Competition of Zener and polaron phases in doped CMR manganites

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Inspired by the strong experimental evidence for the coexistence of localized and itinerant charge carriers close to the metal-insulator transition in the ferromagnetic phase of colossal magnetoresistive manganese perovskites, for a theoretical description of the CMR transition we propose a two-phase scenario with percolative characteristics between equal-density polaron and Zener band-electron states. We find that the subtle balance between these two states with distinctly different electronic properties can be readily influenced by varying physical parameters, producing various “colossal” effects, such as the large magnetization and conductivity changes in the vicinity of the transition temperature.

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I. INTRODUCTION

The transition from a metallic ferromagnetic low-temperature phase to an insulating paramagnetic high-temperature phase observed in some hole-doped manganese oxides (such as the perovskite family \(\text{La}_{1-x}\{\text{Sr}, \text{Ca}\}_{x}\text{MnO}_3\)) is associated with an unusual dramatic change in their electronic and magnetic properties. This includes a spectacularly large negative magnetoresistance (CMR) - which might term colossal magnetoresistance (CMR) - which might have important technological applications \cite{1}. Starting with the pioneer papers of Jonker and van Santen half a century ago \cite{2}, this challenging behavior has stimulated a considerable amount of experimental and theoretical work \cite{3,4}, however, even much of the basic physics of the CMR still remains controversial.

Early studies on lanthanum manganites concentrated on the link between magnetic correlations and transport, and attributed the low-\(T\) metallic behavior to Zener’s double-exchange mechanism \cite{5,6}, which maximizes the hopping of a strongly Hund’s rule coupled Mn \(e_g\)-electron in a polarized background of the core spins (Mn \(t_{2g}\)-electrons). The quantum version of this process has been described by Kubo and Ohata \cite{7}. Although there is no much controversy about the qualitative validity of the double-exchange scenario to stabilize a ferromagnetic state, it has been argued that physics beyond double-exchange is important not only to explain the very complex phase diagram of the manganites \cite{8,9} but also the CMR transition itself \cite{10,11}. The difficulty is that magnetic scattering of itinerant charge carriers (doped holes) from enhanced fluctuations near \(T_c\) is the exclusive mechanism to drive the metal-insulator transition in the double-exchange-only models. However, even complete spin disorder does not lead to a significant reduction of the electronic bandwidth, and therefore cannot account for the observed scattering rate \cite{12,13} (cf. also the discussion in Refs. \cite{14,15}).

