Disorder-induced orbital ordering in doped manganites

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We study the effect of quenched disorder on the ordering of orbital and magnetic degrees of freedom in a two-dimensional, two-band double-exchange model for $e_g$ electrons coupled to Jahn-Teller distortions. Using a real-space Monte Carlo method, we find that disorder can induce a short-range ordering of the orbital degrees of freedom near 30% hole doping. The most striking consequence of this short range ordering is a strong increase in the low temperature resistivity. The real-space approach allows to analyze the spatial patterns of the charge, orbital, and magnetic degrees of freedom, and the correlations among them. The magnetism is inhomogeneous on the nanoscale in the short-range orbitally ordered state.

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I. INTRODUCTION

Hole-doped perovskite manganites RE$_{1-x}$AE$_x$MnO$_3$ (RE=rare earth, AE=alkaline earth) have attracted great attention from the condensed matter community over the last decade [1]. While the initial surge of research activities on these materials was triggered by the discovery of the colossal magnetoresistance (CMR) effect, a rich variety of phases and phase transitions was subsequently uncovered [2, 3]. It is now widely accepted that the interplay among charge, spin, orbital and lattice degrees of freedom is the underlying cause of the complexity and richness of the physical phenomena observed in manganites. Recent efforts from both experiment and theory have highlighted the significance of quenched disorder in these materials [1, 2, 3, 4]. Therefore, analyzing the effects of disorder in manganites has become an active area of research [3, 4, 5, 6].

Disorder is generally viewed as an agent for suppressing the ordering tendencies of the microscopic degrees of freedom. Experiments on the half-doped ($x = 0.5$) manganites show that quenched disorder indeed spoils the long-range ordering of the charge, orbital, and spin variables leading, in some cases, to a short-range ordering of these microscopic degrees of freedom [5, 6]. The opposite effect, however, is observed in manganites near 30% hole doping, where an ordering of the orbital degrees of freedom is induced by the presence of quenched disorder [7].

In manganites, the average $r_A$ and the variance $\sigma^2$ of the A-site ionic radii are known to control the single-particle bandwidth and the magnitude of quenched disorder, respectively [12]. Samples with constant $r_A$ and varying $\sigma^2$ were used in the experiments of Ref. [7] with a combination of La, Pr, Nd, Sm, and Ca, Sr, Ba at the A-site, while keeping $x = 0.3$. An increase in the low-temperature resistivity by four orders of magnitude was attributed to the onset of orbital ordering, which was also evidenced from the structural changes analyzed via powder x-ray diffraction. Magnetism is affected strongly with a reduction in both, the Curie temperature $T_C$ and the saturation value of the magnetization. This doping regime is also believed to be magnetically inhomogeneous, as independently inferred from NMR and neutron scattering experiments [13, 14].

Disorder has been included previously in models for manganites to study its influence on the long-range ordered phases, especially near a first-order phase boundary or in the vicinity of phase separation [9, 15, 16]. The idea that quenched disorder may lead to a partial ordering of the orbital degrees of freedom in manganite models has so far remained unexplored.

In this paper, we study a two-band double-exchange model with quenched disorder using a real-space Monte Carlo method. Disorder is modelled via random on-site energies selected from a given distribution. We consider two different types of distributions, described in detail in the next section. Here and below we refer to these distributions as (i) binary disorder and (ii) random scatterers. While the binary disorder has no significant effect on the orbital degrees of freedom, random scatterers lead to orbitally ordered (OO) regions and a sharp increase in the low-temperature resistivity is found as observed in the experiment [7]. The magnetic structure is inhomogeneous in a restricted doping regime as observed in the NMR and the neutron scattering experiments. Within clusters, staggered orbital ordering is accompanied by ferromagnetism, thus providing an example of Goodenough-Kanamori rules in an inhomogeneous system [18, 19].

