Dynamical correlations in half-metallic manganites

L. Craco\textsuperscript{1}, M. S. Laad\textsuperscript{2} and E. Müller-Hartmann\textsuperscript{2}

\textsuperscript{1} Max Planck Institute for the Physics of Complex Systems, D-01187 Dresden, Germany
\textsuperscript{2} Institut fuer Theoretische Physik, Universitaet zu Koeln, Zuelpicher Strasse, D-50937 Koeln, Germany

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Motivated by the recent optical and photoemission measurements on half-metallic ferromagnetic three-dimensional manganites, we combine a tight-binding fit of the one-particle bandstructure with the dynamical mean-field theory, which treats the dynamical orbital correlations and the combination of Jahn-Teller and doping-induced disorder on the same footing. We show how all of the above effects are necessary to obtain good semi-quantitative agreement with experimental features. As applications, we show how modest external magnetic fields drive drastic changes in the optical spectrum, demonstrating the colossal ac magnetoresistivity. The photoemission lineshape contribution is evaluated, and good agreement with published experimental work is found.

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In view of its potentially large technological applications, the colossal magnetoresistance (CMR) has attracted renewed attention in the past few years. The sensitivity of transport and optical response to small magnetic fields promises to open up new, attractive applications in optoelectronic devices\textsuperscript{[1]}. A proper analysis of this problem requires combination of realistic material aspects (captured in one-electron bandstructure) with strong correlation effects, which are known to determine the physical response of transition metal compounds. Such approaches, recently been applied\textsuperscript{[2]} to the study of photoemission spectra of materials believed to undergo filling-driven Mott transitions, have shown the importance of including proper bandstructure effects into a state-of-the-art correlation calculation. However, generically, one has to deal with the added complication of orbital degeneracy and associated Jahn-Teller distortions, which maybe static or dynamic\textsuperscript{[3]}. This is hard in practice, requiring one to deal with many bands-in this situation, a simpler approach to model the actual bandstructure, keeping the electronically active states intact, is an attractive option. This is the main motivation of the tight-binding (TB) fit to the actual complex bandstructure, a strategy that has been used with success for the case of cuprates\textsuperscript{[4]}.

In this work, we use this strategy to study the field-dependent optical conductivity of well-doped manganites. We concentrate on the well-doped, half-metallic ferromagnetic state in the cubic manganite La\textsubscript{0.7}Ba\textsubscript{0.3}MnO\textsubscript{3}\textsuperscript{[5]}. We combine the TB-fit with dynamical mean-field theory (DMFT) to treat the dynamical effects of local correlations with essential bandstructure features in a self-consistent way. We show how all the above effects are crucial to obtain a consistent picture of the field-dependent optical response and explicitly demonstrate the phenomenon of colossal optical magnetoresistivity. We also compute the photoemission line-shapes, pointing out its possible relevance to the study of the role of orbital correlations in the CMR materials.

We start by noticing that in the full linear augmented plane wave (LAPW)\textsuperscript{[6]} calculation, the majority $e_g$ bands, providing the conduction states, split from the $t_{2g}$ bands, but overlap with the $O$ $p$ bands. However, the two $e_g$ bands are clearly identified along the symmetry directions, making a mapping to an effective two-band model possible. Constructing Mn-centered Wannier functions with strong $O$ $p\sigma$ character, only the $ddg = t_\sigma$ and $ddb = t_\delta$ overlaps are nonzero. Using Slater-Koster tables for the cubic perovskite structure, the $e_g$ bands are: $c^\pm_k = c_k \pm D_k$, where $c_k = (t_\sigma - t_\delta)(c_x + c_y + c_z)$,

$$D_k = -(t_\sigma - t_\delta)\sqrt{c_x^2 + c_y^2 + c_z^2 - c_x c_y - c_y c_z - c_z c_x},$$

and $c_{\alpha} \equiv \cos(k_{\alpha} a)$. The full $e_g$ bandwidth is $2D = 6(t_\sigma + t_\delta)$. To fix parameters above, notice that the dispersion along $\Gamma - X$ is equal to $4t_\delta$. Since the overlap of the majority $e_g$ bands is omitted in the two-band fit, one uses the average value of the $\Gamma - X$ dispersion, giving $4t_\delta = -0.12$, or $t_\delta = -0.03$ eV. From $D = 2.15$ eV, one has $t_\sigma = -0.69$ eV. The Fermi level is set to zero by choosing $\epsilon = 0.81$ eV, completing the simple TB fit to the actual LAPW bandstructure. The corresponding total DOS\textsuperscript{[7]} is clearly different from the model DOS used in usual model hamiltonian treatments, and describes the doubly-degenerate $e_g$ bandstructure in the cubic perovskite geometry. Starting from similar form for the bandstructure Weife et al.\textsuperscript{[8]} concentrate on the polaronic physics while ignoring the undoubtedly strong local, orbital correlations in the FM phase\textsuperscript{[9]}. It is precisely our aim to show how a combination of strong correlations with Jahn-Teller distortion-induced orbital disorder and proper one-electron bandstructure is required for a qualitative (in our case, we found even semi-quantitative) agreement with published experimental results.