In view of this problem, it has been suggested that orbital \cite{16,17} and, in particular, lattice effects \cite{12,18,19} are crucial in explaining the CMR phenomenon. The argument was that the electron-lattice coupling is known to be strong in at least some members of the perovskite manganese family and tends to localize doped hole carriers as small polarons by changing the ratio of the energy gain through polaron formation to the conduction band kinetic energy \cite{20,21}. Clearly this ratio is extremely sensitive to changes of the magnetic correlations by varying magnetic field, temperature and carrier concentration \cite{22,23}. There are two types of lattice distortions which are important in manganites. First the partially filled \(e_g\) states of the Mn\(^{3+}\) ion are Jahn-Teller active, i.e., the system can gain energy from a quadrupolar symmetry elongation of the oxygen octahedra which lifts the \(e_g\) degeneracy \cite{24,25}. A second possible deformation is an isotropic shrinking of a Mn\(^{4+}\) octahedron. This “breathing”-type distortion couples to changes in the \(e_g\) charge density, i.e., is always associated with the presence of an Mn\(^{4+}\) ion. In the lightly doped region of the phase diagram, holes are pinned onto Mn\(^{4+}\) sites by locally “undoing” the cooperative Jahn-Teller distortion, i.e., forming so-called “anti Jahn-Teller polarons” \cite{30}. As pointed out in a recent paper by Billinge \textit{et al.} \cite{31}, this perspective is not appropriate, or at least misleading, in the heavily doped material, when the high-temperature polaronic state is approached from the ferromagnetic metallic (Zener) state, because there are initially almost no Jahn-Teller distorted octahedra. In this regime, both breathing-mode collapsed (Mn\(^{4+}\)) and Jahn-Teller distorted (Mn\(^{3+}\)) sites are created simultaneously when the holes are localized in passing the metal-insulator transition. To avoid any confusion, in what follows, we refer to a “polaron” as a doped charge carrier (hole) which is quasi-localized with an associated lattice distortion \cite{22}. The relevance of small polaron transport above \(T_c\) is obvious from the activated behavior of the conductivity \cite{32}. Consequently many theoretical studies focused on po-
Polaron features have been established by a variety of experiments. For example, high-temperature thermopower [10,11] and Hall mobili-ty measurements [12] confirmed the polaronic nature of charge carriers in the paramagnetic phase. More directly the existence of polarons has been demonstrated by atomic pair distribution [13], X-ray and neutron scattering studies [14–16]. Interestingly it seems that the charge carriers partly retain their polaronic character well below $T_c$, as proved, e.g., by neutron pair-distribution-function analysis [17] and very recent resistivity measurements [18]. Moreover, there have been predictions, based on XAFS data [19], that small octahedral distortions persist at low temperature, forming a nonuniform metallic state. Particularly striking in this respect is the direct relationship between the structural distortions and the magnetism of the CMR perovskites [20]. That means, even the nature of the ferromagnetic low-temperature phase is not yet completely resolved. Obviously manganese oxides, above and below $T_c$, are in the subtle regime where many different tendencies are in competition and it seems that more refined ideas are needed to explain the main properties of these materials.

Realizing that intrinsic inhomogeneities and mixed-phase characteristics exist and might play a key role in manganites, two-phase models describing the coexistence and interplay of localized and itinerant carriers have recently attracted a lot of attention (for a review see Ref. [2]). For example, phase separation scenarios involving phases with different electronic densities have been adopted to describe the mixed-phase tendencies in manganites [23,24]. These approaches are particularly meaningful in the limits of small ($x < 0.1$) and high ($x \sim 1$) hole densities, where nanometer scale coexisting clusters have been reported. In the CMR regime ($0.15 < x < 0.5$), however, several experiments reported clusters as large as micrometers in size [25,26]. Of course, such $\mu$m-sized domains, if charged, are energetically unstable because of the electro-neutrality condition enforced by long-range Coulomb repulsion. Therefore an alternative concept, where the metal insulator transition and the associated magnetoresistance behaviour is viewed as a percolation phenomenon, was analyzed theoretically [27,28], and now has a substantial experimental support by pulsed neutron [29], electron microscopy [30], Mössbauer [31] and scanning tunnel spectroscopy [32] studies.

Motivated by this situation, in the present work we propose a novel approach to the metal-insulator transition in manganites which takes into account the percolative coexistence of two “intertwined” equal-density phases: metallic double-exchange dominated and polaronic insulating. The transition is driven by a feedback effect which, at $T_c$, abruptly lowers the number of delocalized holes, i.e., conducting sites, leading to an collapse of the bandwidth of the Zener state. The physical picture behind our approach is corroborated by X-ray-absorption fine structure [30] and pair distribution [32] data, which indicate that charge localized and delocalized phases coexist close to the CMR transition [33,34]. Further support comes from zero-field muon spin relaxation and neutron spin echo measurements [35], reporting two time scales in the ferromagnetic phase of La$_{1-x}$Sr$_x$MnO$_3$, where the more rapidly relaxing component has been attributed to spins inside the overlapping metallic region and the slower Mn-ion relaxation rate was associated with polarons which occupy a diminishing volume fraction as the temperature is lowered.

**II. TWO-PHASE MODEL FOR THE CMR TRANSITION**

To set up a two-component description of the metal-insulator transition in CMR manganites, let us first characterize the two competing phases.

