Optical spectral weights and the ferromagnetic transition temperature of CMR manganites: relevance of double-exchange to real materials

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(February 26, 2018)

We present a thorough and quantitative comparison of double-exchange models to experimental data on the colossal magnetoresistance manganese perovskites. Our results settle a controversy by showing that physics beyond double-exchange is important even in La\textsubscript{0.7}Sr\textsubscript{0.3}MnO\textsubscript{3}, which has been regarded as a conventional double-exchange system. We show that the crucial quantity for comparisons of different calculations to each other and to data is the conduction band kinetic energy $K$, which is insensitive to the details of the band structure and can be experimentally determined from optical conductivity measurements. The seemingly complicated dependence of $T_c$ on the Hund’s coupling $J$ and carrier concentration $n$ is shown to reflect the variation of $K$ with $J$, $n$ and temperature. We present results for the optical conductivity which allow interpretation of experiments and show that a feature previously interpreted in terms of the Hund’s coupling was misidentified. We also correct minor errors in the phase diagram presented in previous work.

I. INTRODUCTION

The colossal magnetoresistance (CMR) rare earth manganese perovskites first attracted attention in the 1950s because of the range of magnetic and structural transitions they display. Recent interest has been revived by the extremely large ("colossal") magnetoresistance displayed by some CMR materials, coupled with their rich phase diagram. Despite this long and intense study, much of the physics of CMR remains controversial, with basic issues still subject to debate. In this paper we address two such issues: the first is in what sense the standard ‘double-exchange only’ model (defined below) describes the physics in the regions of the phase diagram where the ground state is a ferromagnetic metal. The second is the interpretation of the optical conductivity spectrum, and in particular which portions of the observed spectrum pertain to the conduction band electrons responsible for the interesting physics of the CMR materials. In addition to its direct relevance to CMR, we believe our work is of broader significance for the theory of interacting electrons in solids, as a contribution to the fundamental issues of the quantitative comparison of many-body calculations to the properties of materials, and to the interpretation of the optical spectra of correlated electron materials.

It is generally agreed that a crucial aspect of CMR physics is ‘double-exchange’: the mobile carriers (which, for CMR materials, are $\text{Mn} e_g$ symmetry $d$-electrons) are strongly coupled ferromagnetically to localized core spins ($\text{Mn} t_{2g}$ symmetry electrons). This means that core spin alignment dictates carrier motion, which in general leads to ferromagnetism. What, if any, additional physics beyond double-exchange is important for the manganites is still debated. For example, some authors have argued that double-exchange only models correctly predict the magnetic transition temperatures of CMR materials\textsuperscript{7}, while others have argued that they do not\textsuperscript{6,8}. Published calculations of $T_c$ have not resolved the issue because the results depend on model parameters such as bandwidth, interaction strength and carrier density, often chosen arbitrarily or in a manner inconsistent with the physics of the manganites. We attempt to settle the question by establishing the full phase diagram and by presenting precise calculations of $T_c$ within a specific model. Most importantly, by showing how the parameters on which the calculated $T_c$ depends can be related to measured properties of real materials and to parameters used in other calculations.

Another important set of issues concerns the optical conductivity, which in the CMR materials has a strong dependence on chemical composition, frequency, temperature and magnetic field.\textsuperscript{9–11} It is widely believed that information extracted from these data will be helpful in elucidating the physics of CMR, but this goal has not yet been fully achieved. Even the basic question of which parts of the observed spectrum are due to the conduction band degrees of freedom is still not settled.\textsuperscript{9–15} We show here that a comparison of the data to the theoretically calculated magnitude and temperature dependence of optical spectral weight, and of its relationship to $T_c$, can resolve this issue. Our results indicate that several previous analyses\textsuperscript{9} underestimate the actual conduction band spectral weight, and suggest that a previous paper by several authors including one of us\textsuperscript{8} misidentified a key feature in the data. This misidentification arose pri-
mainly from reliance on a theoretical calculation based on inappropriate system parameters.

In this paper we perform a systematic and quantitative comparison of the predictions of double-exchange-only models to data. We incorporate “real materials” aspects via a tight-binding fit to the band structure; in these materials a simple nearest neighbour hopping model represents the real material parameters adequately. In particular, we show that the crucial quantity for comparison of calculations to data or to other calculations is the conduction-band kinetic energy, a local expectation value related to the electron hopping amplitude and defined more precisely in Eq. (2). Because this kinetic energy is a local quantity, it can be easily and reliably calculated and is insensitive to the fine details of the band structure. When expressed in terms of the kinetic energy, the apparently disparate results of a variety of computations are seen to be in quantitative agreement. More importantly, because the kinetic energy may be determined from optical conductivity experiments, our results enable a detailed comparison of theory to experiment.

To treat the many-body physics of double-exchange, we use the highly successful dynamical mean-field method\cite{1}. This provides a detailed and apparently reliable treatment of the local correlations which are crucial to the conclusions we wish to draw. We will argue that the effect of the omitted physics, which has to do with intersite fluctuations, may be reliably estimated for the issues of interest to us and is not too large.

The application of the dynamical mean-field method to double-exchange systems was pioneered by Furukawa\cite{2}, whose important work established the basic usefulness of this method and presented many valuable results concerning the phase diagram, conductivity and other properties. However, his work was incomplete in some respects, incorrect in some details, and in many cases employed physically irrelevant parameters. The full treatment we present here is therefore needed.

We believe that the analysis presented in this paper may be useful in the general context of the development of a predictive theory of dynamical and ordering phenomena in systems, such as transition metal oxides, with strong interactions. This is an important goal of electronic condensed matter physics; and recent advances in computational power and in the techniques of many-body physics suggest that it may be obtainable. The dynamical mean-field method\cite{1} seems particularly promising in this respect. It is computationally tractable, includes incoherent and inelastic effects, and can be combined with conventional band theory. Investigations of the extent to which this method is useful in the computation of real materials properties are urgently needed. The work described below is such an investigation. In this context we point out that one of the great advances in the theory of equilibrium critical phenomena arose from a careful and detailed application of mean-field theories. We believe that a similar opportunity may be present in the application of the dynamical mean field theory to strong correlation physics.

The rest of the paper is organized as follows. Section II defines the double-exchange-only model, and explains how its parameters should be defined and related to experiments. Section III presents the dynamical mean-field formalism. Section IV presents the numerical and analytical results for the phase diagram, and corrects what seems to be a minor error in the phase diagram proposed in \cite{1}. Section V presents results for the spectral weight (integrated area) in different regions of the optical conductivity spectrum. Section VI provides a summary of our theoretical results and a detailed interpretation of experimental data, particularly of optical conductivity and its relation to $T_c$. Readers interested only in this aspect may proceed directly to this section. Section VII is a conclusion and presents some possible directions for future research.

