The interplay between double exchange, super-exchange, and Lifshitz localization in doped manganites

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Considering the disorder caused in manganites by the substitution Mn → Fe or Ga, we accomplish a systematic study of doped manganites begun in previous papers. To this end, a disordered model is formulated and solved using the Variational Mean Field technique. The subtle interplay between double exchange, super-exchange, and disorder causes similar effects on the dependence of $T_C$ on the percentage of Mn substitution in the cases considered. Yet, in $\text{La}_{2/3}\text{Ca}_{1/3}\text{Mn}_1-y\text{Ga}_y\text{O}_3$ our results suggest a quantum critical point (QCP) for $y \approx 0.1 - 0.2$, associated to the localization of the electronic states of the conduction band. In the case of $\text{La}_x\text{Ca}_y\text{Mn}_1-y\text{Fe}_y\text{O}_3$ (with $x = 1/3, 3/8$) no such QCP is expected.

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I. INTRODUCTION

A continuous phase transition at zero temperature defines a Quantum Critical Point (QCP), with a diverging length-scale for quantum fluctuations and a diverging time scale for dynamics. The Renormalization Group can be extended to the study of QCP's which has become a popular concept among theoreticians, given the enhanced predictive power of the calculations. For instance, the experimental finding of a QCP has been postulated to underlie the physics of high-temperature superconductors and spin-ladders. It has been used to predict the scaling behavior for meta-magnetic quantum criticality in metals, or to study the effects of disorder in quantum spin-chains. The localization transition of non-interacting electrons in the presence of disorder is another type of continuous phase transition at zero temperature. The experimental finding of a QCP compatible with the theoretical expectations for the Anderson metal-insulator transition would be very interesting. Note that the Anderson transition is not the cause of the metal-insulator transition in $\text{La}_{1-x}\text{Ca}_x\text{Mn}_3\text{O}_4$, a well known colossal magnetoresistance (CMR) manganite.

In this paper, we want to address the behavior of typical CMR manganites when doped at the Mn sites

$$\text{La}_{1-x}\text{AE}_x\text{Mn}_{1-y}\text{TR}_y\text{O}_3,$$  \hspace{1cm} (1)

where $\text{AE} = \text{Ca}, \text{Sr}$ and $\text{TR} = \text{Fe}, \text{Al}, \text{Ga}$, in the range $0.3 < x < 0.5$. Some experimental work has already been done for this materials, but, from the present analysis we believe that several interesting features have not still been found experimentally. Specifically, we will show that, from the knowledge gained in the study of the phase diagrams of $(\text{La}_{1-x}\text{RE}_x)_{1-x}\text{Sr}_x\text{Mn}_3\text{O}_4$ and $(\text{La}_{1-x}\text{RE}_x)_{1-x}\text{Ca}_x\text{Mn}_3\text{O}_4$, where RE is a trivalent rare earth, and from our model calculations, one can conclude that:

i) When the $y = 0$ material shows a first order transition from the paramagnetic state (PM) to the ferromagnetic (FM) one, like in $\text{La}_{2/3}\text{Ca}_{1/3}\text{Mn}_3\text{O}_4$, a QCP should appear close to 15% Ga substitution ($x = 1/3$ and $y = 0.15$). Unfortunately previous experimental work did not go beyond 10% of Ga, but more experiments will be done. In this case, it is interesting that the localization transition determines the magnetic behavior and then, through the corresponding magnetic critical exponents, it may be possible to investigate the physical properties of such QCP experimentally. In this context, note that disorder added to a system with a low $T$ first order transition separating ordered phases (see Sec. and Fig. 2) produces quantum-critical like features.

ii) When the $y = 0$ material shows a second-order PM—FM phase transition, as in $\text{La}_{2/3}\text{Sr}_{1/3}\text{Mn}_3\text{O}_4$, the low temperature phase will be ferromagnetic until values of $y$ as large as $\approx 0.4$, and (if Mn is substituted with Ga) a FMmetal—FMinsulator transition at finite temperature should take place (see also Ref. 20).

