langle (S^z)^m n_\downarrow\rangle$$

and

$$(J/2)\langle (S^z)^m(1-(S^z)^2)I_2(S^z)\rangle = -\langle (S^z)^m S^-\sigma^+\rangle$$

for any m, so in (41) we can replace $I_{1\uparrow}(S^z)+JS^zI_2(S^z)/2$ with $n_\downarrow$ and $(J/2)(1-(S^z)^2)I_2(S^z)$ with $-S^-\sigma^+$. A similar procedure can be carried out for (40*b*), and we obtain

$$S^+_\uparrow(\lambda, \mathrm{i}\omega_n) = \left\langle \mathrm{e}^{\lambda S^z} \frac{(E_{n\downarrow}-(J/2)S^z)n_\downarrow + (J/2)S^-\sigma^+}{(E_{n\uparrow}+(J/2)S^z)(E_{n\downarrow}-(J/2)S^z)-(J^2/4)(1-(S^z)^2)} \right\rangle \tag{43a}$$

$$T^+_\uparrow(\lambda, \mathrm{i}\omega_n) = \left\langle \mathrm{e}^{\lambda S^z} \frac{(J/2)((S^z)^2-1)n_\uparrow - (E_{n\downarrow}-(J/2)S^z)S^-\sigma^+}{(E_{n\uparrow}+(J/2)S^z)(E_{n\downarrow}-(J/2)S^z)-(J^2/4)(1-(S^z)^2)} \right\rangle. \tag{43b}$$

It may be seen that the DMFT and CPA expressions for S^+_σ agree, but $T^+_\uparrow(\lambda)|_{\mathrm{DMFT}} = T^+_\downarrow(\lambda)|_{\mathrm{CPA}}$. This discrepancy, due to failure of the decoupling approximations made in the equation of motion for T^α_σ [9], vanishes in the important limit $J\to\infty$ of strong coupling where all $\alpha = +$ Green functions are zero.

5. The Curie temperature

Furukawa [14] finds that the $S = J = D = \infty$ DE model exhibits a transition to ferromagnetism, and in section 4 it was shown that our CPA gives exact expressions for the Green-function equations in this limit. However, in section 3 we showed that there is no magnetic transition in our CPA (figure 1) and found unphysical behaviour at $n = 1$ (figure 2). In this section we resolve this apparent discrepancy and postulate a modification of our CPA that restores magnetic behaviour. We again work in the elliptic DOS case where $E_{n\sigma} = \mathrm{i}\omega_n - (W^2/4)G_\sigma(\mathrm{i}\omega_n)$.

We first derive the $S = J = \infty$ DMFT static susceptibility χ. In this case

$$P(\boldsymbol{S}) = \left(\exp(\beta h S^z)\prod_n[(1+S^z)E_{n\uparrow}+(1-S^z)E_{n\downarrow}]\right) \times \left(\int \mathrm{d}^2S\ \exp(\beta h S^z)\prod_n[(1+S^z)E_{n\uparrow}+(1-S^z)E_{n\downarrow}]\right)^{-1} \tag{44}$$

as was shown in the previous section. The first-order deviation $\delta P(\boldsymbol{S})$ of $P(\boldsymbol{S})$ from its zero-field paramagnetic state value $(1/4\pi)$ is given by

$$\delta P(\boldsymbol{S}) = \left(\beta h + \sum_n \frac{\delta E_n}{E_n}\right)\frac{S^z}{4\pi} \tag{45}$$

where E_n is the zero-field paramagnetic state value and δE_n is the first-order deviation of $E_{n\uparrow}$. From (28) we have in the present case, where $\bar{W} = W/\sqrt{2}$,

$$\delta G_{n\sigma} = \frac{\sigma E_n\,\delta\langle S^z\rangle}{2E_n^2 - W^2/12}. \tag{46}$$

