I. INTRODUCTION

Pseudocubic manganites such as $La_{1-x}Sr_xMnO_3$ are of continuing interest to physicists and materials scientists for many reasons, including the possibility that the materials are 'half metals', in other words, have ferromagnetic ground states with fully spin polarized conduction bands. The possibility of 'half metallicity' was understood in the mid 1990s by Wollan, Kohler, Zener, Goode, and Anderson but remains controversial today, with some calculations and some (but not all) experiments suggesting the presence of minority carriers.

In the pseudocubic manganites such as $La_{1-x}Sr_xMnO_3$ the possible half metallicity arises from correlation and crystal-field effects involving the Mn d-levels, which in a naive formal-valence picture contain $4-x$ d-electrons (see e.g Ref for further discussion). In brief, in these materials the nearly cubic crystal field splits the $t_{2g}$-multiplet into a lower-lying $t_{2g}$ symmetry triplet and a higher lying $e_g$-symmetry doublet. Correlation effects cause the $t_{2g}$ levels to contain three electrons with parallel spin, producing an electrically inert $S = 3/2$ 'core spin', while the remaining $1-x$ electrons go into $e_g$-symmetry orbitals and can move throughout the crystal subject to a Hund's coupling $J$ which favors configurations in which an $e_g$ electron on a given site has its spin parallel to the core spin on that site. Evidently, if the Hund's coupling is strong enough, then a state in which all of the core spins are aligned will also have a fully polarized conduction band; therefore the crucial question becomes: what is the value of $J$?

From the earliest days through the revival of interest in the mid 1990s and including recent literature, many workers have assumed that $J$ is much larger than the electronic bandwidth. Such a large value of $J$ has been argued to lead to interesting observable effects including a temperature dependent optical spectral weight in the physically relevant $\omega < 4eV$ regime and to provide a direct connection between optical conductivity and the values of the magnetic transition temperatures. A temperature dependent optical spectral weight has indeed been observed. Even more importantly, the experimental phenomenology suggests that there is a strong connection between electronic energetics and magnetic order and that in particular changes in electron kinetic energy occurring as the system is driven through ferromagnetic-non-ferromagnetic transitions lead to the observed multiphase behavior, suggesting again a very large value of $J$. On the other hand, Measurements on gas-phase $Mn$ suggest a relatively small $J$ (comparable to the electronic bandwidth), and measurements on transition metal oxides suggest that gas-phase values for $J$ are only weakly renormalized by solid-state effects.

Tokura and co-workers noted that the optical conductivity may contain direct information about the value of $J$: in a paramagnetic state optical processes were possible in which an electron moves from one site, where it was locally parallel to the core spin, to an adjacent site in which (because the core spin is differently oriented) it is locally antiparallel. Such transitions should occur at an energy set by the Hund's coupling $J$, and should become visible as the temperature is raised. Ref analysed this issue in detail, presenting a comparison between theory and experiment which was argued to exclude the possibility that the 'antiparallel' transitions had been observed.

In this paper we reexamine the previous work, using a more realistic tight-binding parametrization of the
relevant portions of the band structure in combination with a dynamical mean field treatment of the interaction between electrons and core spins and employing more detailed and extensive numerics. While various aspects of this physics have been extensively studied, especially for model systems, we believe that this paper is the first to combine a realistic band parametrization with a complete treatment of dynamical and thermodynamic quantities such as the conductivity (for both low-\( T \) and \( T > T_c \)), spin-wave stiffness and transition temperature. (Of course, many important studies of static properties using realistic bands have appeared: for a recent example see\(^{20}\). We also note that in early important work Takahashi and Shiba computed \( T = 0 \) optical conductivities using a similar parametrization of band theory\(^{21}\).) We compute magnetic transition temperatures, optical conductivities and optical spectral weights and the \( T = 0 \) spin-wave stiffness for a range of model parameters and by comparison to experiment estimate the actual physical parameters for the material. We find that contrary to statements made in some previous work\(^{13}\) a moderate value of \( J \) provides a reasonable account of many aspects of the optical spectrum and a plausible account of the magnetic transition temperature and \( T = 0 \) spin wave stiffness. The differences with previous work apparently arise because the calculations of Ref\(^9\)\(^{13}\) were performed on perhaps oversimplified model systems. Also some of the results\(^{13}\) contain numerical errors.