The layout of the rest of this paper is as follows. In the next section, we describe the experimentally known effects of Mn substitution and we will discuss the simplest model that can account for it. We will first consider the relatively simpler case of Fe → Mn substitution, and we will then address the case of Gallium. From now on, we will not be concerned with the Al case, because this substitution causes the lattice to loose oxygen for $y \approx 0.1$, and a significant amount of oxygen vacancies are present. This phenomenon can easily be taken into account with a proper modification of the present model, but this is left for future work. In section III we present our results, while the conclusions, comments, and predic-
core spins, which we assume to be classical.

II. MODEL

A. Fe substitution

In previous works, we have considered the simplest model that describes the magnetic properties of $\text{La}_{1-x}\text{RE}_x\text{MnO}_3$ and we concluded that, for $0.3 < x < 0.5$ such model is the single orbital double exchange model (DEM) on a cubic lattice with isotropic hopping and super-exchange antiferromagnetic first-neighbors interaction:

$$\mathcal{H} = \sum_{ij} t(S_i, S_j) c_i^\dagger c_j + \sum_{(ij)} J_{AF} S_i \cdot S_j,$$

where $c_i$ corresponds to a Mn $e_g$ orbital while $S$ stands for a unit vector oriented parallel to the Mn exchange direction, which we assume to be classical. The function $t(S_i, S_j) = t \cos \theta_i \cos \theta_j + \sin \theta_i \sin \theta_j \cos(\varphi_i - \varphi_j)$ stands for the overlap of two spin $1/2$ spinors oriented along the directions defined by $S_i$ and $S_j$, whose polar and azimuthal angles are denoted by $\theta$ and $\varphi$, respectively. The hopping integral, $t$, is approximately 0.16 eV. As far as the magnetic interactions are concerned, one can trade the effects of phonons by tuning the superexchange constant $J_{AF}$. The phase diagram of the model has been extensively studied and the results are in agreement both with the experimental data of Ref. 14 and by the Hybrid Monte Carlo method. These studies have shown that the parameter $J_{AF}$ is rather constrained. On one hand, if one wants to reproduce the first-order character of the PM-FM phase transition, one should have $J_{AF}$ larger than 0.06$t$. But, on the other hand, the system is FM at low temperatures, only up to $J_{AF} \approx 0.08t$, for a hole concentration of $1/3$. The conclusion is twofold: i) $J_{AF}$ is not a very tunable parameter and, ii) $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ is a rather critical system where small perturbations can produce large changes in physical properties.

Therefore, it is a challenge to this model to give account of the experimental situation of the double series in the range $0.3 < x < 0.5$, without stretching out the values of $J_{AF}$ already determined.

Through the whole series, only the Mn $e_g$ (up) band is electronically active, with the electron hopping between Mn$^{3+}$ and Mn$^{4+}$. In the Fe case, the Fe $e_g$ (up) band is completely filled and electron hopping from Mn$^{3+}$ to Fe$^{3+}$ is forbidden. The situation is similar when TR = Fe$^{3+}$ or Ga$^{3+}$, in which cases the d-band is full. This means that the fraction of Mn$^{3+}$ with respect to the amount of Mn$^{3+}$+Mn$^{4+}$ is increas by a factor $(1-y)^{-1}$ respect to the case $y = 0$. Figure 2 of Ref. shows that, as far as the following argument is concerned, the Curie temperature, $T_C$, does not depend much on the hole density around 40% of carriers. This will allow us to extrapolate the results for TR = Fe from our previous computations. In fact, the similar ionic radii of Fe$^{3+}$ and Mn$^{3+}$ means that lattice distortion effects may be ignored. Thus, let us explore the hypothesis that the substitution of Mn for Fe affects only the antiferromagnetic interaction between the localized spins, besides the irrelevant in this case $(1-y)^{-1}$ factor in the effective number of holes. This change is due to the arising of Mn-Mn, Mn-Fe, and Fe-Fe couplings, with probability $(1-y)^2$, $2y(1-y)$ and $y^2$ respectively. Given that the core spins are $3/2$ for Mn and $5/2$ for Fe, the effective value of the super-exchange constant is

$$J_{\text{eff}} = J_{AF} \left[ (1-y)^2 + \frac{5}{3} 2y(1-y) + \frac{25}{9} y^2 \right],$$