II. MODEL

The double-exchange-only (DE) model involves electrons (with orbital indices $a,b$) moving in a band structure defined by a hopping matrix $t_{ij}^{ab}$ and a chemical potential $\mu$, and connected by a Hund’s coupling ($J > 0$) to core spins $S$, which we take to be classical. We denote the operator creating an electron of spin $\alpha$ on orbital $a$ of site $i$ by $d_{ia\alpha}^\dagger$ and define the double-exchange-only Hamiltonian $H_{DE}$ by:

$$H_{DE} = -\sum_{<ij>,ab} t_{ij}^{ab} d_{ia\alpha}^\dagger d_{jb\alpha} - \mu \sum_{iaa} d_{iaa}^\dagger d_{iaa} - J \sum_{iaa,\beta} \vec{S}_i \cdot \vec{d}_{ia\alpha}^\dagger \vec{d}_{ia\beta}.$$  \hspace{1cm} (1)

Here $t_{ij}^{ab}$ is the amplitude to hop from site $i$, orbital $a$ to site $j$, orbital $b$. The calculated band structure \cite{1} is well fit by a $t_{ij}^{0b}$ which involves only nearest-neighbour hopping.

A crucial quantity is the electron kinetic energy $K$, defined by:

$$K = \frac{-2}{ZN_{\text{sites}}} \sum_{<ij>} \left< t_{ij}^{ab} d_{ia\alpha}^\dagger d_{jb\alpha} \right>$$

$$= \frac{2}{Z} \int \frac{dp}{2\pi d} e^{ip} \left< \vec{d}_{ia\alpha}^\dagger \cdot \vec{d}_{ia\beta} \right>, \hspace{1cm} (2)$$

where $e^{ip}$ is the dispersion obtained by Fourier transforming $t_{ij}^{ab}$, $Z$ is the number of nearest neighbours and $d$ is the spatial dimension. In the limit $J \to \infty$, $K$ is the only relevant energy scale in the model and in particular the magnetic transition temperature $T_c$ has been found to depend only on $K$ \cite{1}. $K$ is also a suitable quantity for comparison to experiment because, in the physically realistic limit of nearest-neighbour hopping, $K$ may be determined from an analysis of the optical
conductivity. We would like to mention that this $K$ includes a sum over all bond directions and is 3 times larger than the $K$ defined in Quijada et al.

### III. FORMALISM

In this paper we study the magnetic transition temperature and kinetic energy of $H_{DF}$ as functions of carrier density $n$ and Hund’s coupling using the dynamical mean field approximation, supplemented by an analytical treatment of the $J_d \to 0$ limit and by comparison to results of Monte Carlo and series expansions.

The dynamical mean field method has been discussed extensively elsewhere and therefore we omit formal details. This scheme was first obtained in the limit $d \to \infty$ with $t_{ij} \sim 1/\sqrt{d}$, and is often referred to as the infinite dimensional limit, but the central approximation is actually the neglect of the momentum dependence of the electron self-energy, $\Sigma$. While this becomes exact in the limit $d \to \infty$, it is quite a good approximation in $d = 3$, where the momentum dependence of $\Sigma$ is quite weak.

To make the importance of the neglect of momentum dependence clear and to establish notation we sketch a derivation of the dynamical mean field equations. A momentum independent $\Sigma$ implies that all of the many-body physics may be derived from the local (momentum-independent) Green’s function $G_{loc}$, defined by

$$G_{loc}(\omega) = Tr \int \frac{d^4 p}{(2\pi)^3} G(p, \omega)$$

where the Green’s function, a matrix in orbital ($ab$) and spin ($\alpha\beta$) space, is

$$G(p, \omega) = [\omega + \mu - \epsilon^a_b - \Sigma^{ab}_{\alpha\beta}(\omega)]^{-1}.$$  \hspace{1cm} (4)

It is convenient to define the density of states summed over bands $N(\epsilon) = \sum_\lambda \int \frac{d^2 p}{(2\pi)^2} \delta(\epsilon - \epsilon^\lambda_p)$, where $\epsilon^\lambda_p$ are the eigenvalues of $\epsilon^a_b$. We shall be interested in the pseudocubic manganese perovskites in the doping regimes where orbital order is not important. For these materials $N(\epsilon) = \int \frac{d^4 p}{(2\pi)^4} \delta(\epsilon - \epsilon^\lambda_p)$ is independent of $\lambda$; thus we use $N(\epsilon) = n_{orb} N(\epsilon)$, where we have introduced the number of orbitals $n_{orb}$. For the physically relevant case, $n_{orb} = 2$, but the case $n_{orb} = 1$ has been considered by other workers, so we retain $n_{orb}$ as a variable to facilitate comparisons. The local Green’s function is then proportional to the unit matrix in $ab$ space, $G = G_{loc,\alpha\beta} \delta_{\alpha\beta}$ and the coefficient is

$$G_{loc,\alpha\beta}(\omega) = \int d\epsilon N(\epsilon) \frac{1}{\omega + \mu - \epsilon - \Sigma_{\alpha\beta}(\omega)}.$$  \hspace{1cm} (5)

$G_{loc}$ depends only on frequency and is therefore the solution of a single-site problem. This problem is specified by a mean-field function $b_{\alpha\beta}(\omega)$ which in the model of present interest, is related to $G_{loc}$ via

$$G_{loc,\alpha\beta}(\omega) = \int d^2 q \frac{P(\tilde{q})}{b_{\alpha\beta}(\omega) - J \tilde{S} \cdot \tilde{\sigma}_{\alpha\beta}}.$$  \hspace{1cm} (6)

with

$$P(\tilde{q}) = \frac{1}{Z_{loc}} \exp \left[ \sum_n Tr \ln \left[ b_{\alpha\beta}(i\omega_n) - J \tilde{S} \cdot \tilde{\sigma}_{\alpha\beta} \right] \right],$$

and $Z_{loc}$ ensures that $\int d^2 q P(\tilde{q}) = 1$. The self-energy is defined by

$$\Sigma_{\alpha\beta}(\omega) = b_{\alpha\beta}(\omega) - G^{-1}_{loc,\alpha\beta}(\omega)$$

and the mean-field function $b_{\alpha\beta}(\omega)$ is fixed by the requirement that Eq. (3) holds with $G_{\alpha\beta}(p, \omega)$ defined by Eq. (4), $\Sigma$ defined by Eq. (7), and $G_{loc}$ by Eq. (5).

Using the momentum independence of the self-energies within this mean-field model, the electron kinetic energy (Eq. (3)) can be written as

$$K = \frac{2}{Z} \sum_n \int \frac{d^4 p}{(2\pi)^3} \epsilon_p Tr[G(p, i\omega_n)]$$

$$= \frac{2}{Z} \sum_n Tr [G_{loc}(i\omega_n)]^2.$$  \hspace{1cm} (9)

In a ferromagnetic state with magnetization direction $\hat{m}$ we have

$$b_{\alpha\beta}(\omega) = b_0(\omega) + b_1(\omega) \hat{m} \cdot \tilde{\sigma}_{\alpha\beta}$$

$(b_1(\omega) = 0$ in the paramagnetic state). If the spin axis is chosen parallel to $\hat{m}$ then $b_{\alpha\beta}$ becomes a diagonal matrix with components parallel ($b_{\uparrow} = b_0 + b_1$) and antiparallel ($b_{\downarrow} = b_0 - b_1$) to $\hat{m}$.