Substituting (46) into (45) using $\delta E_{n\sigma} = -(W^2/4)\delta G_{n\sigma}$, then multiplying by S^z and integrating over the local spin orientation, we obtain

$$\chi = \frac{g\mu_{\mathrm{B}}\,\delta\langle S^z\rangle}{B} = (g\mu_{\mathrm{B}})^2(\beta/3)\Big/\left(1+(W^2/12)\sum_n(2E_n^2-W^2/12)^{-1}\right) \tag{47}$$

upon rearrangement. This gives the correct Curie law $\chi = (1/3)(g\mu_{\mathrm{B}})^2\beta$ at $n = 0$ and $n = 1$ and a transition to ferromagnetism for all n at a temperature

$$k_{\mathrm{B}}T = -(W^2/12)\mathcal{I}[(2E^2 - W^2/12)^{-1}]$$

where $E(\epsilon) = \epsilon - (W^2/4)G(\epsilon)$ [14].

We now consider where the CPA calculation of χ has gone wrong and how to improve it. The CPA equations for the Green functions are exact in the present case ($S = J = \infty$), but to solve them we need expressions for the expectations $\langle \exp(\lambda S^z) n_\sigma \rangle$, $\langle \exp(\lambda S^z) S^{-\sigma} \sigma^\sigma \rangle$, and $\langle \exp(\lambda S^z) \rangle$, as mentioned in section 2. Since the first two of these are obtained using the $\mathcal{I}$ sum rule, a procedure that is exact, the problem must lie with the determination of $\langle \exp(\lambda S^z) \rangle$. Note that the way that we have calculated $\langle \exp(\lambda S^z) \rangle$ only works for S finite, and to calculate it at $S = \infty$ we have worked for finite S and taken the limit at the end.

Now knowledge of $\langle \exp(\lambda S^z) \rangle$ is equivalent to knowledge of $P(\boldsymbol{S})$, so a possible way of improving the CPA expression for χ is to abandon the above self-consistent determination of $\langle \exp(\lambda S^z) \rangle$ and instead to use some expression for $P(\boldsymbol{S})$ that reduces to the $S = \infty$ DMFT result (44) in the classical spin limit. We have so far been unable to derive such an expression, so we instead postulate a natural extrapolation of (44) to finite S, justifying our formula by the resulting behaviour of χ. This procedure will at least force the CPA for χ to become exact in the $S \to \infty$ limit, and the $S = \infty$ magnetic transition is likely to persist to finite S.

From the $S \to \infty$ limit of (19*a*) and (19*b*) it may be seen that the quantity in square brackets in (44) is related to the denominators of the Green-function equations (for $S = \infty$). A natural extension of (44) is thus

$$P(\boldsymbol{S}) = \left(\exp(\beta h \eta S^z) \prod_n [(1/2 + S + S^z)E_{n\uparrow} + (1/2 + S - S^z)E_{n\downarrow}] \right) \times \left(2\pi \sum_{S^z} \exp(\beta h \eta S^z) \prod_n [(1/2 + S + S^z)E_{n\uparrow} + (1/2 + S - S^z)E_{n\downarrow}] \right)^{-1} \tag{48}$$

where η is chosen so as to optimize the behaviour of χ. The explicit energy shifts associated with the field h in $E_\uparrow$ and $E_\downarrow$ are ambiguous and have been neglected. However, some effect of h on the double exchange enters through the Green functions and the factor η allows for the conduction electron contribution to the spin in the Zeeman energy. Then proceeding as above we can calculate

$$\chi = (g\mu_B)^2 \frac{(1/3)\eta\beta S(S+1) - \beta s \mathcal{I}[G'/E] + (1/2)\mathcal{I}[G']}{1 - Q/(2S+1) + 2\beta s R} \tag{49}$$

where $s = (W^2/12)S(S+1)/(2S+1)$, Q is as in (29*b*), and

$$R = \mathcal{I}\left[\frac{1}{(2S+1)E^2 - ((2/3)S+1)\bar{W}^2/4} \right]. \tag{50}$$