Thus, we can easily extrapolate the results of Fig. 3 of Ref. 21 for $x = 0.3$, to the case of Fe substitution (we took $J_{AF}/t$ $\approx 0.07$), where we get $J_{\text{eff}} = 1.17 J_{AF}$ for $y = 0.12$, what implies that $T_C(y = 0.12)/T_C(y = 0) \approx 0.4$ (see Fig. 3 of Ref. 21), in complete agreement both with the experimental data of Ref. 14 as well as with the phenomenological analysis of A. Tzavelas et al.

On the other hand, when TR in (1) is Al or Ga, lattice distortion effects enhance the relevance of the fact that the Mn sites the charge is a mixture of +3 and +4. In fact, we will see, electrostatic effects yielding diagonal disorder in our model turn out to be crucial not only to understand the known experimental facts, but also to produce a new and very interesting physical situation for values of $y$ not studied in Ref. 22.

B. Ga substitution

The effects of Ga doping in the colossal magnetoresistance materials $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$, has been recently studied by preparing the series $\text{La}_{2/3}\text{Ca}_{1/3}\text{Mn}_{1-y}\text{Ga}_y\text{O}_3$ ($y = 0, 0.02, 0.05, 0.1$) and a strong decrease of $T_C$ with growing $y$ was reported. However, the doping fraction never went beyond 10%, while we believe that a QCP will appear for $y \approx 0.1-0.2$. No structural change upon doping was observed nor it is to be expected for larger Ga fraction, because it does not occur in the $\text{LaGa}_y\text{Mn}_{1-y}\text{O}_3$ series. The model used here does not take into account the orbital degeneracy in the $e_g$ band. Thus, strictly speaking it ceases to be valid when the ratio Mn$^{4+}$/Mn$^{3+}$ approaches unity, which, in this case, corresponds to $y \approx 1/3$. Nevertheless we use the results for $y$ up to 0.4 in order to describe better the change in physical properties due to the transition.

The Ga substitution disorders the system (details are given below). In order to describe specifically a disorder effect, we will need a disordered model. The standard way of constructing a disordered model is to choose first a simple model able to describe the ordered situation (i.e. the system with no Gallium), and then to mimic the inclusion of impurities in the real system by the random
modification of some terms in the Hamiltonian that reflect the microscopic effects of the impurity. As said in the previous subsection our reference ordered model, is the single orbital Double-Exchange Model (DEM) on the cubic lattice of Eq. (2).