The precise form of the equation for $b_\sigma$ depends on the form of the density of states $N(\epsilon)$, but the important behaviour of the physical observables do not, as long as $N(\epsilon)$ has a finite second moment ($\int d\epsilon N(\epsilon)^2 < \infty$). We perform our calculations on the Bethe lattice with a semi-circular density of states per orbital

$$N(\epsilon) = \frac{\sqrt{4t^2 - \epsilon^2}}{2\pi t^2}.$$  \hspace{1cm} (11)

Here $t$ is a bandwidth parameter; the full bandwidth of the non-interacting ($J = 0$) problem is $4t$. This corresponds to number of nearest neighbours $Z = 2d$, as $Z \to \infty$ with $tZ$ fixed. For this $N(\epsilon)$, the self-consistent equations for the $b_\sigma$ are

$$b_{\uparrow} = i\omega_n + \mu + \int_{-1}^{1} d(\cos\theta) P(\theta) \times \frac{-b_1 + J\cos\theta}{b_{\uparrow} b_{\downarrow} - (b_{\uparrow} - b_{\downarrow}) J\cos\theta - J^2}$$

$$b_{\downarrow} = i\omega_n + \mu - \int_{-1}^{1} d(\cos\theta) P(\theta) \times \frac{(b_1 + J\cos\theta)}{b_{\uparrow} b_{\downarrow} - (b_{\uparrow} - b_{\downarrow}) J\cos\theta - J^2}.$$  \hspace{1cm} (12)
and the local Green’s function (Eq. 8) can be written in terms of the mean-fields as
\[ G_{loc}^{\sigma}(i\omega_n) = i\omega_n + \mu - b_\sigma(i\omega_n). \] (13)

The self-energies are evaluated using Eq. (8) and the full Green’s function from Eq. (8).

We will also be interested in the optical conductivity, \( \sigma \). The contributions to \( \sigma \) from the states described by \( H_{DE} \) are obtained by coupling an electromagnetic field to \( H_{DE} \) (Eq. 8) via the Peierls coupling \( t_{ij} \rightarrow t_{ij}e^{iA(r_i - r_j)} \) and performing linear response. If the self-energy is momentum independent then there are no vertex corrections in \( \sigma \) and
\[ \sigma_{ij}(i\Omega_n) = e^2 \frac{1}{i\Omega_n} \left[ S(\infty) - \sum_\sigma \int d^dp \left( \frac{1}{2\pi^d} \right)^d \right] \]
\[ T \sum_m Tr \left[ \gamma_\sigma^i G_\sigma(p, i\omega_n) \gamma_\sigma^j G_\sigma(p, i\Omega_n + i\omega_m) \right] \] (14)

where \( \gamma_\sigma \) is the current operator determined by the Peierls substitution; it includes both intra and interband transitions within the manifold of conduction band states. Since we concentrate on materials with cubic symmetry, \( \sigma_{ij} \sim \delta_{ij} \) and hereafter we suppress the spatial indices on \( \sigma \).

This conductivity obeys the sum rule
\[ S(\infty) = \frac{a_{d-2}}{e^2} \int_0^\infty \frac{1}{\pi} d\Omega \sigma(\Omega) = \sum_{i\delta a \alpha} t_{i\delta}^\beta \delta^2 \left( d_{i\delta a} d_{i+\delta \alpha} + H.c. \right) \] (15)

If the nearest neighbour hopping is dominant then \( \int_0^\infty \frac{1}{2} d\Omega \sigma(\Omega) = \frac{e^2}{\pi a_{d-2}^2} K \). The experimentally measured optical conductivity includes additional transitions not described by \( H_{DE} \) and obeys the familiar f-sum rule \( \int_0^\infty \frac{1}{2} d\Omega \sigma(\Omega) = \frac{n_e}{m} \), with \( n \) the total (conduction and valence) electron density and \( m \) the bare mass. Extracting the portion pertaining to the \( \epsilon_g \) electrons from the measured conductivity has been controversial. We will show below how to do this using the information on spectral weights we derive below. Here we concentrate on the optical conductivity (Eq. 14) and the generally valid expression
\[ K = \int d\epsilon_k \frac{d\omega}{\pi} f(\omega)\epsilon_k N(\epsilon_k) ImG(\epsilon_k, \omega). \] (17)

Requiring the two to be equal yields the differential equation
\[ -\frac{\partial}{\partial \epsilon_k} [N(\epsilon_k)\varphi(\epsilon_k)] = N(\epsilon_k)\epsilon_k, \] (18)
the solution of which yields \( \varphi(\epsilon_k) \).

IV. NUMERICAL METHODS

We now outline the numerical methods used to solve Eqs. (12) and compute physical quantities. For computational convenience, we rewrite the self-consistency Eqs. (12) in terms of \( b_0 \) and \( b_1 \) defined in Eq. (8). At \( T > T_c \), there is no magnetic order and \( b_1(\omega) = 0 \). At \( T = 0 \) and non-zero \( J \), all spins are ferromagnetically aligned implying \( P(\theta) \rightarrow \delta(\cos \theta - 1) \), and one can analytically solve for the \( b_{0,1}(i\omega_n) \) from Eq. (12). We initialize \( b_{0,1} \) with the \( T = 0 \) expressions, and solve Eq. (12) on the Matsubara axis by direct iteration.

![Figure 1](image-url)  
**FIG. 1.** The dependence of \( T_c \) on the Matsubara cutoff \( \omega_{max} \), for \( n = 0.75, n_{orb} = 1 \) and \( J = 10t \); \( \omega_{max} \) ranges from \( 4(4t + J) \) to \( 4(4t + J) \). The quadratic dependence on \( t/\omega_{max} \) holds for the \( n_{orb} = 2 \) as well. For our calculations we choose \( \omega_{max} = 3(4t + J) \) which produces a value of \( T_c \) that is within 0.5% of the \( \omega_{max} \rightarrow \infty \) value.

It is convenient to compute \( P(\theta) \) by solving Eq. (12) on the Matsubara points, and then use this converged \( P(\theta) \) to solve the equations on the real axis. Convergence to within an rms error of \( 10^{-4} \) was usually achieved within 12 iterations unless one is close to the magnetic \( T_c \) where there is critical slowing down. The electron density \( n \) and the normalized magnetization density \( m \) are given by...
\[ n = n_{\text{orb}} T \sum_n \left[ G_1^{\text{loc}}(i\omega_n) + G_2^{\text{loc}}(i\omega_n) \right] \]
\[ m = \int_{-1}^{1} d(\cos\theta) \cos \theta P(\theta). \]

An accurate value of the transition temperature \( T_c \) was most conveniently obtained by computing values of \( m \) (Eq. \( \text{[20]} \)) in the range \( 0.17 \leq m \leq 0.3 \) and finding \( T_c \) by fitting to the mean-field expression \( m^2(T) = \alpha(T_c - T) \), with \( \alpha \) and \( T_c \) fit parameters.

The next issue is that of the number of Matsubara points needed to compute \( S_{\text{loc}}(\theta) \), \( n \) and \( K \). At large \( \omega_n \), the asymptotics of the mean fields are

\[ b_0 = i\omega_n + \mu - \frac{1}{i\omega_n + \mu} - \frac{1 + J^2}{(i\omega_n + \mu)^3} \]
\[ b_1 = \frac{Jm}{(i\omega_n + \mu)^2}. \]

In our computations we choose a frequency cutoff \( \omega_{\text{max}} \) and evaluate higher frequency contributions analytically using the asymptotic form given in Eq. \( \text{[21]} \). The errors in physical quantities are of order \( \omega_{\text{max}}^{-2} \). This is illustrated for \( T_c \) in Fig. \( \text{[1]} \). To achieve an accuracy of \( 5 \times 10^{-4} \) in \( n \), it was required to choose \( \omega_{\text{max}} \geq 3(4t + J) \). Choice of this value of \( \omega_{\text{max}} \) also ensures that the value of \( T_c \) is within 0.5% of the \( \omega_{\text{max}} \to \infty \) value.