**A. Delocalized Zener state**

The degeneracy of $e_g$ states in (cubic) manganese oxides implies that the itinerant $e_g$ charge carriers carry an orbital degree of freedom (put in mind that the Jahn-Teller distortion lifting the $e_g$ degeneracy vanishes in the highly-doped low-temperature metallic phase). Consequently the $e_g$ electron transfer amplitudes between two Mn sites depends on the orbital orientation. Within an $\{\theta = |3x^2 - y^2|, \epsilon = |x^2 - y^2|\}$ orbital basis, the anisotropic hopping matrix elements between bonds along the $x, y,$ and $z$ directions are given by [36,37]

$$t^{x/y}_{\alpha\beta} = \frac{t}{4} \left[ \begin{array}{c} 1 \pm \sqrt{3} \\sqrt{3} \end{array} \right] \quad t^{z}_{\alpha\beta} = t \left[ \begin{array}{ccc} 1 & 0 \\ 0 & 0 & 0 \end{array} \right]$$

where $\{\alpha, \beta \in \{\theta, \epsilon\}\}$ denote orbital indices. We note that the orbital pseudospin is not a conserved quantity. The dispersion of the corresponding (noninteracting) two-band ($\zeta = \pm$) tight-binding Hamiltonian is

$$\varepsilon_{k}^{(0)} = -2t \left[ \cos k_x + \cos k_y + \cos k_z \right]$$

$$\pm \left\{ \cos^2 k_x + \cos^2 k_y + \cos^2 k_z \right\}^{1/2}$$

The band structure [33] yields the density of states (DOS) depicted in Fig. 1 (cf. also Ref. [28]). Although we employ this DOS for all the calculations presented below, we would like to emphasize that the results will not change dramatically using an (isotropic) simple cubic or even constant DOS.

$$\begin{eqnarray}$$
Here due to the Kubo-Ohata factor \[9\] the chemical potential \(\mu\) is \(\mu(T = 0)\) is located near the lower maximum of the DOS.

Starting from a Kondo lattice Hamiltonian, in the limits of strong Hund’s rule coupling and large on-site Hubbard interaction \((U \gg J_H \gg t_{x/y/z}\) which reflects the situation in the manganites), an effective transport Hamiltonian for (spinless) \(e_g\) holes can be derived \[9,16\]. Note that because this Hamiltonian acts in a projected Hilbert space without double occupied sites, in Fig. 1 the integrated DOS is normalized to one, leading to an additional prefactor \(1/2\) in the \((k, \zeta)\)-summations. Then, combining the action of Zener’s double-exchange with the percolative approach, the following effective hole band results

\[
\varepsilon_{k\zeta} = p^{(f)} \tilde{\gamma}_S [\tilde{S} \lambda] \varepsilon^{(0)}_{k\zeta}.
\]

That is, in our model the renormalization of the band energy of the Zener state is driven by two mechanisms.

At first, introducing an effective field \(\lambda = \beta g \mu_B H_{\text{eff}}\) that tends to order the ion spins in \(z\)-direction, we have the temperature- and field-dependent band narrowing due to the Kubo-Ohata factor \[3\]

\[
\gamma_S[z] = \frac{1}{2} + \frac{1}{2\pi^2} \coth\left(\frac{2\varepsilon_{k\zeta}}{k_B T}\right) \left[\coth\left(\frac{\varepsilon_{k\zeta}}{k_B T}\right) - \frac{1}{2\pi^2} \coth\left(\frac{2\varepsilon_{k\zeta}}{k_B T}\right)\right].
\]

(4)

Here \(\tilde{S} = S + 1/2\) refers to the total \(3d\) spin of an Mn\(^{3+}\) ion when an \(e_g\) electron is present, and \(S = 3/2\) is the total core spin formed by the three \(t_{2g}\) electrons. The factor \(\tilde{t}\), yielding an effective hole transfer amplitude

\[
\tilde{t} = \gamma_S [\tilde{S}\lambda] \tilde{t},
\]

was obtained averaging the transfer matrix element for a single nearest-neighbour Mn\(^{3+}\)-Mn\(^{4+}\) bond over all values and directions of the total bond spin \(S_T\) (cf. Refs. \[4,14\]).