With an ordered model in our hands, we can discuss the effects of Gallium doping. Given that the sizes of Mn and Ga are not very different, one expects small changes in the Mn–O–Mn angles, and therefore, on the values of the couplings \( t \) and \( J_{AF} \). To keep the model simple we shall assume that the parameter \( J_{AF} \) and \( t \) are not modified at all by Ga substitution. Since the series \( \text{LaGa}_3\text{Mn}_{1-y}\text{O}_3 \) exist for all \( y \), we expect Ga to be in a \( \text{Ga}^{3+} \) oxidation state, which is a filled-shell configuration. This means that the corresponding \( \epsilon_n \) orbital will not be available for a hole in a neighboring Mn to hop, which reduces the Double-Exchange (DE) ferromagnetic interaction. We thus encounter a quantum-percolation problem (see e.g. Ref. 23). However, quantum-percolation studies have shown that the quantum threshold is not far from the classical one (that would be reached at 69% Ga fraction!), and this effect is not expected to be crucial. Moreover, given the filled-shell configuration of Ga, the \( \text{Mn}^{3+} \) core spin \( S_i \) will disappear from Ga-sites, which reduces also the AFM interaction that competes with the DE ferromagnetic one. In the case of Ga, the effective AF coupling of Eq. (3) is \( J_{eff} = (1 - y)^2 J_{AF} \) which, for instance, for \( y = 0.12 \) turns out to be 0.77 \( J_{AF} \). This is really a huge reduction (see Fig. 3 of Ref. 21 and Fig. 3 of Ref. 24), and it is not obvious what the effect of Ga substitution on the Curie temperature would be. Fortunately, the effects of Ga substitution described so far can be straightforwardly studied with the VMF method and it turns out that the reduction on the antiferromagnetic interaction has a stronger effect than the reduction of the DE mechanism, and \( T_C \) rises in the model when the Ga fraction is increased. Since this is in plain contradiction with experiments, it is clear that our microscopic description of the Ga substitution is still incomplete. This suggests that an on-site electrostatic perturbation is induced by Ga ions in Eq. (3). This electrostatic term can be justified by the fact that the average charge on Ga sites is always +3 while on Mn sites it is a mixture of +3 and +4. Therefore holes cannot hop onto Ga sites, but they are attracted to them.

We expect this phenomenon to be more relevant in the case of Ga than in the case of Fe. In fact, if in a cubic lattice of Mn ions we substitute a fraction \( y \) of Mn by TR randomly, the electrostatic potential that appears in the Mn sites is proportional to the number of TR\(^{3+} \) that are neighbors of the considered Mn ion. Let us call \( \epsilon \) the electrostatic potential felt by a hole at a Mn site with only a neighboring TR. In the Fe case, \( \epsilon \) is the same on each Mn site because lattice distortion effects can be ignored, ought to the similar ionic radii of Fe\(^{3+} \) and Mn\(^{3+} \). However, in the Ga case (and even more in the Al case), the ionic radii difference implies that to the randomness in the distribution is added the randomness originated by the lattice distortion which, among other phenomena, makes the potential randomly dependent on the Mn position.

To avoid an overly complicated model, we incorporate the second disorder to the first one (of Lifshitz type) and we work with an effective \( \epsilon \), the same for all Mn sites. Consequently, the \( \epsilon \) is larger for Ga and Al than for Fe; this is precisely what the experimental data required since, as we have said, without a localization mechanism that destroys the double exchange, \( T_C \) would rise with \( y \).

As we will see, a moderate disorder (from our analysis, \( \epsilon < 2t \)) modifies only slightly the decrease of \( T_C \) with \( y \). For this reason we have neglected it in the case of Fe.

In summary, our model for \( \text{La}_{2/3}\text{Ca}_{1/3}\text{Mn}_{1-y}\text{Ga}_y\text{O}_3 \) is as follows: every Ga atom modifies the Hamiltonian in the following ways:

1. Holes cannot hop onto Ga. Therefore the average number of one third of a hole per unit cell, actually means that the fraction of \( \text{Mn}^{4+} \) is increased with respect to the case \( y = 0 \) by a factor \( (1 - y)^{-1} \).

2. The core spins vanish at the Ga sites.

3. A electrostatic potential appears on the Mn sites, proportional to the number of \( \text{Ga}^{3+} \) that are nearest-neighbors of the considered Mn site.

### III. RESULTS

We have studied our disordered model using the VMF technique which is approximate for the spins, but treat charge carriers on each spin texture exactly (up to controlled numerical errors). The Hybrid Monte Carlo study has shown that the VMF overestimates the critical temperature by a 30%, which is the same overestimation factor found in classical statistical mechanics three-dimensional models with short range interactions. In the particular case of a disordered model, the VMF technique has the important merit of allowing the study of very large clusters (96 × 96 × 96 in this work). The self-averaging nature of the electronic density of states makes thus unnecessary to average our results over disorder realizations, which instead would be mandatory in a Monte Carlo study where the study of a single 16 × 16 × 16 cluster is at the very limit of present day computers and algorithms. In order to make the study as unbiased as possible, we shall consider as VMF ansatz all the spin orderings that were found in the Monte Carlo study of model in the absence of disorder, namely FM, G-AFM, A-AFM, C-AFM, skyrmion (SK), flux, twisted and island phases.