\section*{V. Ferromagnetic Transition Temperature}

\subsection*{A. Analytical results}

We first present mean field results and establish the phase diagram, then show how to incorporate fluctuation corrections. We begin with analytical results for small ferromagnetic \( J \). At \( T = 0 \) the core spins are magnetically ordered with a characteristic wavevector \( \vec{q} \). The polarized core spins produce an effective magnetic field on the conduction electrons which polarizes them, leading to a magnetization \( \vec{m}_q \) and to a change in energy, which for small \( J \) is

\[ \delta E = \frac{1}{2} \left( \chi_q^0 \right)^{-1} \vec{m}_q \cdot \vec{m}_{-q} + \frac{1}{2} J \left( \vec{S}_q \cdot \vec{m}_{-q} + H.c. \right) \]

where \( \chi_q^0 \) is the magnetic susceptibility for noninteracting electrons at wavevector \( \vec{q} \). Minimization of Eq. \( \text{[22]} \) leads to \( \vec{m}_q = \chi_q^0 \vec{S}_q \) and \( \delta E = -\frac{1}{2} J^2 S_q^2 \chi_q. \) Thus at small \( J \) the system will order at the wavevector which maximizes \( \chi_q \).

The wavevector at which \( \chi \) is maximal depends on band filling and dimensionality. Here, for consistency and to facilitate comparison to other works, we restrict ourselves to infinite-\( d \). For very large \( d \), \( \chi(q) \) is independent of \( q \) except for regions of width \( O(1/\sqrt{d}) \) about \( \vec{q} = 0 \) and the commensurate antiferromagnetic vector \( \vec{q} = \vec{Q} = (\pi, \pi, \ldots) \); therefore at \( d = \infty \) we need to consider \( \chi_{Q=0}, \chi_{Q=Q} \) and the susceptibility at a typical \( q \), \( \chi_{\text{loc}} \). Fig. \( \text{[2]} \) shows the ferromagnetic (bold line), antiferromagnetic (solid line) and local (dashed line) susceptibilities for \( J = 0 \) calculated for a semicircular density of states. The phase corresponding to the maximal \( \chi \) is the small \( J \) ground state. For \( 0 \leq n/n_{\text{orb}} \leq 0.195, \chi(q = 0) \geq \chi_{\text{loc}, Q}; \) this is the range of dopings where the DE model has a low \( J \) ferromagnetic ground state. At small \( J \) and intermediate \( n \) (0.195 < \( n/n_{\text{orb}} < 0.35 \), \( \chi_{\text{loc}} \) is largest, implying order at wavevector different from both 0 and \( Q \). At \( d = \infty \) all such wavevectors are degenerate, implying some sort of highly degenerate ground state. Finite \( d \) corrections will select a particular wavevector. We therefore identify the phase as incommensurate (IC). For \( n/n_{\text{orb}} > 0.35, \chi_Q \) is largest and the small \( J \) phase is a commensurate antiferromagnet.

\[ T_c = \frac{J^2}{3} \left[ \chi_Q - \chi_{\text{loc}} \right]. \]

An explicit analytic expression for ferromagnetic \( T_c \) can be obtained by working out the susceptibilities in the mean-field Eq. \( \text{[2]} \) for \( Q = 0 \), and one finds that
for the semicircular density of states, the state energy vanishes, because no hopping is possible. The relevant band structure, and is therefore of order $t$. For the Bethe lattice with $Z$ nearest neighbours and near neighbour hopping $t$, the energy is

$$E_{AF} = -\frac{2Zt^2}{J} - \frac{2t^2}{J} - O \left( x^2 t \right) \quad (26)$$

where the arrow indicates the $d \rightarrow \infty$ limit. Further, one may consider an incommensurate state which for the purpose of $d = \infty$ energetics has a random spin arrangement, leading to

$$E_{IC} = \frac{t^2}{J} - \sqrt{2} xt + O \left( x^2 t \right) \quad (27)$$

Equating $E_{FM}$ to $E_{IC}$ implies a ferromagnet-incommensurate transition at $x_{F-I} = t / ((2 - \sqrt{2})J)$; equating $E_{AF}$ to $E_{IC}$ yields incommensurate-anitferromagnetic transition at $x_{I-A} = t / (\sqrt{2}J)$. Thus at small $1 - n$ the sequence of spatially homogeneous phases as was also found at small $J$ and small $n$. However, at small $1 - n$ and large $J$, inhomogeneous phases are favoured: the standard Maxwell construction applied to Eqs. (25,26,27) shows that in this model, which neglects long ranged Coulomb interactions, the whole small $x$ and large $J$ regime is in fact phase separated into commensurate $AF$ and $F$ with $x \sim 1/2$, as previously noted by [9]. Combining the small $J$ and large $J$ results, we obtain the phase diagram shown in Fig. (3). The qualitative behaviour is correct; we do not have precise numerical values for the IC-AF or PS phase boundaries.

We emphasize that the large-$J$ antiferromagnetism and phase separated behaviour is tied to the regime $n \approx n_{orb}$, where the conduction band of the ferromagnetic state is almost completely full. The physically relevant regime for the manganites is $n < n_{orb}/2$, where the large $J$ behaviour is simply ferromagnetic. Several published papers have asserted that behaviour associated with the regime $n \approx n_{orb}$ is relevant to the manganites. In our view, these assertions are unjustified because $n_{orb} = 2$, so the physical density corresponds to $n/n_{orb} < 0.5$.

It is possible that additional interactions, neglected here, could change the effective orbital degeneracy from 2 to 1. We do not believe this happens in the manganites. In support of our view we cite the case of LaMnO$_3$. This is an insulator which has a frozen Jahn-Teller distortion which acts to quench the orbital degrees of freedom. This material is a $(0,0,\pi)$ antiferromagnet: its magnetic structure is ferromagnetic planes antiferromagnetically coupled. The ferromagnetic in-plane coupling means that virtual hopping to the empty orbital is the dominant process. The much weaker antiferromagnetic bond perpendicular to the plane is believed to be due to an additional $t_{2g} - t_{2g}$ superexchange which becomes important because the Jahn-Teller order strongly suppresses the out of plane hopping. In other words, even in the material in which the tendency to quench the orbital degree of freedom is strongest, there is no conduction band mediated antiferromagnetism in the physically relevant regime. We believe, therefore, that the physics of $n \approx n_{orb}$ is simply not relevant to the manganites, in contrast to the assertions made in [9].

