Doped manganites show many unusual features, the most striking being the colossal magnetoresistivity (CMR) in the ferromagnetic (FM) phase. In addition, the manganites have a rich phase diagram as function of band filling, temperature and chemical composition. The broad features of these phase diagrams can be understood in terms of the double exchange model (DEM), although Jahn–Teller deformations and orbital degeneracy may also play a role. A remarkable property of these compounds is the existence of inhomogeneities in the spin and charge distributions in a large range of dopings, compositions and temperatures. At band fillings where CMR effects are present, $x \sim 0.2 - 0.5$, these compounds can be broadly classified into those with a high Curie temperature and a metallic paramagnetic phase, and those with lower Curie temperatures and an insulating magnetic phase.

The DEM is a simplification of the FM Kondo lattice, where the FM coupling between core spins and conduction electrons is due to Hund’s rule. When this coupling is larger than the width of the conduction band, it is a divalent ion, and the probability of finding two carriers in neighboring sites is a function of band filling, temperature and chemical composition. The model is described in the next section, and the method of calculation is introduced in the following section. The main results are presented in section IV, and the main conclusions are discussed in section V.
in a given ion has all the \( \epsilon_g \) orbitals in the next ions available. Then, the anisotropies associated to the differences between the two inequivalent \( \epsilon_g \) orbitals should not play a major role. On the other hand, if \( x \geq 0.5 \), we expect a significant dependence of the hopping elements on the occupancy of orbitals on the nearest ions. In this regime, the equivalence of the two \( \epsilon_g \) orbitals in a cubic lattice can be broken, leading to orbital ordering \( \epsilon_g^5 \) (see, however \( \epsilon_g^6 \)). We will show that the main features of the FM-FM phase transition, for \( x \leq 0.5 \), can be understood without including orbital ordering effects. Moreover, in this doping range, anisotropic manganites show similar features \( \epsilon_g^5 \) \( \epsilon_g^6 \), which suggest the existence of a common description for the transition. We will also neglect the coupling to the lattice. As mentioned below, magnetic couplings suffice to describe a number of discontinuous transitions in the regime where CMR effects are observed. These transitions modify substantially the coupling between the conduction electrons and the magnetic excitations. Thus, they offer a simple explanation for the anomalous transport properties of these compounds. Couplings to additional modes, like optical or acoustical phonons \( \epsilon_g^5 \), and dynamical Jahn-Teller distortions \( \epsilon_g^6 \) will enhance further the tendency towards first order phase transitions discussed here.

We consider that a detailed understanding of the role of the magnetic interactions is required before adding more complexity to the model.

III. METHOD.

At finite temperatures, the thermal disorder in the orientation of the core spins induces off-diagonal disorder in the dynamics of the conduction electrons. The calculation of the partition function requires an average over core spin textures, weighted by a Boltzmann factor which depends on the energy of the conduction electrons propagating within each texture. We have simplified this calculation by replacing the distribution of spin textures by the one induced by an effective field acting on the core spins, which is optimized so as to minimize the free energy. The electronic energy includes accurately the effects of the core spin disorder on the electrons. Our calculation follows from the inequality

\[
\mathcal{F} \leq \mathcal{F}_0 + \langle \mathcal{H}_{\text{eff}} - \mathcal{H}_0 \rangle_0,
\]

where \( \mathcal{F}_0 \) is the free energy of the system with Hamiltonian \( \mathcal{H}_0 \), and the expectation value \( \langle \cdot \rangle_0 \) are calculated with the Hamiltonian \( \mathcal{H}_0 \). The mean-fields \( \{ h_i \} \) are chosen to minimize the right-hand side of (4). The calculation of the right-hand side of (4), requires the average of the density of states (see Eq.(3)) on spin configurations straightforwardly generated according to the Boltzmann weight associated to the Hamiltonian \( \mathcal{H}_0 \) and temperature \( T \). The key point is that \( g(E; \{ S \}) \) can be numerically calculated on very large lattices without further approximations using the method of moments \( \{ h_i \} \) (complemented with an standard truncation procedure \( \{ h_i \} \)).

We have extracted the spin-averaged density of states on a \( 64 \times 64 \times 64 \) lattice (for these sizes, we estimate that finite size effects are negligible). For simplicity on the analysis, we have restricted ourselves to four families of fields: uniform, \( h_i = h \), giving rise to FM ordered textures; \( h_i = (-1)^i h \), originating A-AFM order, i.e., textures that are FM within planes and AFM between planes; \( h_i = (-1)^{x_i+y_i} h \), producing C-AFM order, that is, textures that are FM within lines and AFM between lines; and staggered, \( h_i = (-1)^{x_i+y_i+z_i} h \), which originate G-AFM order, i.e., completely AFM textures. We have chosen fields of these kind since they produce the expected kinds of order, although this is not a limitation of the method. Once the spin-averaged density of states is obtained, it is straightforward to obtain the values of the mean-field that minimize the right-hand size of Eq.(4), and the corresponding value of the density of fermions. Expressing the right-hand side of Eq.(4) as a function of the magnetization (or staggered magnetizations), we obtain the Landau’s expansion of the free energy on the order parameter.