II. MODEL

A. Hamiltonian

We consider electrons moving among sites of a simple cubic lattice according to a tight binding model with hopping matrix elements obtained from band theory and coupled to a local classical 'core spin'. Thus we write:

\[
H = H_{\text{band}} + H_{\text{int}}
\]

For the interaction we take a local coupling of core spins to conduction electrons:

\[
H_{\text{int}} = -J \sum_{a,b,\alpha,\beta,i} \tilde{S}_i \cdot C_{a,i,\alpha}^+ \sigma_{\alpha \beta} C_{a,i,\beta}.
\]

We obtain \( H_{\text{band}} \) from LSDA band calculations\(^5,22\), which are consistent with straightforward quantum chemical (Goodenough-Kanamori) considerations. Both approaches suggest that the itinerant electrons lie in two symmetry related bands which may be thought of as arising from electrons hopping among \( e_g \) symmetry local orbitals and are reasonably well described by a nearest neighbor tight binding model\(^{13,23}\). To write this hopping it is convenient to adopt a Pauli matrix notation in which the up state is the \( |x^2 - y^2 > \) orbital and the down state is the \( |3z^2 - r^2 > \) orbital. The basic hopping is orbital dependent so that

\[
H_{\text{band}} = -\frac{1}{2} \sum_{i,\alpha,\beta,\gamma} (t(\delta)^{ab}_{\alpha\beta} c_{a,i,\alpha}^+ \sigma_{\alpha\beta} c_{b,i,\beta} + H.c.)
\]

where \( \delta \) is a vector connecting site \( i \) to a nearest neighbor and the hopping is determined by a 2x2 matrix given after Fourier transformation by

\[
\varepsilon = \varepsilon_0(p) + \vec{\varepsilon}(p) \cdot \vec{\tau}
\]

with \( \vec{\varepsilon} \) the usual Pauli matrices, \( b \) the lattice constant,

\[
\varepsilon_0(p) = -t(\cos(px) + \cos(py) + \cos(pz))
\]

and \( \vec{\varepsilon}(p) = (\varepsilon_x(p), 0, \varepsilon_z(p)) \) with

\[
\varepsilon_x(p) = -\sqrt{3}t \frac{1}{2}(\cos(px) - \cos(py))
\]

\[
\varepsilon_z(p) = -t(\cos(px) - \frac{1}{2}(\cos(px) + \cos(pz)))
\]

The energy eigenvalues are

\[
E_{\pm} = \varepsilon_0 \pm \sqrt{\varepsilon_x^2 + \varepsilon_z^2}
\]

Note that along the zone diagonals ((1,1,1) etc) \( \varepsilon_{x,z} \) is 0 so the two bands are degenerate and along the line to any cube face ((1,0,0) and equivalent) one of the two bands is dispersionless. These two features occur, to a high degree of accuracy, in the calculated band structure, lending support to the nearest neighbor tight binding modelling.

The best fit to the calculated band theory leads to \( t = 0.67eV \). This, for example, implies that at band filling \( n = 0.7 \) the density of states per \( Mn \)-ion is 0.57/eV which reproduces almost exactly the local spin density approximation value 0.58/eV per \( Mn \)-ion quoted on p. 1154 of Ref\(^5\). (Note also that \(^5\) contains a misprint\(^{24}\) in the value of the Drude plasma frequency \( \Omega_D = 1.9eV \). The correct band theory value is very close to the \( \Omega_D = \sqrt{4\pi e^2 D/b} = 3.1eV \) which follows using \( t = 0.67eV \) in Eq. 12 below).