In the same way, the phase separation discussed in Refs. [15,20,21] is crucially dependent on the existence of a commensurate antiferromagnetic order; the $(0,0,\pi)$ order observed in the actual materials would not lead to the same sort of phase separation, because it would permit metallic in-plane conduction.
B. Numerical results

By solving the dynamical mean field equations numerically, we have computed the ferromagnetic transition temperature $T_c$ as a function of the Hund’s coupling. Fig. 4 shows ferromagnetic $T_c$ vs $J$ for fillings of $n = 0.7$ (solid circles) and $n = 0.25$ (squares), for doubly degenerate $e_g$ orbitals ($n_{orb} = 2; n/n_{orb} = 0.35$ or 0.125). As we noted in the analytic treatment of the low $J$ limit, for not too low densities ($n > 0.2n_{orb}$), a ferromagnetic solution cannot be sustained. Thus for a modest $n$, as $J$ is decreased the ferromagnetic $T_c$ vanishes and the ground state changes from ferromagnetic to incommensurate to antiferromagnetic. As noted previously, we find $n_c \approx 0.195n_{orb} \approx 0.2n_{orb}$. We therefore suspect that the $n = 0.2(n_{orb} = 1)$ curve in the $T_c - J$ diagram of Ref. 3, which indicates that $T_c$ drops to 0 at a finite $J$ of order 1, is incorrect at the low $J$ end; the FM solution should be sustainable down to a very small $J$.

The stars in Fig. 4 correspond to Eq. (24), with the chemical potential $\mu = -1.52t$ for $n = 0.25; n/n_{orb} = 0.125$ (the curve is magnified in the inset). We notice that one cannot sustain a ferromagnetic transition for $n = 0.7$ as $J \to 0$.

The ferromagnetic transition temperature has an apparently complicated dependence on interaction strength and doping. We now show that this seemingly complicated dependence simply reflects the variation of the kinetic energy, $K$, with these parameters. Fig. 5 plots $T_c$ against the change in kinetic energy between the paramagnetic and the $T = 0$ ferromagnetic state ($\Delta K = K(0) - K(T_c)$), both being normalized by the $K$ of the noninteracting ($J = 0$) system at $T = 0$.

FIG. 4. $T_c$ vs $J$ plot of the double-exchange model for doubly degenerate $e_g$ orbitals. The $n = 0.7; n_{orb} = 2$ curve is relevant for La$_{0.7}$A$_{0.3}$MnO$_3$ compounds. For $n \leq 0.39$, the model can sustain a ferromagnetic solution down to the lowest $J$. The inset amplifies the low $J$ end of the $n = 0.25$ curve, to illustrate how the numerics (open boxes) patch on to the analytical expression (stars) from Eq. (24). The solid lines are a guide to the eye. $T_c(J = \infty) = 0.079$ for $n = 0.7$ and 0.055 for $n = 0.25$.

![FIG. 4. T_c vs J plot of the double-exchange model for doubly degenerate e_g orbitals. The n = 0.7; n_orb = 2 curve is relevant for La_0.7A_0.3MnO_3 compounds. For n ≤ 0.39, the model can sustain a ferromagnetic solution down to the lowest J. The inset amplifies the low J end of the n = 0.25 curve, to illustrate how the numerics (open boxes) patch on to the analytical expression (stars) from Eq. (24). The solid lines are a guide to the eye. T_c(J = \infty) = 0.079 for n = 0.7 and 0.055 for n = 0.25.](image)

FIG. 5. $\Delta K = K(0) - K(T_c)$ vs. $T_c$ for the DE model for $n = 0.7; n_{orb} = 2$. Both the quantities are scaled with the $K$ for the non-interacting system at $T = 0$ ($J_{\infty}(T = 0) = 1.01t$ for $n = 0.7$).

To understand this curve, consider the general expression for the free energy, $F = E - TS$ of $H_{DE}$ (Eq. 1), where $S$ is the entropy and $E = -K - J$ where $K$ is given by Eq. (25) and

$$J = \sum_{i,\alpha,\beta} J \left( \vec{S_i} \cdot \vec{d_{i\alpha}} \sigma_{\alpha\beta} d_{i\beta} \right)$$

(28)

where the latter expression is the dynamical mean-field result. Entropy favours the disordered state; ferromagnetic order is driven by a decrease in energy. $T_c > 0$ implies $E_{fm} < E_{nm}$, where $E_{fm}$ is the ground state energy of the ferromagnet and $E_{nm}$ that of "non-ferromagnet" (incommensurate or antiferromagnetic order). At low $T$ all spins are aligned, and carriers can move freely, while at high $T$ the random spin arrangement and large $J$ means carrier hopping is somewhat blocked. $T_c$ is therefore set by the change in $K(T_c)$, which is a simple number ($\approx 1/3$) of $K(T = 0)$ as $J \to \infty$.

As $J \to \infty$, the local spin is always parallel to the core spin and $J$ does not change between ferromagnetic and paramagnetic states. The transition is entirely driven by the change in $K$ between the paramagnetic and ferromagnetic states. In the non-ferromagnetic (NM) phase the band is narrower than in the FM phase, but at finite $J$ the electron has some possibility to hop onto the "wrong" spin site. This is equivalent to the on-site mag-
netization not being saturated in the NM phase, thus \( J_{fm} > J_{nm} \). Since \( T_c \sim E_{nm} - E_{fm} = \Delta K + \Delta J \), it implies that \( E_{fm} = E_{nm} \) when \( T_c = 0 \) and we get \( K_{fm} < K_{nm} \) at this point. So at high \( J \), we start with fully polarized bands and with the magnetic state having more kinetic energy. The high \( J \) expansion suggests that everything comes from virtual hops: \( \Delta K \sim t^2/J \) and \( \Delta J \sim \Delta n \sim J \cdot t/J \sim t \). Since \( \Delta J/J \) is independent of \( J \) at large \( J/T_c \), we consider the Sokolov limit of \( J \) for \( J \gg 1 \). As we reduce \( J \) to a value where the polarization isn’t complete, first \( K' \)’s cross and then \( T_c \) → 0. We have used the dynamical mean-field equations to determine the points at which \( E_{fm} = E_{nm} \) in Fig. 2. For \( n = 0.7(n/\text{orb} = 0.35) \) the critical \( J \approx 0.6t \).

Since \( K \) is of crucial interest to us, we present its values at \( T = 0 \) for the soluble limits \( J = 0, \infty \) in Table I for a variety of fillings. At \( J = 0 \) the system is always paramagnetic, and the kinetic energy increases with \( n \) in the range \( 0 < n < 1 \). For \( J = \infty \) however, the kinetic energy is maximal for \( n = \text{orb}/2 \). As \( K(T_c) = K(0)/\sqrt{2} \) for \( J = \infty \) and the transition temperature is tied to the kinetic energy (Fig. 3), \( T_c \) also is a maximum at the same filling, as has been noted in earlier works.

Table I. The kinetic energy \( K \) at \( T = 0 \), evaluated using a dynamical mean-field method with a semicircular density of states, for \( J = 0 \) and \( J = \infty \) at different fillings.

| \( n/\text{orb} \) | \( K(J = 0)/\text{orb} \) | \( K(J = \infty)/\text{orb} \) |
|------------------|-----------------|-----------------|
| 0.125            | 0.2160          | 0.1958          |
| 0.25             | 0.3016          | 0.3248          |
| 0.35             | 0.5086          | 0.3889          |
| 0.50             | 0.6496          | 0.4244          |
| 0.75             | 0.7994          | 0.3248          |

VI. OPTICAL CONDUCTIVITY

Next we discuss the optical conductivity of the double-exchange model. At large \( J \) the density of states of this model consists of two nearly semi-circular bands corresponding to conduction electrons parallel (\( \uparrow \)) and antiparallel (\( \downarrow \)) to the core spin. The bands are separated by an energy \( 2J \). This structure is shown in Fig. 4, which we now use to give a qualitative discussion of \( \sigma \).