II. MODEL AND METHOD

We consider a two-band model for itinerant $e_g$ electrons on a square lattice. The electrons are coupled to Jahn-Teller (JT) lattice distortions, $t_{2g}$-derived $S = 3/2$
core spins and quenched disorder as described by the Hamiltonian:

\[ H = \sum_{\langle ij \rangle \sigma} t^{ij}_{\alpha \beta} \gamma^{\dagger} \gamma_{j \beta} + \sum_i \epsilon_i \sigma_i + J_s \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \]
\[ - J_H \sum_i \mathbf{S}_i, \sigma_i - \lambda \sum_i \mathbf{Q}_i, \tau_i + \frac{K}{2} \sum_i \mathbf{Q}_i^2. \]  

(1)

Here, \( c \) and \( c^\dagger \) are annihilation and creation operators for \( e_g \) electrons, \( \alpha = \uparrow, \downarrow \) is the spin index and \( \alpha, \beta \) are summed over the two Mn-\( e_g \) orbitals \( d_{x^2-y^2} \) and \( d_{3z^2-r^2} \), which are labelled \((a)\) and \((b)\) in what follows. \( t^{ij}_{\alpha \beta} \) denote the hopping amplitudes between \( e_g \) orbitals on nearest-neighbor sites and have the cubic perovskite specific form: \( t^{x}_{aa} = t^{y}_{bb} \equiv t, \quad t^{y}_{ab} = t^{x}_{ba} = t/3, \quad t^{x}_{ab} = t^{y}_{ba} = -t/\sqrt{3}, \quad t^{y}_{ab} = t^{x}_{ba} = t/\sqrt{3}, \) where \( x \) and \( y \) mark the spatial directions \[20\]. The \( e_g \)-electron spin is locally coupled to the \( t_{2g} \) spin \( \mathbf{S}_i \) via the Hund’s rule coupling \( J_H \). The \( e_g \)-electron spin is given by \( \sigma_{\mu} = \sum_{\alpha \beta} \gamma_{i \alpha \sigma} \Gamma_{\mu}^{\alpha \beta} \gamma^\dagger_{i \beta \sigma}, \) where \( \Gamma_{\mu} \) are the Pauli matrices. \( J_s \) is the strength of the superexchange coupling between neighboring \( t_{2g} \) spins. \( \lambda \) denotes the strength of the JT coupling between the distortion \( \mathbf{Q}_i = (Q_{ix}, Q_{iz}) \) and the orbital pseudospin \( \tau^{\mu}_{i \alpha} = \sum_{\alpha \beta} \gamma_{i \alpha \sigma} \Gamma_{\mu}^{\alpha \beta} \gamma^\dagger_{i \beta \sigma} \). \( K \) is a measure of the lattice stiffness, and we set \( t = 1 \) as our reference energy scale.

The following two forms of on-site disorder modelling are used: (i) binary disorder: \( \epsilon_i \) takes equally probable values \( \pm \Delta \), (ii) random scatterers: a fraction \( x \) of the sites are taken to have \( \epsilon_i = D \), while for the other sites \( \epsilon_i = 0 \). Although the first choice of disorder is the simplest from the model point of view, the second appears more realistic. In real materials a fraction \( x \) of the rare-earth ions is replaced by alkaline-earth ions at random locations. Therefore, it is likely that the disorder arising as a consequence of this substitution is connected to the amount of doping. This situation is modelled by placing repulsive potentials on a fraction \( x \) of the sites, which are randomly selected. A typical measure of the strength of a disorder distribution is its variance. For the binary distribution the variance is \( \Delta \), while for the finite density \( x \) of scatterers with potential strength \( D \) it is \( D \sqrt{x(1-x)} \). These two models for disorder were previously employed in a study of half-doped manganites \[11\]. The JT distortions and the \( t_{2g} \) derived core spins are treated as classical variables, and we set \( |S_i| = 1 \). Guided by earlier estimates for the JT coupling strength in manganites, we fix \( \lambda = 1.5 \) \[21\], and explore the variation in the parameters \( \Delta, D, J_s \).