To gain some insight on the physics of our model, it is useful to consider (see Fig. 1) the hole density of states (DOS) for the fully FM spin-polarized system with a 20% Ga substitution, upon variation of the electrostatic parameter \( \epsilon \). For \( \epsilon = 0 \), we have the usual DOS of the cubic lattice, slightly contracted and smoothed by the
percolative disorder (holes cannot jump onto a Ga). We have a sharp contribution at midband, due to the localized states produced by the rare configurations of Mn sites fully surrounded by Ga sites (thereafter this very rare configurations were taken out by hand). If the electrostatic potential at a given site is large enough (in a Mn site with \( k \) neighboring Ga, it is \( k\epsilon \)), a hole state of energy \(-k\epsilon\) will get localized in this Mn site, as the reader can check in Fig. 1. This effect can be investigated within the small defect-density expansion, where \( \epsilon \) is allowed to be arbitrarily large. It turns out that a localized state will form if \( k\epsilon \) is larger than the edge of the \( \epsilon = 0 \) DOS. Therefore, Fig. 1 tells us that for \( \epsilon = 2t \), a Mn site should have at least three neighboring Ga to trap a hole on it, while for \( \epsilon = 3t \) it is enough to have two neighboring Ga (Mn sites with two neighboring Ga sites are deceivingly common, even for modest values of \( y \), because of the large combinatorial factor). The electrostatic potential tends also to localize the electronic states even near to the edges of the main part of the DOS, depicted in Fig. 1 (for \( y = 0.2 \) the Fermi level is very close to the edge of the main part of the DOS). The calculations show that the double exchange interaction is strongly suppressed when holes occupy these localized states, leading to a decrease in the value of the \( T_C \). We find that the drop in \( T_C \) upon Ga doping is best reproduced using \( \epsilon \sim 3t \), and, considering our earlier results for manganites with a perfect Mn lattice, we take \( J_{AF} \sim 0.06t-0.07t \). On the other hand, for \( \epsilon < 2t \) disorder is not the main agent of the eventual decrease of \( T_C \) as \( y \) increases (as happens in the case TR = Fe).

In Fig. 2 we show the phase diagram of our model, for four values of \( J_{AF} \), namely \( J_{AF} = 0.05t \), which is a reasonable value for La\(_{2/3}\)Sr\(_{1/3}\)Mn\(_{1-y}\)Ga\(_y\)O\(_3\), at least for \( y = 0.2 \) \( J_{AF} = 0.06t \) that marks the change from second to first order character of the PM-FM transition, and \( J_{AF} = 0.064t-0.07t \) which we believe is an appropriate range for La\(_{2/3}\)Ca\(_{1/3}\)Mn\(_{1-y}\)Ga\(_y\)O\(_3\). There are several points to be noted. First, see how at \( y = 0 \), the different values of \( J_{AF} \) are enough to explain the 30% differences in \( T_C \) between La\(_{2/3}\)Sr\(_{1/3}\)MnO\(_3\) and La\(_{2/3}\)Ca\(_{1/3}\)MnO\(_3\). As we see, for La\(_{2/3}\)Sr\(_{1/3}\)Mn\(_{1-y}\)Ga\(_y\)O\(_3\), \( J_{AF} \sim 0.05t \), we have a more or less monotonous decreases of the \( T_C \), the ground-state being FM in all the interesting \( y \) range. On the other hand, the scenario is completely different for La\(_{2/3}\)Ca\(_{1/3}\)Mn\(_{1-y}\)Ga\(_y\)O\(_3\) (\( J_{AF} = 0.064t \) and 0.07t). In this case, the FM ordering disappears for \( y_t \approx 0.1-0.2 \), where the PM phase is stable until the lowest temperatures. For larger values of \( y \) a very complicated situation is reached with different, almost degenerated phases. This region might look somehow glassy experimentally.
The ordering temperature in the large $y$ region is about one order of magnitude smaller than for $y = 0$. More remarkably, for $y$ larger than $y_c$, the spins are not completely polarized even at zero temperature (technically, this means that the mean-field at low temperature is $\alpha T$, where $\alpha$ is a large but not infinite constant[2]), and, also at zero temperature, the magnetization in the FM phase decrease upon approaching $y_c$ although it does not get below 80% polarization. This effect will be amplified by the neglected quantum fluctuations of the spins, especially at very low temperatures. Thus, given the wedge of PM phase that reaches near zero temperature at $y_c$ and our underestimation of quantum fluctuations, it seems fairly plausible that a quantum critical point will be found in La$_{2/3}$Ca$_{1/3}$Mn$_{1-y}$Ga$_y$O$_3$, for $y = 0.1-0.2$. In order to emphasize how crucial the electrostatic attraction of holes to the Ga sites is, let us show in Fig. 3 (top), the equivalent of Fig. 3 (bottom), for $\epsilon = 2t$. In this case, the system is ferromagnetic in all the explored range $0 \leq y \leq 0.4$, in agreement with the scarcity of localized states shown in Fig. 3.