The energy bands extend from \(-3t\) to \(+3t\). Useful quantities to characterize the state of the system include the particle density

\[
n = \sum_{a,\sigma} < c_{iaa}^+ c_{iaa} >
\]

the density of states

\[
D(\varepsilon) = \frac{\partial n}{\partial \mu}
\]
FIG. 1: Upper panel: $T = 0$ density of states for a ferromagnetic ground state, plotted against chemical potential for several different Hund’s couplings $J$. Lower panel: Density of states in paramagnetic phase plotted against energy for several $J$ (whose $T = 0$ value is shown for different $J$ values in the upper panel of Fig. 1), the ’kinetic energy’ per site $K = -\frac{<H_{\text{band}}>}{N}$ (N is the number of sites) given by

$$K = \frac{1}{2N} \sum_{\delta a \sigma i} t_{ab}^{\delta}(\delta) c_{i+\delta a \sigma}^{+} c_{i \sigma} + H.c.$$  (11)

and the ’Drude weight’ $D$ which for a spin polarized system and current flowing in the $z$ direction is

$$D_{z} = b^{3} \int \frac{d^{3} p}{(2\pi)^{3}} \sum_{\lambda=\pm} \left(\frac{\partial E_{\lambda}(p)}{\partial p_{z}}\right)^{2} \delta[(E_{\lambda}(p) - \mu)]$$  (12)

Figure 2 shows the kinetic energy (divided by 3 to facilitate comparison to optics) and Drude weight, plotted against particle density for a fully spin polarized ground state.

B. Conductivity

To obtain the conductivity we require a coupling between the electric field and the electronic states. We represent the electric field by a vector potential $A$ and adopt the Peierls phase approximation, $t_{i-j} \rightarrow t_{i-j} e^{i A R_{i-j}}$. This approximation has been argued to be accurate in other transition metal oxide contexts $^{23,25}$ and was also used by Takahashi and Shiba $^{21}$. The current density operator in the $z$ direction is

$$\hat{J}_{z} = b^{3} \int \frac{d^{3} p}{(2\pi)^{3}} \sum_{\lambda=\pm} \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} (E_{\lambda}(p) - \mu)$$  (13)

The expectation value of the term in $J$ proportional to $A$ gives the total oscillator strength, $S(\infty)$ in the conduction band contribution to the optical conductivity (see $^{14,25,26}$ for details). We have

$$S(\infty) = \frac{\epsilon^{2}}{3b} K$$  (14)

The conductivity is

$$\sigma(\Omega) = \frac{S(\infty) - \chi_{jj}(\Omega)}{i\Omega}$$  (15)

with $\chi_{jj}$ the usual Kubo formula current-current correlation function evaluated using $J$ evaluated at $A = 0$.

C. Spin Wave stiffness

To compute the $T = 0$ spin wave stiffness we follow the standard procedure outlined in $^{27,28}$ We compute the energy cost of a small amplitude, long wavelength rotation of the order parameter away from the fully polarized ferromagnetic state, which we take to be aligned with the $z$ axis. The calculation is most easily carried out by locally rotating the spin quantization axis to align with the local spin direction, so that the $T = 0$ Hamiltonian becomes (note repeated orbital and spin indices $a, b, \alpha, \sigma, \sigma'$ are summed over)

$$H = -\frac{1}{2} \sum_{i,\delta} t_{ab}^{\delta}(\delta)c_{i+\delta a \sigma}^{+} c_{i \sigma} + \sum_{i,\alpha,\sigma} R_{i+\delta \alpha \sigma}^{+} R_{i \alpha \sigma'} + H.c.$$  (16)

where $R$ are the usual $S = 1/2$ rotation matrices. We find two terms. The first one arises from the term in

FIG. 2: Solid line: Kinetic energy (from Eq 11 but divided by 3) in units of hopping parameter $t$ plotted vs. particle density $n$. Dashed line: Drude weight $D_{z}/t$ (from Eq 12)
The spin wave stiffness $D_{SW}$ as conventionally defined is given by twice the total coefficient of $(q)^2 \vec{m}_q \cdot \vec{m}_{-q}$ divided by the ordered moment, i.e.

$$D_{SW} = \left[ \frac{K}{12} - 2I_{SW}(J) \right] b^2$$

This expression applies to the tight binding band structure and to all $J$ such that the ground state is fully polarized. It is consistent with those obtained by Kubo and Ohata and Furukawa who considered simpler models in the infinite $J$ and (Furukawa) first $1/J$ correction. We have evaluated $D$ (Eq 22) for the chemical potential $\mu = -1$ corresponding to the widely studied 0.7 doping; results, made dimensionless by dividing by $tb^2$ are shown in Fig. 3.