It is finally worth mentioning when our calculation and the Dynamical Mean Field Approximation \( \{ h_i \} \) \( \{ h_i \} \) are expected to yield the same results. It is clear that the key point is the calculation of the density of states in Eq.(4). For this problem of classical variables, the dynamical Mean-Field is known to yield the same density of states as the CPA approximation \( \{ h_i \} \). Under the
hypothesis of spatially uncorrelated fluctuations of the spins, which holds in any Mean-Field approximation, the CPA becomes exact on the Bethe lattice with large coordination number. However, one cannot conclude that with our calculation we would get the same results on the Bethe lattice, since one has still to specify the probability distribution for the spins to be used in the CPA calculation of the average density of states. In Refs. [22,23] the calculation is done by identifying an effective Heisenberg-like mean field, which becomes exact when the magnetization is very small. Then, the distribution of spin orientations is equivalent to the one generated by an effective magnetic field. In this limit, our ansatz should reproduce the calculations reported in [32,19], when implemented in a Bethe lattice.

In order to study first order transitions, one must consider solutions at finite magnetizations. Then, the optimal Boltzmann weights need not coincide with the effective field ansatz made here. Detailed DMFA calculations for the double exchange model [23,27], however, show that the differences between the optimal DMFA distribution and that obtained with an effective field are small throughout the entire range of magnetizations. Thus, the scheme used in this work includes the same physical processes as the DMFA, but it is also able to describe effects related to the topology of the three-dimensional lattice, like those associated to the Berry phase, which arises from the existence of closed loops. Furthermore, the present scheme allows us to study the relative stability of phases, like the A and C antiferromagnetic phases described below, which can only be defined in a cubic lattice.

IV. RESULTS.

The model, Eq.(1), contains two dimensionless parameters, the doping $x$, and the ratio $J_{AF}/t$. The range of values of $x$ is $0 \leq x \leq 1$, and the Hamiltonian has electron-hole symmetry around $x = 0.5$. The zero temperature phase diagram, shown in Fig. 1, is calculated minimizing the effective Hamiltonian at fixed chemical potential and zero temperature (we take the limit of zero temperature in Eq.(3) obtaining the grand-canonical Hamiltonian), within the four Mean-Field ansatzs previously defined. At zero $J_{AF}/t$, only the ferromagnetic phase is found, and the system is stable at all compositions. When $J_{AF}/t$ is finite, there is a value of the chemical potential for which the empty system with a perfect G-type AFM spin ordering has the same grand-canonical energy that a system with a perfect FM spin ordering and a finite value of $x$. At this value of the chemical potential the system is unstable against phase separation [2], as shown in Fig. 1. Notice that the phase-separation region can never reach $x = 0.5$, due to the hole-particle symmetry. For larger values of $J_{AF}$ a small region of A-type AFM is found for $x \sim 0.25$, and a much larger region of C-type AFM for $x$ close to half-filling. Finally, a G-type AFM-region is eventually reached by further increasing $J_{AF}/t$. However, this is not a saturated antiferromagnetic phase since the mean-field that minimizes the grand-canonical energy has a finite $h/T$ when $T$ tends to zero [34] (notice that one cannot have a continuously varying value of $x$ in a perfect AFM configuration).

![FIG. 1. Calculated phase diagram at $T = 0$. The A-AFM phase has ferromagnetic alignment within planes, and antiferromagnetic alignment between parallel planes. The C-AFM phase has ferromagnetic alignment along chains, and antiferromagnetic alignment between neighboring chains.](image)

Let us now discuss the phase diagrams at non-zero temperatures for the different values of $J_{AF}/t$ shown in Fig. 2. For $J_{AF} = 0$, we obtain a maximum transition temperature of $T = 400K$ for a width of the conduction band $W = 12t \approx 2eV$, which is consistent with a density of states of $p(E_F) = 0.85 \ eV^{-1}$ calculated in [38] for La$_{1/3}$Ca$_{2/3}$MnO$_3$ (see also [39]). Note that the bandwidth calculated in this way is probably an overestimate, as it does not include renormalization effects due to lattice vibrations [40]. There is some controversy regarding the value of $J_{AF}$. The reported value of $J_{AF}$ for the undoped compound, LaMnO$_3$, is $J_{AF} \approx 0.58 \text{meV}$, so that $J_{AF} \approx 0.005t$ [11], although calculations give higher values [12]. In the doped compounds, there is an additional contribution of order $J_{AF} \sim t^2/U_{ex}$, where $U_{ex} \approx 1-2eV$ is the level splitting induced by the intra-atomic Hund’s coupling [3]. Thus, $J_{AF} \sim 0.01t - 0.08t$, although higher values have been suggested [7].