It is then easy to see that if we take $\eta = 1 + (n/2)/(S+1)$ then the correct Curie laws $\chi = (g\mu_B)^2 \beta S(S+1)/3$ and $\chi = (g\mu_B)^2 \beta (S+1/2)(S+3/2)/3$ are obtained at $n = 0$ and $n = 1$ respectively. Note that $S\eta = S + n/2 + O(1/S)$, so if we regard our extrapolation from $S = \infty$ as a kind of $1/S$ expansion this is a very natural value—it corresponds to the average spin size in the system to leading order. With this form (49) for χ we also obtain a magnetic transition for all S at a temperature determined by

$$k_B T = \frac{2sR}{Q/(2S+1) - 1}. \tag{51}$$

The Curie temperature is plotted against filling n for various S in figure 4 (top figure) below. It agrees with Furukawa's result in his case of $S = \infty$. Clearly for finite-S ferromagnetism is more stable for $n > 1/2$ than for $n < 1/2$, in agreement with the findings of Brunton and Edwards [18]. We have also calculated T_C via the spin-wave dispersion in the (assumed)

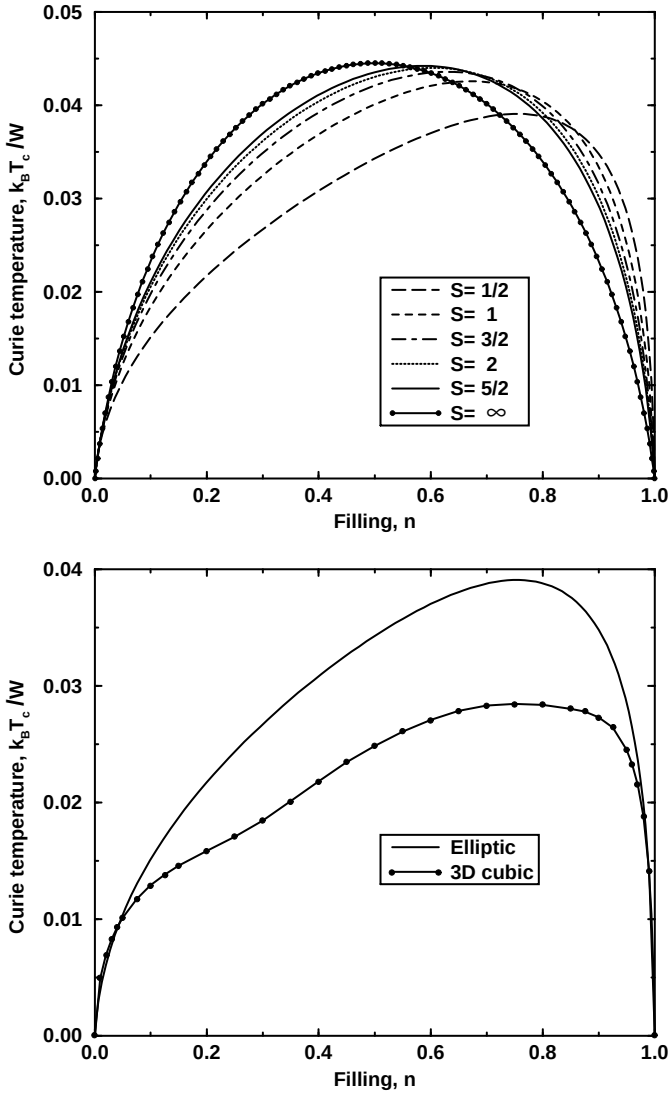

Figure 4. The Curie temperature $k_B T_C / W$ calculated using the elliptic bare DOS plotted against filling n for various S (top figure), and the effect on T_C for $S = 1/2$ of changing the bare DOS to the 3D cubic DOS (bottom figure).

saturated ferromagnetic ground state, using a method similar to that of Sakurai [19, 20] for the Hubbard model. The Curie temperature obtained is similar in magnitude to T_C in figure 4 and decreases with increasing S, as is the case here for n near 1. This work will be published elsewhere.