### III. Method of Evaluation

#### A. Overview

To evaluate the non-ground-state properties of $H$ we use the dynamical mean field method\textsuperscript{13,31–33} This method is extensively described and justified elsewhere, and is relevant here because the principal interactions are local. In brief the central approximation concerns the electron Green function. For the band structure of present relevance the electron Green function is written in general as

$$G(z,p) = (z - \Sigma(p,z) - \varepsilon(p) + \mu)^{-1}$$

with $\Sigma(p,z)$ the self energy. In the dynamical mean field method one takes $\Sigma$ to be $p$–independent, i.e. to depend only on $z$. The important quantity is then the momentum integrated Green function

$$G_{mom-int}(z) = b^3 \int \frac{d^3p}{(2\pi)^3} G(z,p)$$

The local Green function, being a function of frequency only, may be derived from a local quantum field theory, which is specified by a partition function $Z_{loc}$ given in terms of a mean field function $a(\tau)$. This is in general a matrix in orbital and spin indices (which we do not explicitly write here)

$$Z_{loc} = \int Dc^+cExp[\int d\tau d\tau' c^+(\tau)a(\tau - \tau')c(\tau') + \int d\tau H_{int}]$$

From this action one may extract a local Green function $G_{loc}$ and self energy $\Sigma$ via

$$G_{loc}(\tau) = \frac{\delta \ln Z_{loc}}{\delta a(\tau)} = (a - \Sigma)^{-1}$$

The mean field function $a$ is fixed via the constraint that the local Green function calculated from $Z_{loc}$ is
identical to the momentum integrated Green function obtained from Eq. 24, using the local self energy defined in the second equality in Eq. 26, i.e.

\[ G_{\text{loc}}(z) = G_{\text{mom-int}}(z) \]  

(27)

In the present problem the two orbitals are degenerate so it is not necessary to consider orbital indices in \( G_{\text{loc}} \). For \( T=0 \text{K} \) a ferromagnetic core spin configuration yields a spin dependent but frequency independent \( \Sigma \). For \( T > T_c \) Eq. 27 is an integral equation for \( \Sigma \), which is solved by numerical iteration.

B. Conductivity

In the dynamical mean field approximation there are no vertex corrections for the current current correlation function\(^{33,34} \), so it is given by

\[ \chi_{\text{J0}}(i\Omega) = T \sum_{n} \int \frac{d^3p}{(2\pi)^3} \right. \]

\[ \left. Tr[J_zG(p,i\Omega + i\omega_n)J_zG(p,i\omega_n)] \right] \] (28)

with \( G \) given by Eq. 23 using the momentum independent self energy from the final solution of Eq. 26.

The Matsubara sum and analytical continuation may easily be performed, and the dissipative part of the conductivity is given by \( \sigma(\Omega) = Im[\chi_{\text{J0}}(\Omega-i\delta)]/\Omega \). It is convenient to transform within the trace to a orbital basis which diagonalizes the Green functions. In the model considered here the self energy is the same for both orbitals, and so we may split the result into an intra- and interband part, writing

\[ \sigma_{\text{inter/intra}}(\Omega) = \int \frac{d\omega}{\pi} \theta(\omega, \Omega) \int \frac{d^3p}{(2\pi)^3} \]

\[ \frac{i^2 \sin^2(p_z) e^2}{\epsilon^2_z + \epsilon^2} \Phi_{\text{inter/intra}} \] (29)

with

\[ \theta(\omega, \Omega) = \frac{f(\omega) - f(\omega + \Omega)}{\Omega} \] (30)

\[ \Phi_{\text{intra}} = \frac{\epsilon^2_z A^{(1)}(\omega) A^{(1)}(\omega + \Omega)}{\epsilon^2_z + \epsilon^2 - \epsilon^2_z} \]

\[ + \frac{\epsilon^2_z A^{(2)}(\omega) A^{(2)}(\omega + \Omega)}{\epsilon^2_z - \epsilon^2_z} \] (31)

\[ \Phi_{\text{inter}} = A^{(1)}(\omega) A^{(2)}(\omega + \Omega) \]

\[ + A^{(2)}(\omega) A^{(1)}(\omega + \Omega) \] (32)