We have argued that the kinetic energy is the crucial parameter determining \( T_c \). To further substantiate this we show in Table II results obtained for \( T_c(J \rightarrow \infty) \) by a variety of techniques in a range of models \( d = 3 \), expressed in terms of \( K_{d=\infty}(T = 0) \). Roder et al. used a series expansion technique. Yunoki et al. studied thermodynamic properties of the classical spin model in \( d = 3 \) with a single orbital nearest neighbour hopping using Monte Carlo on \( 6 \times 6 \times 6 \) clusters. Calderon and Brey have performed similar Monte Carlo calculations on \( 4 \times 4 \times 4 \) and \( 20 \times 20 \times 20 \) lattices, and argued that Yunoki et al. underestimated \( T_c \) by a factor of \( \approx 1.6 \). The results of Calderon et al. are in agreement with those of Roder et al. Note that in all calculations the doping dependence of \( T_c \) is essentially the doping dependence of the kinetic energy.

We now consider the dynamical mean-field results. These yield somewhat higher \( T_c/K \) results, as expected because fluctuations are neglected. Calderon and Brey partitioned the hopping term \( t_{ij} \) into average (\( \langle \bar{t}_{ij} \rangle \)) and random (\( \delta t_{ij} \)) components and argued that fluctuations lead to a \( \approx 25\% \) correction to \( T_c \). Applying this correction leads to the numbers given in brackets, which are in good agreement with those found in Refs. 7 and 18.

Table II. \( T_c/K(T = 0) \) for \( J = \infty \); comparison of the different methods. The bracketed terms for \( d = \infty \) indicate \( T_c \) values that are \( \approx 25\% \) fluctuation corrected.

| Method       | \( n/\text{orb} \) | \( T_c/K_{d=\infty}(T = 0) \) |
|--------------|-----------------|-----------------|
| \( d=\infty \) | 0.5             | 0.070(0.053)     |
| Series       | 0.25            | 0.063(0.050)     |
| Series \( d=\infty \) | 0.5             | 0.036            |
| Yunoki       | 0.25            | 0.033            |
| Calderon     | 0.5             | 0.056            |
| Calderon \( d=\infty \) | 0.25            | 0.056            |

FIG. 6. The large \( J \) density of states, consisting of bands of electrons parallel (\( \uparrow \)) and antiparallel (\( \downarrow \)) to the core spin. The bands are separated by an energy \( 2J \). The "wrong-spin" transitions become allowed as \( T \) is increased from 0. For large \( J \), these two pieces are well separated.

The crucial point is that the optical process conserves electron spin. At \( T = 0 \) in the fully polarized ferromagnetic state all spins are aligned, making it impossible for an optical process to create a final state with an \( e_g \) electron anti-aligned to a core spin. Further, the perfect spin alignment means that no scattering processes are present; thus the \( T = 0 \) conductivity is simply given by the band theory of the spin polarized \( e_g \) electron manifold. The spectral weight in these transitions follows directly from...
the band theory kinetic energy. Impurities, weakly coupled phonons etc. will change the form of the conductivity but will not significantly affect the total spectral weight.

As $T$ is raised, the spin disorder increases, and an electron moved to an adjacent site may find itself anti-aligned to the new core spin.

Thus as $T$ is raised the resulting spin disorder leads to a broadening of $\sigma$, a decrease in total spectral weight, and also to a shift of oscillator strength to the peak at $\sim 2J$. For $2J \gg t$, the wrong-spin peak is well separated from the parallel spin transitions and we shall derive its spectral weight in this limit.

The wrong-spin transitions produce the peak at $2J$, $\sigma(2J)\sim J$ also to a shift of oscillator strength to the peak at $\sim 2J$. For $J >> t$, the inter-band weight falls off as $J^{-3}$. The straight line has a slope of $n/2$, corresponding to the large $J$ analytic expression Eq. (31).

Now, $A_+$ is an analytic function and its integral gives the real part evaluated at $\omega = J$, which is $1/(2J)$ in the large $J$ limit. Thus

$$\int \frac{2}{\pi} d\Omega \sigma(\Omega) = \int \frac{d\epsilon_n (\epsilon_n^2 - 4)}{12\pi J} \int_{-\infty}^{\mu} \frac{d\omega}{\pi} A_-(\epsilon_n, \omega).$$

The $\omega$-integral gives the momentum space occupancy $n(\epsilon_n)$. If the scattering is strong enough, this is just $n$. So, for large $J$ and at the magnetic transition we get

$$\int \frac{2}{\pi} d\Omega \sigma_{2J}(\Omega) = \frac{n}{2J}$$

which falls off as $1/J$ and is proportional to the filling $n$. The numerical results are shown in Fig. 7(b); we see that the analytic value for the slope, $n/2 = 0.35$ (straight line) from Eq. (29) overlays the numerical curve for large values of $J$. This analysis shows that the spectral weight in the wrong-spin transitions depends crucially on $n$. A previous calculation of $\sigma$ used $n_{orb} = 1$ and $n = 0.7$. The resulting oscillator strength in the wrong-spin transition is therefore not applicable to manganites with $n/n_{orb} = 0.35$ (and indeed the oscillator strength in this case is larger than the one we calculated for the physical $n/n_{orb}$ by a factor of two). To aid in comparison to experiments we give in Table. III the total $T = 0$ spectral weight, the total $T > T_c$ spectral weight and, for $T > T_c$, the spectral weight in the same-spin and wrong-spin transitions, calculated using the dynamical mean field theory for various values of $J$.

**TABLE III.** The total $T = 0$ spectral weight $K(0)$, the total $T > T_c$ spectral weight $K(T_c)$, and $K_{ant}$ and same-spin ($K_{par}$) transitions for various values of $J$, and $n = 0.35 n_{orb}$.

| $J$ | $K(0)$ | $K(T_c)$ | $K_{ant}$ | $K_{par}$ |
|-----|--------|----------|-----------|-----------|
| 1   | 0.7778 | 0.8080   | 0.1209    | 0.6850    |
| 2   | 0.7778 | 0.6994   | 0.1067    | 0.5926    |
| 3   | 0.7778 | 0.6541   | 0.0833    | 0.5707    |
| 4   | 0.7778 | 0.6291   | 0.0661    | 0.5629    |
| 5   | 0.7778 | 0.6136   | 0.0547    | 0.5588    |
| 6   | 0.7778 | 0.6030   | 0.0467    | 0.5562    |
| 7   | 0.7778 | 0.5952   | 0.0408    | 0.5543    |
| 11  | 0.7778 | 0.5778   | 0.0275    | 0.5502    |
VII. COMPARISON WITH EXPERIMENTS

A. Overview

In this section we relate our results to experiments. The measurements we analyse are the value of the ferromagnetic transition temperature and the magnitude and temperature dependence of the spectral weight (integrated area) in different regions of the optical conductivity spectrum. The physics issues we are able to clarify include the extent to which the double-exchange-only model describes the behaviour of the manganites, the proper interpretation of the optical spectrum, and the value of the Hund’s coupling. The remainder of this section is organized as follows. In sub-section B we summarize our results in a manner suited to comparison to data, in sub-section C we outline what is known about unresolved issues arising in interpretation of experiments and in sub-section D we present the comparison between our results and data.