With the single-particle dispersion relation from the TB-fit, the one-electron part of the hamiltonian is,

$$H_0 = \sum_{k\alpha} \epsilon_{k\alpha} c^+_k c_k, \quad \epsilon_{k\alpha} = \epsilon_{\alpha} + \sum_{\beta} V_{\alpha\beta} c^+_k \sum_{\delta} V_{\delta\beta} c_k,$$

where $\alpha = a, b$ label the orbital indices for the doubly-de-
generate $e_g$ sector for the case of the cubic manganites. The usual spin index is dropped since one of the spin species is projected out of the problem in the double-exchange limit. In this situation, the interaction part of the Hamiltonian containing inter-orbital coulomb interaction and the Jahn-Teller coupling is given by:

$$H_{int} = U_{ab} \sum_i n_{ia} n_{ib} - g \sum_i (Q_{i2} \tau_i^2 + Q_{i3} \tau_i^3) + \frac{k}{2} \sum_i (Q_{i2}^2 + Q_{i3}^2).$$

The $J_H \to \infty$ limit introduces a DE projection factor into the hopping term, which depends on the nearest neighbor core-spin correlation function: $t_{ij} \to t_{ij}/1 + <\mathbf{S}_i, \mathbf{S}_j>/2S^2$, and is temperature and magnetic-field dependent. The one-particle part is now written as:

$$H_0 = \sum_{\langle ij \rangle \neq \alpha \beta} t_{ij}^{\alpha \beta} (M) (c_{i \alpha}^\dagger c_{j \beta} + h.c.).$$

The total Hamiltonian of the system in the FM phase is thus $H = H_0 + H_{int}$. To simplify matters further, we make a local rotation in the two-dimensional $Q_2 - Q_3$ space, so that the local quantization axis is parallel to $z$. One uses a local unitary transformation $U_i^\dagger [Q_i, \bar{\Sigma}_i] U_i = Q_i \tau_i^3$ and simultaneously transforms the electronic operators as $a_{ia} = U_i c_{i \alpha}$. The Hamiltonian then reads,

$$H = - \sum_{\langle ij \rangle \neq \sigma \sigma'} t_{ij}^{\sigma \sigma'} (M) U_i^\dagger U_j (a_{i \sigma}^\dagger a_{j \sigma'} + h.c.) + U_{ab} \sum_i n_{i \uparrow} n_{i \downarrow} - g \sum_i Q_i \sigma n_{i \sigma},$$

omitting the purely phononic part, and relabelling $a_{i \uparrow} = a_i$ and $a_{i \downarrow} = b_i$. With this, we have simplified the two-band model in the DE limit to a Hubbard-like model with a disordered "magnetic field" whose source is the disordered Jahn-Teller distortions which exist in the well-doped FM state at low $T$.

A plausible parameter range for the problem at hand is $U_{ab} > 2D$ and $Q \equiv \sqrt{V_0} \leq U_{ab}$, requiring one to treat the dynamical effects of strong local correlations and repeated scattering produced by moderately strong, local disorder on the same footing. This is achieved by using the DMFT as an approximation to our 3d problem. In $d = \infty$, the full lattice problem is mapped onto a single site problem, with the "impurity" embedded self-consistently in a dynamical bath. In the multiband case, and without symmetry breaking (orbital ordering in our case), the Green function and the purely local self-energy are functions of frequency only: $G_{nm'} (\omega) = G (\omega) \delta_{nn'}$ and $\Sigma_{nn'} (\omega) = \Sigma (\omega) \delta_{nn'}$. In this situation, the DMFT self-consistency condition becomes,

$$G_{nm'} (\omega, M) = \frac{1}{N} \sum_k \frac{1}{\omega - \Sigma (\omega) - H_{B} (k, M) - \delta_{nm'}}.$$

The remaining problem is to compute the single-particle self-energy, $\Sigma (\omega, M)$, in a situation where strong electronic correlations and disorder-induced strong scattering are simultaneously dominant, as above. We treat the strong orbital correlations at the local level using the iterated perturbation theory (IPT) away from half-filling, and generalized to finite temperature, and the repeated scattering effects of the local Jahn-Teller (disordered) distortion by a proper combination of the IPT with the coherent-potential approximation (CPA), which solves the static disorder problem exactly in $d = \infty$.