The quantities \( A, \epsilon \) and \( \Phi \) depend on momentum \( p \) which we have not written explicitly to avoid confusing clutter in the equations: for example, the band-n \( n = 1, 2 \) spectral function \( A^{(n)}(p, \omega) = Im[\Sigma(\omega)]((\omega - E_n(p) - Re[\Sigma(\omega)])^2 + Im[\Sigma(\omega)]^2)^{-1} \).

At \( T > T_c \) the conductivity is calculated using the self energy obtained from the solution of the mean field equations. For \( T \to 0 \) the spin-induced self energy vanishes (except for the spin-dependent energy shift) and we have added a modest impurity broadening (scattering rate \( \Sigma''(\omega) = \tau^{-1} = 0.1t \)) so the Drude peak is visible on the same scale as the interband term. The calculated \( T = 0 \) conductivity is shown in Fig. 4.

C. Transition Temperature

To determine the transition temperature it is convenient to write the mean field function in the presence of a uniform magnetization \( \mathbf{m} \), which is in principle a matrix in spin space, as

\[ \mathbf{a} = a_0 \mathbf{1} + a_1 \sigma \cdot \mathbf{m} \] (33)

and to take the magnetization direction to be parallel to \( z \) so that after integrating out the fermions we obtain for the impurity model action (the two is for orbital degeneracy)
\[ S_{\text{imp}} = 2 \sum_n T_r \ln[a_0 + a_1 \sigma_z + J (\vec{S} \cdot \vec{\sigma})] \]  

(34)

We can evaluate the trace over spins, getting (\(\theta\) is the angle between the core spin direction and the magnetization direction, which is taken to be the z direction)

\[ S_{\text{imp}}(\theta) = 2 \sum_n \ln[a_0^2 - a_1^2 - J^2 S^2 - 2a_1 JS \cos(\theta)] \]  

(35)

Assuming (as detailed studies of simpler model systems have confirmed) that the transition is second order we may study it by linearizing in \(a_1\), so that the magnetization \(m\) is given by

\[ m = \frac{\langle d \cos \theta (\cos(\theta) \exp[S(\theta)]) \rangle}{\langle d \cos \theta \exp[S(\theta)] \rangle} = -\frac{4JS}{3} \sum_{n} a_1 a_0^2 - J^2 S^2 + O(a_1^3) \]  

(36)

The impurity (local) Green function corresponding to Eq 35 is

\[ G_{\text{loc}} = [(a_0 + a_1 \sigma_z + J (\vec{S} \cdot \vec{\sigma}))^{-1}] \]

\[ = \frac{(a_0 - \sigma_z[a_1(1 + \frac{4JS^2}{3a_0^2 - J^2 S^2}) + Jm])}{(a_0^2 - J^2 S^2)} + O(a_1^3) \]  

(37)

where the average has been taken over the action defined by Eq 35. From Eq 37 we may compute the self energy \(\Sigma = -G_{\text{loc}} + a_0 + a_1 \sigma_z\) finding \(\Sigma = \Sigma_0 + \Sigma_1 \sigma_z\):

\[ \Sigma_0 = \frac{J^2 S^2}{a_0} \]  

(38)

\[ \Sigma_1 = \frac{1}{3} \frac{J^2 S^2}{a_0^2} a_1 - \frac{a_0^2 - J^2 S^2}{a_0} JS m \]  

(39)