Our results show four types of first order transitions: i) In pure DEM ($J_{AF} = 0$) the magnetic transition becomes discontinuous at sufficiently low densities, in agreement with the analysis presented in [21]. The phase coexistence region shrinks to zero and the critical temperatures vanish as $x$ goes to zero, as expected.

ii) The competition between antiferromagnetism and ferromagnetism when $J_{AF} \neq 0$ leads to a discontinuous transition which prevents the formation of canted phases, as reported in [13-17]. This transition also takes place...
at low dopings.

![Graph](image)

FIG. 2. Transition temperatures as function of electronic density and strength of the AFM couplings. The dashed lines correspond to continuous transitions. Solid thick lines are drawn for first order transitions, and the stripes correspond to phase coexistence regions. The onset of first order transitions at \( x \approx 0.5 \) is \( J_{AF}/t \approx 0.06 \).

iii) At moderate to high dopings, the FM-PM transition becomes discontinuous, if the AFM couplings are sufficiently large. The onset for first order transitions at \( x = 1/2 \) is \( J_{AF}/t \approx 0.06 \). Unlike the previous two cases, this transition takes place between phases of similar electronic density. First and second order transition lines are separated by tricritical points.

iv) In an interval of \( J_{AF}/t \), which depends on the doping level, we also find phase transitions between the PM and A-AFM and C-AFM phases, that are of second order (see Fig. 3). At low temperatures there appear FM, C-AFM, A-AFM, and G-AFM phases separated by first order transitions with its associated phase separation regions, as shown in Fig. 3.

As we see, the DEM complemented with AFM superexchange interactions between the localized spins give rise to a very rich magnetic phase diagram that contains first and second order transitions between phases with different magnetic order.

In order to set a common frame for comparison with with standard approximations [14,20], we note the free energy of the system is made up of an entropy term, due to the thermal fluctuations of the core spins, an almost temperature independent contribution from the electrons and another temperature independent term due to the direct AFM coupling between the core spins. For instance, in the PM-FM case, we can write: \( \mathcal{F} = 3J_{AF}M^2 + E_{\text{elec}}(M) - TS(M) \) where \( S(M) \) is the entropy of a spin in an effective magnetic field producing magnetization \( M \). We can expand: \( S(M) = (\frac{3}{2}M^2 + \frac{9}{20}M^4 + \frac{99}{350}M^6 + ...) \) and \( E_{\text{elec}}(M) = c_1M^2 + c_2M^4 + c_3M^6 + ... \) where \( c_1, c_2 \) and \( c_3 \) are functions of the band filling, and \( c_1 \) is always negative (\( c_1, c_2 \) and \( c_3 \) are obtained fitting the numerical results for \( E_{\text{elec}} \)). If there is a continuous transition, the critical temperature is given by \( T_C = (2|c_1| - 6J_{AF})/3 \). The transition becomes discontinuous when the quartic term in \( \mathcal{F}(M) \) is negative. This happens if \( c_2 < 0 \) and \( T < 20/9|c_2| \). Thus, if \( J_{AF} > |c_1|/3 - 10/|c_2|/9 \), and \( c_2 < 0 \), the transition becomes of first order. A tricritical point appear in the transient between first and second order transitions.

The fact that \( c_2 < 0 \) is due to the energetics of the electrons in the disordered spin background. In a fully polarized system, \( M = 1 \), the electrons propagate in a perfect lattice. If \( M = 0 \) the spins are completely disordered, and our results reduce to those reported in [14,45].

Standard approximations [14,20] to the phase diagram of the DEM use the virtual crystal approximation, in which the cubic density of states is scaled by the average value \( \langle T(S_i, S_j) \rangle \), defined in Eq. (1). This approximation suffices to describe the main features of the phase diagram when \( J_{AF} = 0 \), but overestimates the kinetic energy of the electrons moving in the disordered spin background. The effect is more pronounced near half filling, when the electronic contribution is the largest, and \( c_2 \) is positive on the virtual crystal scheme. As our calculation takes fully into account the propagation of the electrons in a disordered environment, we think that the existence of a first order PM-FM transition when \( T_C \) is suppressed is a robust feature of the model.