Next, the percolative aspects of the metal-insulator transition discussed above imply that, at least just above \(x_c\) (critical concentration for the occurrence of the ferromagnetic metallic state at \(T = 0\)) or below \(T_c\), there exist insulating (polaronic) enclaves sparsely embedded in the conducting ferromagnetic (Zener) phase. Thus the ferromagnetic phase occupies a volume smaller than the sample volume, i.e. \(N^{(f)} < N\), where \(N\) is the total number of sites and \(N^{(f)}\) denotes the number of ions in the FM phase. Of course the sizes and shapes of the insulating microscopic islands are difficult to access quantitatively. For simplicity, within our effective two-fluid approach, we assume, that the hole hopping amplitude has the value \(\tilde{t}\) inside the conducting region (corresponding to the effective transport Hamiltonian \(28\) of previous work \[4\]) and zero elsewhere. Using Fourier analysis, the homogeneous component of this spatially dependent hopping amplitude, \(t = N^{(f)} \tilde{t}\), gives the renormalization of the bandwidth, whereas the long-wavelength Fourier-components cause the scattering of the carriers. Naturally the size of the ferromagnetic region, or equivalently the “probability”

\[
p^{(f)} = \frac{N^{(f)}}{N},
\]

has to be determined self-consistently (see Sec. II C). This introduces a feedback effect. Additional justification for the assumed reduction of the bandwidth proportional to the fraction of the ferromagnetic region can be obtained from the numerical calculation of the density of states outlined in the appendix for a simple tight-binding site percolation model without feedback.

B. Localized polaronic state

Let us denote the number of ions and localized electron vacancies (holes) in the polaronic phase by \(N^{(p)}\) and \(N^{(h)}\), respectively. Of course, \(N^{(p)} = N - N^{(f)}\) and \(N^{(h)} = N_h - N^{(f)}\) hold, where \(N_h\) is the total number of holes, i.e., \(x = N_h/N\). Then the energy gain due to the Jahn-Teller splitting on localized electron sites without the influence of vacancies is weakened according to \((E^{(p)} - E^{(h)}) = (x^{-1} - 1)E_1 N_h^{(p)}\) where \(E_1\) describes an effective Jahn-Teller energy in the polaronic regions. At the same time, if a doped hole is localized at a certain site, forming a small polaron, a breathing distortion may occur which lowers the energy of the unoccupied \(e_g\) level by the familiar polaron shift, \(E_p = -g^2 \omega_0\) (see, e.g., Ref. \[24\]), relative to its energy in an ideal structure. In addition, a reduction of the Jahn-Teller distortions in the neighborhood of the localized electron vacancy takes place. Both effects add up to an effective polaron binding energy \(E_2 < 0\) per hole. Neglecting the exponential small polaronic bandwidth, the polaronic phase, realized only in a fraction \(p^{(p)} = N^{(p)}/N\) of the sample, can be represented approximately by spinless fermions (holes) having

\[
\begin{align*}
FIG. 1. Density of states corresponding to the band dispersion \([3]\) \([W = 12t\) denotes the bandwidth\]. Dotted lines give the DOS of the two \(\zeta\)-subbands. At optimal doping \(x \approx 0.3\), the chemical potential \(\mu_0 \equiv \mu(T = 0)\) is located near the lower maximum of the DOS.
\end{align*}
\]
the following site-independent energy

\[ \varepsilon_p = (x^{-1} - 1) E_1 + E_2. \tag{7} \]

As follows from a fitting of the resistivity data to a small polaron model \[33\], in the region \(0.2 < x < 0.5\), the \(x\)-dependence of the parameter \(E_2\) is rather weak, and therefore has been omitted in \[16\].