Finally, \(a_1\) is fixed from

\[ G_{\text{loc}} = \frac{1}{2} Tr \int \frac{d^dp}{(2\pi)^d} [\omega - \Sigma_0 - \Sigma_1 \sigma_z - \varepsilon_p]^{-1} \]

\[ = \frac{1}{2} Tr \int \frac{d^dp}{(2\pi)^d} [\omega - \Sigma_0 - \varepsilon_p]^{-1} + [\omega - \Sigma_0 - \varepsilon_p]^{-1} \Sigma_1 \sigma_z [\omega - \Sigma_0 - \varepsilon_p]^{-1} \]

\[ = I_1(\omega - \Sigma_0) + \Sigma_1 \sigma_z I_2(\omega - \Sigma_0) \]  

(40)

with \(I_n(z) = \int \frac{d^dp}{(2\pi)^d} (\frac{1}{z - \varepsilon_p})^n \).

Eqs. 36-40 give an equation for \(T_c\), which contains only \(a_0\) and the paramagnetic solution of Eq. 27 which we denote here as \(G_0\).

![Diagram](image_url)  

FIG. 5: Calculated magnetic transition temperature plotted against band filling for several different \(J\) values, along with \(J = \infty\) Bethe-lattice result (for comparison to previous work).

\[ T_c = -\frac{4J^2 T_c}{3} \sum_{n} \frac{G_0^2 - I_2}{(3a_0^2 - J^2)G_0^2 + J^2 I_2} \]  

(41)

This formula shows explicitly that in weak coupling \(T_c \propto J^2\).

IV. RESULTS

As noted above, the band calculation is well fit by a \(t\) of about 0.67eV. The physically relevant densities correspond to \(n < 1\) and the most experimentally relevant density range is \(n = 0.6 - 0.8\) where the ground state is in fact a ferromagnetic metal. We have calculated the magnetic transition temperature as a function of band filling for different values of the Hund’s coupling \(J\). Representative results are shown in Fig. 5. We see that the calculated transition temperatures become noticeably higher than the experimental range (\(\sim 400\)K) once \(J\) becomes greater than a number of the order of unity. Ref\(^{13}\) argued that the calculated transition temperatures for \(J \gg t\) were consistent with experimental data. We now believe that this conclusion was based on normalizing to an incorrect value for the kinetic energy and should be disregarded. The present calculation reveals, in agreement with results obtained by previous workers, that even modest values of \(J\) suffice to push transition temperatures into a range higher than the highest observed \(T_c\). Corrections to the mean field approximation are believed\(^{13}\) in this problem to reduce \(T_c\) by of the order of 30% and do not change this conclusion.

The transition temperature data therefore suggest that a relatively modest \(J\) corresponding to a local spin flip energy \(2J \sim 2 - 3\)eV and less than the full bandwidth.
\(~ 4\text{eV}\), might be appropriate for the CMR manganite materials. Such a value of \(J\) would be consistent with the value \(J \approx 2.7\text{eV}\) appropriate for gas-phase \(Mn\) (note that spectroscopic studies such as\(^7,\!^{18}\) indicate that the Hund’s coupling is much less renormalized by solid-state effects than is the on-site Coulomb interaction). Our computed values for the \(T = 0\) spin wave stiffness also support a lower value for \(J\). Values in physical units may be obtained by multiplying the data shown in Fig 3 by \(tb^2 \approx 9.67\) (using the numerical values \(t = 0.67\text{eV}\) and \(a = 3.84\)). A \(J = 2.5t\) yields a \(D = 300meV\cdot\AA^2\), already much higher than the \(D \approx 160 - 190eV\cdot\AA^2\) observed\(^9\) in optimally metallic manganites.