![Graph](image)

FIG. 3. Transition temperature as function of the value of \( J_{AF}/t \) for concentrations \( x = 1/3 \), \( x = 3/8 \), and \( x = 1/2 \). The dashed (solid) lines correspond to continuous (first order) transitions and a circle has been plotted at the tricritical PM-FM point. In the top panel we sketch experimental results from Ref. [13] where \( x = 1/3 \). The compound \((La_{1-x}Pr_x)_{5/3}Ca_{3/8}MnO_3\) studied in Ref. [3] has \( x = 3/8 \).
At zero temperature, our calculation leads to a richer phase diagram than that calculated within the Dynamical Mean Field Approximation [10]. As mentioned in the preceding section, our method coincides with this approximation when implemented in a Bethe lattice. The topology of a cubic lattice allows for the possibility of A-AFM and C-AFM phases.

We have developed an exact Monte Carlo algorithm to study the DEM. This approach is based on a Path Integral formulation that allows to simulate on lattices much larger than in an usual Hamiltonian formulation. Full details of the method will be given elsewhere [7]. The first data of the Monte Carlo computation confirm the robustness of the present results. Simulations in the parameter region depicted in Fig. 3 show a very clear evidence for a first order transition in lattices up to $12 \times 12 \times 12$ sizes. In Fig. 4 we show data on a $L = 8$ lattice at half-filling at several temperatures. Note the large region of metastability marked by the vertical lines. It is also clear that fluctuations lower the transition temperatures from their mean-field values, as it also happens in the three-dimensional Heisenberg model [18]. In addition, we find a helicoidal spin structure at sufficiently low temperatures, which replaces, partially, the A-AFM and C-AFM phases discussed earlier.

![FIG. 4. Monte Carlo results for the squared magnetization (bottom), and the $k = (2\pi/8, 0, 0)$ squared Fourier component of the magnetization (top) in $8 \times 8 \times 8$ lattices, as function of $J_{AF}/t$, for $x = 1/2$ at different temperatures.](image)

Turning again to the Mean-Field approach, let us recall that while a continuous transition is changed into a smooth crossover in an applied field, a first order transition survives until a critical field is reached. The transition takes place between two phases with finite, but different, magnetization, in a similar way to the liquid-vapor transition. The PM-FM line of first order transitions for dopings close to $x = 0.5$ ends in a critical point, $(T_c, H_c)$. For $J_{AF} = 0.08t$, the critical field varies from $H_c = 0.00075t \approx 2.2T$ at $x = 0.5$ to $H_c = 0.0002t \approx 0.6T$ at $x = 0.3$, while $T_c = T_c(x)$ is the Curie temperature at zero field, shown in Fig. 3.

![FIG. 4. Monte Carlo results for the squared magnetization (bottom), and the $k = (2\pi/8, 0, 0)$ squared Fourier component of the magnetization (top) in $8 \times 8 \times 8$ lattices, as function of $J_{AF}/t$, for $x = 1/2$ at different temperatures.](image)

V. CONCLUSIONS.

We have shown that the phase diagram of double exchange systems is richer than previously anticipated, and differs substantially from that of more conventional itinerant ferromagnets. We have described first order transitions which are either intrinsic to the double exchange mechanism, or driven by the competition between it and AFM couplings. In particular, we find that, in the doping range relevant for CMR effects, AFM interactions of reasonable magnitude change the PM-FM transition from continuous to first order. The existence of such a transition has been argued, on phenomenological grounds, in order to explain the observed data in a variety (but not all) of doped manganites in the filling range $x \sim 0.3 - 0.5$ [49,50]. The generic phase diagram that we obtain is consistent with a number of observations:

i) Materials with a high transition temperature (low AFM couplings) have a continuous PM-FM transition, with no evidence for inhomogeneities or hysteretic effects. The paramagnetic state shows metallic behavior.

ii) The PM-FM transition in materials with low transition temperature (significant AFM couplings) is discontinuous. Near $T_C$ inhomogeneities and hysteretic behavior are observed. The transport properties in the paramagnetic phase are anomalous.

iii) Substitution of a trivalent rare earth for anionic changes that do not modify the doping level) diminishes the $Mn-O-Mn$ bond angle, reducing the conduction bandwidth, $W = 12t$ [39,51]. Assuming that the AFM coupling, $J_{AF}$, does not change significantly, the ratio $J_{AF}/t$ increases; therefore, the doping level $y$ in series of the type $(RE_{1-y}RE_y)_{1-x}AE_x\MnO_3$ might be traded by $J_{AF}/t$. The top panel of Fig. 3 shows the experimental magnetic phase diagram of $(La_{1-x}Tb_{y})_{2/3}Ca_{1/3}\MnO_3$, as taken from Ref. [13]. We note the similarities with the phase diagrams of the DEM in the plane $(J_{AF}/t, T/t)$ at fixed $x$. The phases A-AFM and C-AFM at intermediate $J_{AF}/t$ could become spin glass like phases in presence of disorder.

iv) The first order PM-FM transition reported here survives in the presence of an applied field. A critical field is required to suppress it (hysteretic effects in an applied field have been reported in [52]).

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