C. Self-consistency equations

The basic assumption of our mixed-phase scenario is the coexistence of metallic and insulating clusters with equal hole density, i.e., in accordance with recent experimental results \[5\], \[12\], (see also Ref. \[54\]), we assume that there is no large-scale separation of \(\text{Mn}^{3+}\) and \(\text{Mn}^{4+}\) ions in the CMR doping regime. This means

\[ x = \frac{N_h}{N} = \frac{N^{(f)}_h}{N^{(f)}} = \frac{N^{(p)}_h}{N^{(p)}}, \tag{8} \]

but, of course, in general we have \(N^{(f)} \neq N^{(p)}\) and \(p^{(f)} \neq p^{(p)}\).

Denoting by \(\langle H\rangle\) the energy of the whole two-phase system within mean-field approximation and by \(S\) the corresponding entropy, \(\mathcal{F} = \langle H\rangle - TS\) is an upper bound for the free energy. Introducing the grand-canonical potentials

\[ \Omega^{(f)} = -\frac{1}{2\beta} \sum_{k,\xi=\pm} \ln \left[ 1 + e^{\beta(\mu - \xi \epsilon_k)} \right] \tag{9} \]

and

\[ \Omega^{(p)} = -\frac{N}{\beta} \ln \left[ 1 + e^{\beta(\mu - \epsilon_p)} \right] \tag{10} \]

for holes in the ferromagnetic and polaronic phases, respectively,

\[ \mathcal{F} = N_h \mu + \Omega^{(f)} + \Omega^{(p)} - TS^{(s)} \tag{11} \]

results, where

\[ S^{(s)} = -N \left\{ p^{(f)} \left[ (1 - x) \left( \ln \nu_S[\tilde{S}] - \lambda \tilde{S} B_S[\tilde{S}] \right) \right. \right. \]

\[ + x \left. \left( \ln \nu_S[\tilde{S}] - \lambda \tilde{S} B_S[\tilde{S}] \right) \right\} + p^{(p)} \left[ (1 - x) \ln \nu_S[0] + x \ln \nu_S[0] \right] \tag{12} \]

represents the mean-field ion-spin entropy, and

\[ \nu_S[z] = \sinh(z) \coth(\frac{\beta z}{2}) + \cosh(z), \tag{13} \]

\[ B_S[z] = \frac{2^{3+1} \coth(\frac{2^{3+1} z}{2}) - \frac{1}{2^{25}} \coth(\frac{z}{25})}. \tag{14} \]

If we use, instead of the model \[28\], the spin-dependent transport Hamiltonian \[29\] of our previous work \[13\], assuming that the correlations of the spin background change on a time scale large compared with the hole hopping frequency, i.e., we replace the effective hopping amplitude \(\tilde{t}\) by

\[ \tilde{t} = \frac{\left[ \tilde{S}(1 + B_S[\tilde{S}]) \right]^2}{(2S)(2S + 1)} t, \tag{15} \]

the ion-spin entropy takes the form

\[ S^{(s)} = -N \left\{ p^{(f)} \left[ \ln \nu_S[\tilde{S}] - \lambda \tilde{S} B_S[\tilde{S}] \right] + p^{(p)} \ln \nu_S[0] \right\}. \tag{16} \]

Most significantly, at given temperature \(T\) and doping level \(x\), both the ordering field for the ferromagnetic state \((\lambda)\) and the size of the Zener phase \((N^{(f)})\), or alternatively the hole fraction \(N^{(f)}_h\), have to be determined in a self-consistent way, minimizing the free energy \(\mathcal{F}\) on the hyperplane \(\mu(\lambda, N^{(f)})\) given by

\[ x = \frac{1}{2N} \sum_{k,\xi=\pm} \frac{1}{e^{\beta(\xi \epsilon_k - \mu)} + 1} + \frac{1}{e^{\beta(\epsilon_p - \mu)} + 1}. \tag{17} \]

After all, the magnetization (more exact the averaged spin \(z\)-component) can be calculated from

\[ M = (1 - x) \tilde{S} \left\{ p^{(f)} B_S[\tilde{S}] + p^{(p)} B_S[0] \right\} \]

\[ + x S \left\{ p^{(f)} B_S[\tilde{S}] + p^{(p)} B_S[0] \right\}. \tag{18} \]