Further evidence that a smaller value of \(J\) might be appropriate comes from our calculated optical conductivity, shown in Fig. 6 for \(T > T_c\) and various values of \(J\). Comparison to Fig 4 shows that if \(J\) is larger than about \(1\text{eV}\), then as \(T\) is raised a new feature becomes visible at an energy of order \(2J\). This feature arises from motion of carriers from one site to another site with an antialigned spin, and its existence was noted by Tokura and co-workers\(^9\). Such a feature is visible in experimental data at an energy of about \(3\text{eV}\)\(^1\). Comparison of Fig. 7 to the data of\(^1\) shows that a \(J = 2.5t \approx 1.5\text{eV}\) would approximately reproduce the observed peak position and magnitude. For smaller values of \(J\) the peak becomes difficult to distinguish from the interband transition; for larger values the peak moves to higher energies than is observed and the oscillator strength decreases below what is observed. The Table shows the oscillator strength in the “peak at \(2J\)”\(^7^\), obtained by integrating \(\sigma_{T_c} - \sigma_0\) from the highest zero-crossing up to \(w = \infty\) and converting to kinetic energy via Eq 14 (e.g. the integration for \(J = 3.33\text{eV}\) and \(n = 0.7\) includes the area between \(5.8\text{eV}\) and \(9.6\text{eV}\)). It is also convenient to define \(K_{T_c}\), the total spectral weight of \(\sigma\) at \(T_c\), by evaluating Eqs 11, 14 at \(T = T_c\). The ratio \(K_{anti}/K_{T_c}\) can be compared with the experimental results of Ref\(^1\).

Ref\(^1\) argued, on the basis of calculations based on an oversimplified Bethe-lattice model, that the observed oscillator strength in the putative ‘peak at \(2J\)” was too large to be consistent with the calculation. The present calculations, which are based on the more realistic tight binding model, suggest that this is not the case.

Previous work\(^9,\!^{13}\) has drawn attention to a relationship between optical spectral weight (i.e. kinetic energy)
FIG. 8: Comparison of optical spectral weight (expressed as a kinetic energy) at $T_c$ (dashed and dotted lines) and 0 K (solid line) and various J; the minimal J value, which gives a halfmetallic ground state is dependent on $n$ (e.g. for $n=0.7$ is $J_{min} = 0.64eV$)

changes and the value of the magnetic transition temperature. Our new results indicate that this result is of less generality than suspected. Fig. 8 compares, for different values of $J$, the optical spectral weight at $T = T_c$ (dashed and dotted lines) and $T = 0$ (solid line). The $J = \infty$ limit reveals the expected approximately 30% decrease in spectral weight between $T = 0$ and $T = T_c$, but the other traces reveal that the situation is more complicated in general. Indeed, at $T = 0$ and for $J$ sufficiently large, only one spin band is occupied, and the band is therefore filled to a higher level, thereby losing kinetic energy. On the other hand, at $T > T_c$ both spin directions are equally probable, so electrons can redistribute themselves to the lower parts of the band, thereby gaining kinetic energy. However, in the paramagnetic phase the non-vanishing $J$ blocks some hopping processes. At intermediate $J$ these two processes compete, and as seen in Fig. 8, which one is dominant depends on both the value of $J$ and the band-filling. The blocking process is always dominant only at very large $J$.

The non-systematic change of kinetic energy upon magnetic ordering observed for reasonable values of $J$ suggests that the relation, proposed in previous work\textsuperscript{13},

between change in kinetic energy and value of transition temperature is not as general as expected. The lower panel of Fig. 9 shows the value of the transition temperature, plotted against change in kinetic energy for several different carrier densities, with $J$ as an implicit parameter. Also shown, as the solid line, is the result obtained in Ref\textsuperscript{13} (the different slope occurs because the results of\textsuperscript{13} are for an orbitally non-degenerate model). A linear dependence seems to be reasonable and is consistent with the ideas in Ref\textsuperscript{13} but we observe, that the slope is doping dependent. The upper panel in Fig. 9 shows the same plot for several different $J$, with carrier density as an implicit parameter. A much more complicated appearance is obvious. We see that the relation between $T_c$ and kinetic energy is not at all universal, but depends strongly on electron density, essentially for the reasons given in the paragraph above.

| $J$[eV] | $-K_{anti}[meV]$ | $K_{anti}/K_{Tc}$ |
|---------|------------------|------------------|
| 0.67    | 13.3             | 0.038            |
| 1.33    | 55.6             | 0.17             |
| 2.00    | 56.7             | 0.19             |
| 2.67    | 48.9             | 0.17             |
| 3.33    | 42.2             | 0.15             |