Brunton and Edwards found that the stability of the spin-saturated state at $T = 0$ is strongly dependent on the bare DOS used: approximating the true cubic tight-binding DOS with the elliptic DOS qualitatively changed the form of their spin-flip excitation gap. Accordingly we check the effect on T_C of using the true tight-binding DOS. The bare elliptic and cubic tight-binding DOSs and the corresponding full (zero-field paramagnetic state, $S = 1$, $n = 1/2$, and $J = \infty$) CPA DOSs are shown for comparison in figure 5 below. Now it is straightforward to

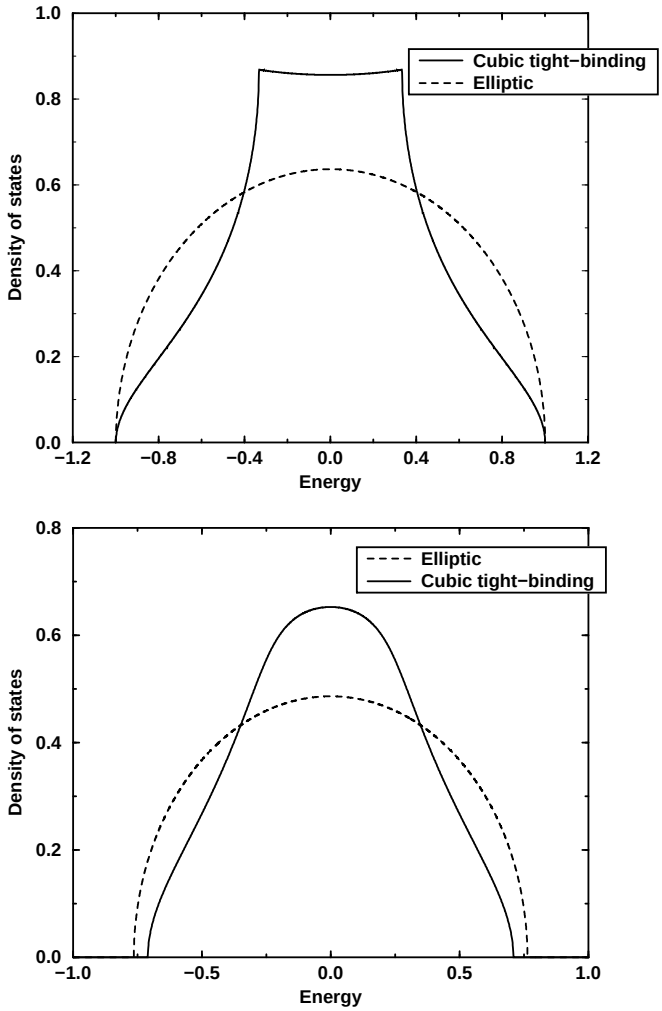

Figure 5. Bare (top figure) and full CPA (bottom figure, for the $S = 1$, $n = 1/2$, $J = \infty$, $W = 1$ zero-field paramagnetic state) DOSs.

extend the derivation of the T_C equation to the case of a general DOS; the only effect on (51) is to replace $W^2/4$ with $(\bar{W}/(WG))^2 + 1/G'$ inside the $\mathcal{I}$-functionals. Hence for general DOS

$$k_B T_C = \frac{2}{3}\frac{S(S+1)}{2S+1}\frac{\tilde{R}}{\tilde{Q}/(2S+1) - 1} \tag{52}$$

where

$$\tilde{R} = \mathcal{I}\left[\frac{(\bar{W}/(WG))^2 + 1/G'}{(2S+1)E^2 - ((2/3)S+1)(\bar{W}^2/W^2)((\bar{W}/(WG))^2 + 1/G')}\right] \tag{53a}$$

$$\tilde{Q} = \mathcal{I}\left[\frac{E}{E^2 - \nu^2(\bar{W}^2/W^2)((\bar{W}/(WG))^2 + 1/G')}\right]. \tag{53b}$$