III. NUMERICAL RESULTS AND DISCUSSION

The changes in magnetization as a function of temperature are shown in Fig. \[3\] for different doping levels. The efficiency of the feedback effect is quite obvious. Omitting the \(p^{(f)}\)-factor in Eq. \(\mathcal{F}\), a rather smooth variation of \(M(T)\) results and the critical temperatures for the disappearance of the Zener state, \(T_c(x)\), are too high as compared with the experimentally observed ones. Combining percolative and double-exchange mechanisms within our simple two-phase model, the magnetization exhibits a first-order transition, whereby \(T_c\) is reduced substantially. It can be expected that the inclusion of a small but finite polaron bandwidth (instead of the localized level \(\tilde{t}\)), softens the abrupt transition to some extent but in any case we will find a rather sharp drop of \(M(T)\) near \(T_c\).

Naturally the bandwidth of the Zener state, displayed in Fig. \[3\] for a doping level \(x = 0.3\), reflects the behavior of the magnetization. Without feedback we found a moderate band narrowing, whereas a radical shrinking occurs in the feedback model, which can be traced back to the collapse of the ferromagnetic metallic region at \(T_c\). The use of the spin-dependent hopping amplitude \(\tilde{t}\) leads to a modification of the Zener bandwidth (and \(\mu\)) only
for the scenario without feedback. Here, temperature-independent band edges correspond to complete spin disorder. Including the $p^{(J)}$ factor, both models (5) and (15) yield nearly identical results, which again indicates the dominance of the percolative feedback mechanism.

FIG. 2. Magnetization $M$, normalized by $M_0 = \bar{S} - x/2$, as a function of temperature $T$ at various doping levels $x = 0.175, \ldots, 0.4$. Results are shown for the models with (bold lines) and without (thin lines) feedback using parameters $E_1 = -0.125$ eV and $E_2 = -0.25$ eV, and $t = 0.3$ eV.

FIG. 3. Temperature-dependences of the Zener band (shaded region) and of the positions of the polaronic level ($\varepsilon_p$) and chemical potential ($\mu$) without (a) and with (b) feedback. Results are presented at $x = 0.3$ ($E_1 = -0.125$ eV, $E_2 = -0.25$ eV; $t = 0.3$ eV). Dashed lines display the band edges resulting from (15) instead of (5).

FIG. 4. Phase diagram of the mixed-phase Zener-polaron model with feedback. The inset shows the fraction of the Zener phase as a function of temperature.

Figure 4 presents the central result of our work, the $x$-$T$ phase diagram of the percolative two-phase model. First of all we would like to state that it is clearly beyond the scope of the present approach to account for the complete phase diagram of real manganites, which contains a great variety of charge, spin and orbital ordered states. The focus is on the doping region $0.15 < x < 0.5$, where at low temperatures a band-description seems to be appropriate. In this regime, the phase diagrams of Fig. 4, determined for two characteristic parameter sets, $(E_1 = -0.125$ eV, $E_2 = -0.25$ eV, $t = 0.3$ eV) and $(E_1 = -0.15$ eV, $E_2 = -0.175$ eV, $t = 0.3$ eV), describe the major features of the metal-insulator transition lines in $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ and $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ [65]. Furthermore, even the absolute values of the critical concentrations $x_c(T = 0) \simeq 0.17$ and of the transition temperatures $T_c$ agree surprisingly well with the experimental data.