In this procedure, the IPT self-energy is fed into the CPA local potential, which now becomes $V_i = v_i - \Sigma_{int} (\omega) - \Sigma_{CPA} (\omega)$. The IPT local propagator is then used along-with this in the disorder-averaged $T$–matrix (CPA) equation (but with $\Sigma (\omega) = \Sigma_{int} (\omega) + \Sigma_{CPA} (\omega)$): the local GF used in the CPA equation is

$$G (\omega, M) = \frac{1}{N} \sum_{k, \pm} \omega - \Sigma_{int} (\omega, M) - \Sigma_{CPA} (\omega, M) - \epsilon_k^\pm.$$

Solution of the CPA equation $\langle T_{ii} [\Sigma (\omega)] \rangle = 0$, gives the total self-energy corrected simultaneously for scattering caused by interactions (IPT) and disorder (CPA). This is fed back into the IPT subroutine that calculates a new self-energy, $\Sigma_{\text{new}} (\omega)$ and a new local GF, $G_{\text{new}} (\omega)$. These are fed back into the modified CPA routine, and the procedure is iterated to selfconsistency. The interacting density of states (DOS) is then obtained from the usual equation, $\rho (\omega, M) = -Im G (\omega, M)/\pi$.

We now present the results. Below, we concentrate on a non-half-filled band $n = 0.6$ at low temperature regime $T = 0.01D$. At "half-filling", the true state is a Neel-ordered orbital antiferromagnet with additional correlated distortions coming from the JT coupling. To study the well-doped FM regime with "melted" orbital order, we consider only the para-orbital state. In Fig. 1 (a), we show results for the local spectral function (DOS) for $U_{ab} = 2.0D$. For $\nu = 0$, it shows features associated with the development of a correlated Fermi liquid metal associated with collective screening of "orbital" moments, in analogy with what happens in the usual $d = \infty$ Hubbard model. However, we observe a rich structure of the DOS directly related to the use of the TB-bandstructure instead of idealized model input bandstructure used in model hamiltonian treatments.

The effect of moderate local JT disorder is modeled by a binary disorder distribution: $P (Q_i) = x \delta (Q_i) + (1 - x) \delta (Q_i - Q)$, where $x$ is the concentration of the $Mn^{3+}$ sites upon doping. This is justified because hole doping creates locally JT-inactive $Mn^{3+}$ sites, which are randomly distributed in the host lattice of JT active $Mn^{4+}$ ions. To make close contact with experiment, we
choose \( x = 0.3 \). In Fig. 1 (a) we show our results for the two values of \( t_{ij} \), i.e., for \( t_{ij} = D \) and \( t_{ij} = \sqrt{2}D \) and for \( v = 0, U_{ab}/2 \). In particular, beyond a critical \( v = v_c \), the spectrum is completely incoherent, due to strong (resonant) scattering off the disordered JT potentials. The DOS is characterized by a low-energy pseudogap, resulting in an anomalous response.

![FIG. 1](image)

**FIG. 1.** (a) shows the spectral DOS for \( n = 0.6 \) and \( U_{ab} = 2D \). (b) shows the corresponding photoemission lineshape in the non-half-filled regime for \( T = 0.01D \). The two values of the JT disorder and the external magnetic field are shown on the top of the figure.

Next, we turn to the photoemission lineshape: \( I_{PES}(\omega) = f(\omega - \mu)\rho(\omega) \). With \( v = U_{ab}/2 \) and \( t_{ij} = D \), we see only a small “coherent” weight at the Fermi level \( (\mu) \), as shown in Fig. 1 (b). The overall details of the calculated spectrum resemble the experimental result [12] quite well. An external magnetic field, \( H_{ext} = h_{sat} \) results in increased coherence, as shown by the increased coherent component at \( \omega = \mu \). The role of dynamical orbital correlations and JT distortions is again clear. In the pure DE models, one expects a large quasiparticle contribution at low-\( T \), and the DE model combined with disordered JT distortions yields a pseudogapped spectrum with no coherent contribution at low energy. Within our approach, we have succeeded in obtaining the quasicoherent low-energy and the incoherent high-energy satellite features in semiquantitative agreement with experimental work [12]. Notice the additional shoulder structure in our calculations on the higher-energy side of the coherent feature. This is a direct consequence of the realistic features in the TB-fit spectral DOS and would be missed by model bandstructures. Finally, in actual practice, surface effects and possible inhomogeneities in the sample correspond to a larger \( U_{ab} \), further enhancing the pseudogap feature in agreement with experiment.