IV. REMARKS AND PREDICTIONS

In this work we have accomplished a systematic study of doped manganites begun in previous papers and formulated a microscopically motivated disordered model to study the effects of Ga substitution in La$_{2/3}$Ca$_{1/3}$Mn$_{1-y}$Ga$_y$O$_3$. We have studied this model using the VMF technique in $96 \times 96 \times 96$ clusters. A result of this work is the prediction that a QCP is expected to appear for $y_c \approx 0.1-0.2$, which is induced by Anderson localization of the electronic states below the Fermi level that suppresses the double exchange mechanism. Thus, we expect localization effects in the transport properties at low temperatures, where phonons are static (i.e. polarons could exist[2], but they would play the same role of our $\epsilon$ diagonal static disorder term). For larger values of $y$, a different phase is reached which is difficult to analyze because many different phases are almost degenerate. Nevertheless one can conclude safely that the ordering temperature for $0.3 < y < 0.4$ will be in the 20 to 40 K range. It is worth noting that the main features of the predicted phase-diagram are in quantitative agreement with preliminary experimental work[6].

A completely different behavior is expected for La$_{2/3}$Sr$_{1/3}$Mn$_{1-y}$Ga$_y$O$_3$. In such case the localization phenomenon that can be inferred from Fig. 3 for $\epsilon = 3t$, besides to the results of Fig. 3 corresponding to $J_{AF} = 0.05t$, strongly suggest that the fact that holes are localized does not necessarily imply that the long range magnetism disappears; in fact, to maintain it, it is enough that effective ferromagnetic interactions between close spins remain. Hence, for La$_{2/3}$Sr$_{1/3}$Mn$_{1-y}$Ga$_y$O$_3$ we expect a FM-metal-FM-Insulator transition at low temperatures (see also Ref. 20).

In the Ga case, it is worthwhile to remark that in our model, which considers spins coupled to charge carriers, the localization transition determines the magnetic critical exponents too. Experimentally, it is very promising that one be able to study a localization transition through a magnetic order parameter. If there is really a QCP for $y \approx 0.1-0.2$ there would be presumably only one characteristic length in the system, which would set the scale both of the magnetic and transport phenomena.

It is therefore interesting the theoretical and experimental determination of the magnetic critical exponents associated to the magnetic transition at very low $T$ and its universality class.

Finally, it is worth noting that the model used in this work includes the simplest combination of interactions compatible with the experimental data for the manganites, in the absence of Ga doping. The confirmation of the predictions proposed here for Ga doped materials can illustrate the usefulness of simple models in studying the phase diagram of these compounds.

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