As can be seen from the inset, the low-temperature phase contains besides the ferromagnetic metallic region finite domains of the localized polaronic phase ($p^{(J)} < 1$, and accordingly $p^{(p)} > 0$), the fraction of which increases with increasing temperature. Finally, at $T_c$, there is an abrupt spill-over of holes between the delocalized and localized phases, which drives the metal insulator transition. Since the conductivity is proportional to the number of delocalized holes $N^{(f)}$, a dramatic change at $T_c$ results. In the insulating high-temperature phase, the band description breaks down, and the transport properties are dominated by incoherent small polaron hopping processes. For doping levels $x < x_c$, the system behaves as a short-range correlated Jahn-Teller insulator.
IV. SUMMARY

Although the theoretical understanding of the CMR phenomenon is still incomplete, double-exchange, electron-phonon and orbital effects are commonly accepted as the main ingredients. Based on an in-depth analysis of important new experimental information about the inhomogeneous microscopic structure of the ferromagnetic metallic state, we come to the conclusion that in addition mixed-phase tendencies and percolative behavior play an important role in doped manganites, in particular in the vicinity of the metal-insulator transition. To simulate these effects, we have studied a simple semi-phenomenological model, describing coexisting polaronic insulating and double-exchange dominated metallic phases with equal hole density. The fraction of localized and delocalized states was determined self-consistently. Below the transition temperature $T_c$, we found polaronic inclusions embedded in a dominant macroscopic metallic phase. Our approach, which is distinct from that of the electronic phase separation concept involving states with different carrier density, provides an alternative explanation of the ferromagnetic metal to polaronic insulator transition. The abrupt change, revealed in various electrical and magnetic properties at $T_c$, was attributed to a collapse of the Zener state mainly caused by a percolative feedback mechanism. At $T = 0$ the transition is driven by doping and occurs at $x_c \simeq 0.15 - 0.18$. At finite temperatures, disorder due to intrinsic inhomogeneities and magnetic scattering act in combination to reduce the mobility of the charge carriers. The calculated values of $T_c$ agree fairly well with the experimental ones. In conclusion, we believe that a further elaboration of the proposed percolative mixed-phase scenario, e.g., towards a direct calculation of the transport properties, may help to clarify many puzzling aspects of the CMR compounds.

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APPENDIX: PERCOLATIVE PICTURE

To support the assumption that the bandwidth of the Zener state depends approximately linear on the fraction of the ferromagnetic region (see Sec. II B, Eq. (3)), let us consider a site percolation model on a finite hypercubic lattice with $64^3$ sites and periodic boundary conditions. The lattice points are occupied with probability $p$. Adjacent occupied sites will be connected by a hopping matrix element. The density of states of the resulting random tight-binding model,

$$\mathcal{H}_p = \sum_{ij} t_p (c_i^\dagger c_j^\dagger + c_j c_i)$$

$t_p \in \{0, 1\}$, is determined numerically in two different ways, using kernel polynomial and maximum entropy methods [63,64]. On the one hand, we start from a single realization $\mathcal{H}_p$ and determine 400 Chebyshev moments of $\mathcal{H}_p$, using 100 random start vectors. On the other hand, we use 10 realizations of $\mathcal{H}_p$ and compute for each realization 200 moments from 50 random start vectors. The results basically agree.

Figure 5 displays the (averaged) density of states for various values of $p$. Self-evidently, $p$ now is simply a model parameter, i.e., there is no feedback effect as considered in Sec. II B. A significant change of the spec-
trum around the site percolation threshold, $p_c \approx 0.31$, is observed. Below $p_c$, the DOS is mainly confined to the interval $[-2,2]$, like for a one-dimensional model, and mostly consists of isolated peaks with different spectral weight. This can be seen more clearly from the steplike behaviour of the integrated density of states, $N(E) = \int_{-W/2}^E \text{DOS}(E') dE'$, shown in the lower panel. The steps can be attributed to metallic “islands” (finite clusters) containing delocalized electrons. They form a cluster comparable to the system size first at $p = p_c$. Above $p_c$, $N(E)$ becomes a continuous function and the DOS is evocative of that of a simple cubic tight-binding model but with a reduced bandwidth. That is, although the percolative cluster occupies most of the bulk, making the whole system metallic, smaller conducting and insulating regions are embedded into it. These enclaves act as small scattering centers, causing the band narrowing. The inset demonstrates the almost perfect linear dependence of $W$ on $p$, in particular at large $p$, which in reality is the region of interest (cf. the inset of Fig. 2).

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