B. Summary of theoretical results

We have studied the double-exchange-only model defined by Eq. 1. This model involves itinerant electrons hopping among sites of a lattice and coupled ferromagnetically to electrically inert core spins. No other interactions are explicitly included. This model captures some aspects of the CMR manganite perovskites and of other ‘double-perovskite’ systems. Whether other physics is important is the subject of present debate.

The double-exchange-only model was shown to have two crucial parameters: the electron kinetic energy $K$ defined in Eq. 1 and the itinerant electron-core spin coupling $J$. $K$ is the expectation value of a local operator. It depends on parameters like temperature and $J$, but is insensitive to details of band structure. For the $J$ values and carrier concentrations of physical relevance, the ground state is a fully polarized ferromagnet and $K(T = 0)$ is independent of $J$ and in the double-exchange-only model may be computed via a simple band structure calculation. The value appropriate to La$_{0.7}$Sr$_{0.3}$MnO$_3$ is $K \approx 0.84$eV. We calculate $T_c$ as a function of $K(T = 0)$ and $J$ using a mean field method which treats local dynamics exactly and by comparison to published Monte Carlo calculations, were able to estimate the corrections to $T_c$ from non-mean-field spatially dependent fluctuations. The dynamical mean field results are often published in terms of a bandwidth parameter $t$; the $t$ corresponding to the band theory $K$ is $t \approx 1.07$eV.

By comparing our results to those obtained by other techniques, we found that in the $J \to \infty$ limit $T_c/K(T = 0) \approx 0.16$, essentially independent of model details. For finite $J$, we found (for carrier concentrations relevant to the CMR materials with ferromagnetic ground states) that $T_c$ was linearly proportional to the change, $\Delta K$, of $K$ between $T_c$ and $T = 0$. We further found that as $J \to \infty$, $\Delta K/K \to 1/3$; as $J$ is decreased, $\Delta K$ decreases. At a critical $J$, $\Delta K = 0$ and below this $J$ a ferromagnetic ground state cannot be sustained.

As previously noted, $K$ is measurable in optical experiments, and therefore a quantitative test of the double-exchange-only model is possible and conversely constraints on optical conductivity may be extracted from measured $T_c$’s. The issues involved in this analysis will be treated in the next sub-section. To conclude the present sub-section we discuss the additional information concerning the strength of non-double-exchange interactions which may be obtained from the comparison of $T_c$ and the measured $K(T)$.

We may represent the electronic energy of a solid, $E_{el} = -K + I$ where $K$ was defined in Eq. 1 and $I$ represents the expectation value of all interactions, including the $J$ term and other interactions not included in the double-exchange model. In the rest of the discussion we assume $J$ is large enough to have a spin-polarized ground state.

In the double-exchange-only model at $T = 0$, $K$ is saturated and given by the band theory value (assuming spin-polarized electrons). As $T$ is increased, the entropy driven spin-disorder leads to a reduction in $K$; the change in $K$ between $T = 0$ and $T > T_c$ depends on $J$, and becomes as large as $1 - 1/\sqrt{2} \approx 30\%$ in the $J \to \infty$ limit. Other interactions do not commute with $K$, and therefore change the electronic state in a way which reduces $K$. Thus, we argue that $K$ cannot be greater than the spin-polarized band theory value, and that a $K(T = 0)$ appreciably less than this value indicates other interactions are important.

As temperature is increased from $T = 0$, spin disorder leads to a decrease in $K$ and therefore to an increase in the relative strength of the interaction terms which in turn causes a further decrease in $K$. This self-consistent effect says that for a given $J$, the relative change $\Delta K/K$ in kinetic energy between $T_c$ and $T = 0$ increases with increasing interaction strength. This physics was investigated in Ref. 4 for $J = \infty$ in the particular case of electron-phonon interaction, but is expected to be more general. Further, because kinetic energy is what decides $T_c$, the interaction induced decrease in $K$ must decrease $T_c$ below the value predicted by double-exchange.

To summarize, the double-exchange-only model predicts a definite set of relationships between $T_c$, $K(T = 0)$, and the $T$-dependence of $K$ and $\Delta K$. These are summarized in Fig. 3 and Tables. II and III.

Adding other interactions causes $T_c$ and $K(T = 0)$ to decrease and $\Delta K$ to increase.
C. Optical conductivity

This subsection discusses the interpretation of optical conductivity data. As we have shown, the magnitude and temperature dependence of the spectral weight in the $e_g$ contribution to the optical conductivity contains crucial information about the electron kinetic energy and Hund’s coupling. In order to extract this information one must identify the $e_g$ contribution to the measured conductivity, which is not straightforward because of the overlap between the $e_g$ transitions of interest and processes involving other bands. In this subsection we analyze the issues arising in the interpretation of the measured conductivity, with emphasis on how our theoretical results can be used to resolve some of the difficulties in interpreting data.

The optical conductivity $\sigma(\omega)$ is the linear response function relating spatially uniform frequency dependent current $j(\omega)$ to applied electric field $E(\omega)$. In simple terms, the conductivity describes how electrons move in response to an electric field, and therefore contains information about interactions which may hinder this motion. In metals it is useful to distinguish between intra-conduction-band processes (those which involve scattering of electrons between conduction band states, which in many cases including the CMR manganites are the states of immediate physical interest) and other processes which involve scattering of electrons from conduction band states to other (empty) bands, from other (filled) bands to the conduction bands, or which do not involve the conduction bands at all. The other processes are usually called interband, but in the manganites there are two orbitals per unit cell and therefore some of the intra-conduction-band processes are, strictly speaking, interband.

In any event, one would like to extract from the measured conductivity the portions pertaining to transitions between the states of interest (in the manganite case, the intra-conduction-band processes) and analyse only these. However in many cases involving transition metal oxides it has not been clear how to separate the interesting conduction band contributions from other un-interesting processes. The CMR materials are a promising system in which to investigate this issue because the conduction band contribution to $\sigma$ has a strong temperature dependence and a definite relation to $T_c$, which can be used to distinguish it from other contributions.

The main point is this: the double-exchange-only model predicts a definite relation between the $T = 0$ $e_g$ oscillator strength and $T_c$. Interaction corrections only reduce $T_c$ below the double-exchange value and $K$ below the band theory value. Therefore the part of the optical spectrum assigned to the $e_g$ electrons must contain at least enough kinetic energy to reproduce the observed $T_c$, but cannot contain more kinetic energy than predicted by spin-polarized band theory results.