We further adopt the simplifying limit \( J_H >> t \), which is justified and frequently used in the context of manganites \[4\] \[20\], \[22\]. In this limit the electronic spin at site \( i \) is tied to the orientation of the core spin \( \mathbf{S}_i \). Transforming the fermionic operators to this local spin reference frame leads to the following effectively ‘spinless’ model for the \( e_g \) electrons:

\[ H = \sum_{\langle ij \rangle} \tilde{t}^{ij}_{\alpha \beta} \gamma^{\dagger} \gamma_{j \beta} + \sum_i \epsilon_i n_i + J_s \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \]
\[- \lambda \sum_i \mathbf{Q}_i, \tau_i + \frac{K}{2} \sum_i \mathbf{Q}_i^2. \]  

(2)

The new hopping amplitudes \( \tilde{t}^{ij}_{\alpha \beta} \) have an additional dependence on the core-spin configurations and are given by:

\[ \tilde{t}^{ij}_{\alpha \beta} = \cos \frac{\theta_i}{2} \cos \frac{\theta_j}{2} + \sin \frac{\theta_i}{2} \sin \frac{\theta_j}{2} e^{-i(\phi_i - \phi_j)}. \]  

(3)

Here, \( \theta_i \) and \( \phi_i \) denote polar and azimuthal angles for the spin \( \mathbf{S}_i \). From now on the operator \( c_{i \alpha} (c^\dagger_{i \alpha}) \) is associated with annihilating (creating) an electron at site \( i \) in the orbital \( \alpha \) with spin parallel to \( \mathbf{S}_i \).

The model given by Eq. (2) is bilinear in the electronic operators and does not encounter the problem of an exponentially growing Hilbert space, since all many-particle states can be constructed from Slater determinants of the single-particle states. The difficulty, however, arises from the large phase space in the classical variables \( \mathbf{Q} \) and \( \mathbf{S} \). Exact diagonalization based Monte Carlo (ED-MC) is a numerically exact method to treat such problems, and has been used extensively in the past \[6\] \[23\] \[24\]. The classical variables are sampled by the Metropolis algorithm, which requires the exact eigenenergy spectrum. Therefore iterative ED of the Hamiltonian is needed, which leads to \( N^4 \) scaling of the required cpu time, \( N \) is the number of lattice sites. The \( N^3 \) scaling makes this method very restrictive in terms of the achievable lattice sizes, with the typical size in previous studies being \( \sim 100 \) sites. Since a study of larger lattices is essential for analyzing the nature of inhomogeneities in manganite models, several attempts have been made to devise accurate approximate schemes \[23\] \[24\] \[25\]. In the present study we employ the travelling cluster approximation (TCA) \[25\], which indeed has been very successful in analyzing similar models in the recent past \[10\] \[11\] \[20\].

III. RESULTS AND DISCUSSION

We begin with results for bulk quantities describing the ordering of the magnetic and the lattice degrees of freedom. We focus on the 30\% hole-doped system \((x = 0.3)\) for a close correspondence to the experiments in Ref. \[7\]. Fig. 1(a) shows the effect of binary disorder on the temperature dependence of the magnetization \( m \), defined via \( m^2 = \langle (N^{-1} \sum_i \mathbf{S}_i)^2 \rangle_{av} \). Here and below \((...)_{av}\) denotes the average over thermal equilibrium configurations and additionally over realizations of quenched disorder. Results for disordered systems are averaged over \( 4 - 6 \) realizations of disorder. Clearly, the magnetism is not affected much by the presence of weak binary disorder.
This is in agreement with previous studies, which find that the reduction in $T_C$ is proportional to $\Delta^2$ for weak disorder \cite{27, 28, 29}.

Fig. 1(b) shows the temperature dependence of the $q_0 = (\pi, \pi)$ component of the lattice structure factor $D_q(q_0) = N^{-2} \sum_{ij} <Q_i \cdot Q_j> e^{-i q_0 \cdot (r_i-r_j)}$. $D_q(q_0)$ is a measure for the staggered distortion order in the system. The lattice ordering leads to orbital ordering via the JT coupling. An increase with $\Delta$ in the low-temperature value of $D_q(q_0)$ suggests the appearance of orbital order. However, this effect is too weak to explain the experimental resistivity data \cite{3}. Moreover, the increase at low $T$ in $D_q(q_0)$ is not monotonic, which becomes clear by comparing the results for $\Delta = 0.4, 0.8$ and $1.0$ in Fig. 1(b).

Now we explore the results for the disorder arising from random scatterers of strength $D$. Since the disorder originates from the replacement of RE$^{3+}$ by AE$^{2+}$ ions, the density of random scatterers is kept equal to the doping concentration $x$. $m(T)$, shown in Fig. 1(c), is affected strongly upon increasing $D$, with a decrease in the saturation value of the magnetization pointing towards a magnetically inhomogeneous groundstate. More importantly, a monotonic increase with $D$ is observed in the low-temperature values of $D_q(q_0)$ (see Fig. 1(d)). The rise in $D_q(q_0)$ clearly indicates the emergence of orbital ordering in the system, with the area and/or strength of the ordered regions increasing with increasing $D$.