FIG. 9: Upper panel: Open dots: calculated transition temperature plotted against change in kinetic energy between $T = T_c$ and $T = 0$ for different $J$ as shown with density $n$ as implicit parameter. Dashed lines are guides to the eye. Along each line the doping increases by 0.1 steps along the lines from 0.1 up to 1 or 0.9. Lower panel: Filled dots: calculated transition temperature plotted against change in kinetic energy between $T = T_c$ and $T = 0$ for different fixed densities, as shown, with Hund’s coupling $J$ as implicit parameter. Dashed lines are results of a linear regression for the $T_c$ vs $\Delta K(J)$ curves. Solid line: results published in\textsuperscript{13} for a Bethe lattice; the difference seems to arise because of an omitted factor of two in\textsuperscript{13}.
V. CONCLUSIONS

We have presented a detailed analysis of the optical conductivity and magnetic transition temperature of electrons using a quasirealistic band structure and Hund’s coupling to classical ‘core spins’. Our main finding is that, in contrast to a widely-made assumption in the manganite literature but in agreement with quantum chemical estimates and gas-phase measurements, a moderate value of the Hund’s coupling \( J \approx 1.3 - 1.5eV = 2.5t \) (large enough to fully polarize the conduction band at \( T = 0 \) but less than the LSDA band width) suffices to account both for the order of magnitude of the magnetic transition temperature and for a feature observed in the optical conductivity in the paramagnetic phase. We have also presented predictions for the evolution of this feature as the carrier density is varied. Errors in previous work\(^{13}\) (arising from use of an oversimplified band structure and from numerical errors) have been corrected. We have also presented results for the spin-wave stiffness expected for a fully polarized ground state and a tight-binding band structure at arbitrary \( J \).

We observe that even with a modest \( J \) the spin wave stiffness and transition temperature are rather overestimated. It is possible to fix up these discrepancies by adding a phenomenological antiferromagnetic core-spin-core-spin coupling, as has been proposed by many authors. However, in our view a more serious problem remains: if these estimates for \( J \) are accepted, then our basic understanding of the physics of the manganites must be reconsidered, because for these value of \( J \) the change in kinetic energy between ferromagnetic and paramagnetic state is very small. A series of experiments has made it clear that the ‘colossal’ magnetoresistance is associated with a change in electronic state from a more or less fermi-liquid like state to a state with strong local lattice distortions, leading to either short ranged or long ranged charge and orbital ordering. Previous work\(^7\), based on a large-\( J \) limit, had predicted this behavior as a consequence of the decrease in kinetic energy as spins were disordered, allowing the localizing tendency of the electron-phonon interaction to overcome the banding tendency of the electrons and produce a new phase. An approximately 40 per cent decrease in conduction band oscillator strength, which is naturally associated to a decrease in electronic kinetic energy, is indeed observed, and was argued to be consistent with this physics. However, if \( J \) is in the range proposed here, then the disordering of the spins does not significantly change the electronic kinetic energy and another explanation for the ‘colossal’ magnetoresistance must be sought.

One possibility is that a ‘Hubbard U’ interaction is important, and reduces the electronic kinetic energy and band width to the point where even the modest \( J \) we find here can have a strong effect. However, the total conduction band spectral weight observed at low \( T \) is close to the full band theory value, and indeed the while our simple calculation does not reproduce the detailed line-shape, the calculated magnitude of the conductivity in the 1-3eV is in reasonable agreement with data, suggesting that Hubbard-U effects are perhaps not so important. It is possible, however, that the good agreement is accidental, and that contributions from other orbitals (for example, ‘wrong spin’ \( t_{2g} \) electrons) are important in the real materials, although they are absent in our calculations. We also note that the susceptibility of the finite \( J \) paramagnetic state to local (or long ranged) lattice distortions has not been computed. It might be that even though the kinetic energies of the \( T = 0 \) and \( T > T_c \) phases are similar (at moderate \( J \)), the \( T > T_c \) phase is more susceptible to local lattice distortions. This possibility urgently deserves further investigation.

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