The optical conductivity is computed from the Kubo formula. In \( d = \infty \), the vertex part does not enter the Bethe-Salpeter equation for the conductivity [13], so that,

\[
\sigma_{xx}(\omega, M) = \sigma_0 \sum_s \rho_0(\epsilon)d_0 \int d\nu D^2(M)A_{Ms}(\epsilon, \omega') \times A_{Ms}(\epsilon, \omega + \omega') \frac{f(\omega') - f(\omega' + \omega)}{\omega}
\]

![FIG. 2](image)

**FIG. 2.** (a) shows the optical conductivity for the same parameters as in Fig. 1 (b). (b) shows our results for the collosal optical magnetoconductivity for two values of the JT disorder.

The calculated \( \sigma_{xx}(\omega) \) in \( d = \infty \), clearly shows up the magnetic-field induced spectral weight transfer in the “bad metal” state above \( T_{cF} \) for \( x = 0.3 \). In Fig. 2 (a), we show \( \sigma_{xx}(\omega, M) \) with the above parameters for two values of \( t_{ij} \), which correspond to \( H_{ext} = 0 \) \( (D(M) = D) \) and \( H_{ext} = h_{sat} \) \( (D(M) = \sqrt{2}D) \), as explained above. With \( H_{ext} = 0 \) and \( v = U_{ab}/2 \), the Drude-like part in \( \sigma_{xx}(\omega) \) at \( T > T_{cF} \) is strongly suppressed. This suppression is caused by the combined effects of the decrease of \( D(M) \) at high-\( T \), the \( T \)-dependent reduction of the spectral weight (due to the \( T \)-dependence of the quasicoherent collective Kondo peak in our Hubbard-like model in \( d = \infty \)) and strong scattering off disordered (local) JT distortions, which by itself would tend to open up a dip feature in the DOS. With \( H_{ext} = h_{sat} \), we see a strong enhancement of the optical spectral weight in the low- and mid-infrared part of the spectrum. This is understood in terms of the physics of the \( d = \infty \) Hubbard-like model, where \( U_{ab}/D(M) \) controls the distribution of spectral weight; for large \( U_{ab}/D(M) \), most of the weight is concentrated in the high-energy part, and a small fraction comprises the coherent part (in the para- orbital metallic state), in agreement with the fact that the resistivity magnitude places the material in the “bad metal” class. With increasing \( D(M) \), high-energy spectral weight is transferred over energies on the scale of \( U_{ab} \) to the quasicoherent part. This transfer scales with the form of \( D(M) \) as \( D^2(M) = D^2(1 + M^2)/2 \), completely consistent with experiment.
What is interesting is that at low energies, the fractional increase \( \Delta \sigma_{xx}(\omega, M)/\sigma_{xx}(\omega, 0) \) is a few hundred percent! (see Fig. 2 (b)). We call this colossal optical magnetoconductivity (COM). Amazingly, exactly such a phenomenon has been observed by Boris et al. [14] for \( La_{0.67}Sr_{0.33}MnO_3 \). In practice, \( H_{\text{ext}} \) is a few teslas, and, alongwith the fully spin-polarized nature of the ferromagnetic metallic state, this phenomenon makes for interesting field-dependent optical applications. We emphasize the crucial role of dynamical orbital correlations and JT effects here: the \( T \) and \( M \)-dependent transfer of spectral weight has no analogy in a non-interacting system. A similar calculation of \( \sigma_{xx}(\omega) \) by Millis et al. [15] for the case \( U_{ab} = 0 \) gives a completely incoherent optical response, in contradiction with experiment, and approaches based on the pure DE model in the cubic geometry of the perovskites [16] overestimate the quasicoherent part vis-a-vis experiment, and completely miss the mid-IR features [17].

In our picture, the extreme sensitivity of the ac response to a field of a few teslas arises via the change in the hopping, \( t_{ij}^{\sigma\sigma}(M) \), which transfers optical spectral weight over large energy scales of \( O(D) \). The observation of Simpson et al. [18] also has a natural interpretation in terms of Hubbard model like physics in the orbital sector, as shown above. In Fig. 2 (b), we also show how the COM is reduced by static JT disorder, demonstrating that the COM is related to the increased field-induced itinerance of the \( e_g \) holes via \( t_{ij}^{\sigma\sigma}(M) \).

In conclusion, we have shown how a combination of essential bandstructure aspects of the cubic perovskite structure with a DMFT treatment of strong orbital correlations in the \( e_g \) sector in the DE limit describes the optical and photoemission spectra of the metallic CMR manganites in a semiquantitative way. Additional interesting applications of our modelling to investigate field-dependent magneto-optical response across the paraferro transition are being studied, and will be reported elsewhere. Our treatment can be extended to include other broken symmetries in the spin and orbital sectors, and should be generally applicable, with suitable modifications, to other half-metallic TM-oxide ferromagnets.

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