A further constraint is provided by the change $\Delta K$ in kinetic energy between $T = 0$ and $T_c$. For $H_{DE}$ (Eq. 1), $\Delta K \leq 0.3K(T = 0)$ and the maximal value occurs as $J \to \infty$. Limited information is available concerning models with additional interactions, but published calculations for the double-exchange plus phonon problem at $J \to \infty$ show that the fractional change $\Delta K/K(T = 0)$ can become slightly larger than 0.5, but the magnitude of $\Delta K$ is never much larger than the double-exchange-only value. This is useful because the change in optical spectral weight can be accurately measured, and the observed changes can with confidence be attributed to the $e_g$ electrons of interest. A final constraint comes from the position and spectral weight of the “wrong-spin” transitions. The point is that as $T \to 0$, all of the spins are aligned in both the ground state and all states accessible from it via the optical matrix element. However, for $T > T_c$, the core spins are completely disordered and therefore when an electron hops (or is pushed via the optical matrix element) from one site to another it has a probability for landing in the “wrong-spin” configuration, i.e. with $e_g$ electron anti-parallel to the core. The probability depends on the hopping matrix element (i.e., the $T = 0$ kinetic energy) and the value of $J$. This physics was first pointed out by Okimoto et al., who, however, obtained what we argue below was an incorrect value of $J$. We have determined the optical oscillator strength in this transition, as a function of $K(T = 0)$ and $J$. The oscillator strength has a strong dependence on carrier density not noticed in previous work.

D. Analysis of data

Optical conductivity has been measured in a wide range of manganites by several groups. Results are qualitatively similar, but there are substantial quantitative differences between results of different groups. Some of the differences seem to be experimental artifacts associated with surface preparation, others seem to relate to differences in physics among various members of the CMR family of materials. As we shall see, our results provide consistency checks which allow one to separate artifacts from intrinsic behaviour and to make some statements about the underlying physics.

To be concrete, we discuss the data of Quijada et al., who measured $\sigma(\omega,T)$ for three pseudo-cubic manganite films La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO), La$_{0.7}$Ca$_{0.3}$MnO$_3$ (LCMO) and Nd$_{0.7}$Sr$_{0.3}$MnO$_3$ (NSMO). The qualitative features of the data are (i) at $\omega \leq 3eV$, an absorption with a pronounced frequency dependence and an intensity that shifts to lower frequency and increases markedly as $T$ is decreased from $T_c$ to a low temperature, (ii) a strong feature centered at $\omega \approx 4eV$ with little apparent $T$-dependence, and (iii) a weak feature at $\omega \approx 3eV$, visible as a decrease in absorption intensity as $T$ is decreased below $T_c$. The interpretations offered in Ref. were that (1) the integral of the low $T$ conductivity between $\omega = 0$
and $\omega = 2.7\text{eV}$ was a good representation of the total low $T$ conduction-band spectral weight, (2) that the strong feature at $\omega \approx 4\text{eV}$ was a Mn-O interband transition and (3) the weak feature at $\omega \approx 3\text{eV}$ was the "peak at $J$" (it is actually at $2J$ in our units, as in Fig. 3) due to "wrong-spin" transitions characteristic of the spin disordered state. This identification is controversial. Okimoto et al. identified a lower energy feature as the peak at $J$.

We begin our discussion with the "peak at $J$". The identification offered by Okimoto et al. implies $J \approx 0.1-0.13\text{eV}$ in LSMO, $0.1-0.12\text{eV}$ in LCMO and $0.1\text{eV}$ in NSMO. The changes that we find for $J/t > 2$ are of the order of $1-2\%$, too small to explain the observed feature. An alternative possibility is a polaronic shift associated with a fluctuation corrected for these materials as well. The lower $T_c$'s ($\approx 270\text{K}$ for LCMO, $\approx 250\text{K}$ for NSMO) would then imply $J = 1.5 - 1.8\text{eV}$, again yielding a $\Delta K$ that is too small, and a "peak at $J$" visible in the spectrum.

We would like to add here that our values of $K$ are a factor of 3 higher than that of Quijada et al. This is because of the $1/\sqrt{d}$ dependence of each current vertex in the expression for $\sigma(\omega)$. Thus, for $d = 3$, the kinetic energies of Quijada et al should be multiplied by $(\sqrt{d})^2 = 3$ to compare with our numbers. We do this in Table IV for facilitate comparison.

To conclude this section we invert the logic, using the observed transition temperatures and $\Delta K$ to obtain bounds and estimates for the conduction band spectral weights. We argue that for a given $K(T = 0)$ and $J$, the double-exchange-only model gives an upper bound for $T_c$. Thus values of $T_c$ and $J$ yield a lower bound for $K(T = 0)$. These bounds are shown in the Table IV for $J = 2\text{eV}, 3\text{eV}$ and $4\text{eV}$ for the systems studied by Quijada et al. Note that an upper bound of $K(T = 0)$ is given by the band theory value.

An alternative estimate may be obtained by considering the limit $J \rightarrow \infty$. In this case, as noted in Figs. 3, $T_c$ is a universal function of $K(T_c)$ (which may itself be affected by other interactions). Further, $T_c$ must decrease as $J$ is decreased; thus, we may obtain a lower bound on $K(T_c)$ from the $J = \infty$ result for $T_c$. These bounds are also listed in Table IV.

| $K_{\text{min}}(T_c)$ | $J = 2\text{eV}$ | $J = 3\text{eV}$ | $J = 4\text{eV}$ |
|-----------------------|-----------------|-----------------|-----------------|
| $K_{\text{min}}(0)$  | $6.30$          | $5.35$          | $5.14$          |
| $K_{\text{expt}}(0)$ | $780$           | $660$           | $639$           |
| $K_{\text{expt}}(T_c)$ | $477$         | $393$           | $390$           |

It is interesting to note that the experimental $K(T_c)$ for LSMO is $477\text{meV}$, which exactly saturates the lower
bound on $K(T_c)$ from double-exchange. Because the infinite-$J$ double-exchange-only $T_c$ is expected to be the upper bound to the true $T_c$ of an interacting model we expect that in the actual material the $J$ is larger than 2eV (so the model is not far from the $J = \infty$ limit and we suspect that the integration up to $\omega = 2.7$eV does not capture quite all of the spectral weight.

VIII. CONCLUSIONS

We have given a complete and correct treatment of the phase diagram, kinetic energy and some aspects of the optical conductivity of the double-exchange-only model of mobile carriers coupled to core spins. We have determined the physics operating in different regions of the phase diagram and have demonstrated the importance of choosing parameters (especially carrier density) appropriate to the materials of interest by exhibiting the incorrect results obtained by the use of wrongly chosen parameters. We have shown that the crucial quantity is the electron kinetic energy and have used our results to estimate the kinetic energy of several manganite systems. Our results also provide insight into the crucial question of which portions of the spectrum pertain to the low energy electronic degrees of freedom.

There are several directions for future work. One is to combine the dynamical mean field method with a realistic band structure, to obtain a treatment of the frequency dependence of $\sigma$. Another is to employ the methods presented here to models where double-exchange is combined with other interactions. If this were carried through, it seems likely that the combination of $T_c$ and the changes in optical spectral weight could be analysed to provide detailed knowledge of the strength, energy scale, and nature of any additional couplings.

IX. ACKNOWLEDGEMENTS

We thank H.D. Drew, B.G Kotliar and H. Monien for helpful discussions and the University of Maryland MR-SEC and NSF-DMR-9705482 for support.

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