Note that for the elliptic DOS case $(\bar{W}/(WG))^2 + 1/G' = W^2/4$. We plot T_C obtained from (52) in the most sensitive case of $S = 1/2$ in figure 4 (bottom figure) for the elliptic and cubic

DOSs. It may be seen that changing the form of the bare DOS does not have a large effect on T_C. The dip in T_C near $n = 0.3$ for the cubic DOS is interesting since near this filling Brunton and Edwards [18] found an instability of the saturated ferromagnetic state for $S = 1/2$.

6. Summary and outlook

In this paper we have extended our many-body CPA treatment [9] of the DE model to the case of general S and magnetization. In our original approach we were faced with $4S + 1$ algebraic equations to solve for the Green functions in the case of non-zero magnetization. A correspondingly large number of correlation functions had to be determined self-consistently. Consequently in [9] we only considered $S = 1/2$ for the magnetized state and subsequently [15] calculated the paramagnetic susceptibility in this case. The generalization to arbitrary S in section 2 of this paper is achieved by introducing generating Green functions involving a parameter λ. The $4S + 1$ coupled algebraic equations are then replaced by a single first-order linear differential equation in λ whose solution yields the CPA equations for the Green functions. Only three correlation functions have to be determined, as functions of λ, and two of these may be obtained directly from the Green functions. The indirect determination of the third $\langle \exp(\lambda S^z) \rangle$, from the approximate EOM for the Green functions, is less reliable. It seems to be the origin of difficulties in section 3, where the paramagnetic susceptibility is calculated for $J = \infty$. No ferromagnetic transition is found for any n or S and for $n = 1$ the correct Curie law, with spin $S + 1/2$, is found only at high temperature. On the other hand in section 4 it is shown that for $S = \infty$, where dynamical mean-field theory has been implemented [14], our CPA equations for the Green functions agree with DMFT. Furthermore, DMFT leads to a ferromagnetic transition for $0 < n < 1$ and to a correct Curie law for $n = 1$. In section 5 this paradox is resolved by abandoning the apparently unreliable self-consistent determination of $\langle \exp(\lambda S^z) \rangle$ and using instead a probability distribution $P(S^z)$ to evaluate the required expectation values. The form of $P(S^z)$ used for finite S is a reasonable extension of the form which arises in DMFT for $S = \infty$. We then find a finite Curie temperature T_C for $0 < n < 1$, and correct Curie laws for $n = 0$ and 1, for all S. Naturally the results agree with DMFT for $S = \infty$. The maximum in T_C, as a function of band filling n, moves from $n = 0.5$ for $S = \infty$ to larger values of n as S decreases.

This work completes our present study of the paramagnetic state and ferromagnetic transition of the DE model within our many-body CPA. With some effort we could pursue the calculations into the ferromagnetic state. However, this has already been done for $S = \infty$ within DMFT [14] and the rewards might be slight, particularly since for finite S the CPA never gives a ground state of complete spin alignment. It seems more profitable to repair some defects of the DE model itself. One should include both coupling to phonons and the double degeneracy of the e_g band. It is likely, as originally proposed by Millis *et al* [4], that phonon coupling is essential for an understanding of the insulator-like paramagnetic state in the manganites. We showed [9] that, without phonons, the DE model gives much too small a resistivity. The introduction of phonons is therefore a high priority and it is in fact easier to include coupling to local phonons in our CPA approach than to consider degenerate orbitals. This is our next objective.

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Note added in proof. We note that a similar use of the generating function form $\exp(\lambda S^z)$ was made by Callen in a Green-function decoupling scheme for the Heisenberg model [21].

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