It is expected that these orbital ordering correlations are reflected in the transport properties. We therefore compute the dc resistivity $\rho$ approximated by the inverse of low-frequency optical conductivity, which is calculated by using exact eigenstates and -energies in the Kubo-Greenwood formula \cite{30}. Fig. 2(a)-(b) show $\rho$ as a function of temperature for the two disorder models described above. The low-temperature resistivity increases upon increasing the binary disorder strength $\Delta$ (see Fig. 1(a)). For small values of $\Delta$ the resistivity curves appear parallel to each other below $T \sim 0.1$. The resistivity therefore follows Mathiessen’s rule, i.e. $\rho(T)$ for the disordered system is obtained from $\rho(T)$ for the clean system by simply adding a constant contribution arising from the scattering off the disorder potential. $d\rho/dT$ remains positive at low temperature indicating a metallic behavior. This oversimplified description, however, does not take into account the disorder induced changes in the orbital ordering correlations and the related changes in the density of states (discussed below).

Random scatterers lead to a drastically different behavior. The low-temperature rise in $\rho(T)$ covers several orders of magnitude (see Fig. 2(b)). The negative sign of $d\rho/dT$ for $D > 1$ signals an insulating behavior. Upon increasing the disorder strength $D$ we therefore observe a metal to insulator transition. For $x = 0.3$ both disorder models have the same variance, if $\Delta \sim 0.46 D$ holds. Comparing, therefore, the results for $D = 2$ and $\Delta = 1$, we have to conclude that the drastic rise in the resis-
tivity for random scatterers cannot be attributed to the
strength of the disorder potential. In fact, a large increase
in the low-$T$ resistivity was one of the experimental in-
dications for the onset of disorder-induced orbital ordering

Figs. 2(c)-(d) highlight the difference between the
densities of states (DOS) for the two choices of dis-
order modelling. The DOS is defined as $N(\omega) =
\langle N^{-1} \sum_i \delta(\omega - E_i) \rangle_{av}$, where $E_i$
denotes the eigenvalues of the Hamiltonian. Here we approxi-
mate the $\delta$-function by a Lorentzian with width $\gamma = 0.04$:

$$\delta(\omega - E_i) \simeq \frac{\gamma/\pi}{\gamma^2 + (\omega - E_i)^2}. \quad (4)$$

The DOS for the clean system has a pseudogap structure
near the chemical potential. For binary disorder, the pseudogap
slowly fills up with increasing $\Delta$. In con-
trast, it deepens upon adding random scattering centers
and even leads to a clean gap for $D \geq 3$. This opposite
behavior is partly responsible for the drastically different
low-temperature resistivity discussed above. The three-
peak structure for large values of $D$ in Fig. 2(d) can be
understood as follows: A fraction $2x$ of electronic states
split off and form a narrow impurity band centered at an
energy $D$ above the Fermi level of the undoped system.
The lower band now contains a fraction $2(1-x)$ of the
states with the Fermi level located in the middle of the
band. This leads to a situation similar to the undoped
system, and an energy gap originating from staggered
orbital ordering opens at the Fermi level.

To gain further insight into the nature of the states
in the presence of the two types of disorder, we plot the
distribution functions for the lattice variables in Fig. 3.
Panel (a) shows the distribution of the nearest neighbor
lattice correlations, $C_Q(i) = \langle 1/4 \sum_\delta Q_{i+\delta} \cdot Q_{i+\delta} \rangle$ for
binary disorder; here $\delta$ denotes the four nearest neighbor
sites of site $i$. A negative value of $C_Q(i)$ indicates an
antiferro pattern of JT distortions, and hence a pattern of
staggered orbital ordering. The distribution function for
$C_Q$ is defined as $P(C_Q) = \langle N^{-1} \sum_i \delta(C_Q - C_Q(i)) \rangle_{av}$; the
$\delta$-function is again approximated by a Lorentzian with
width $\sim 0.04$. A peak in $P(C_Q)$ centered near $C_Q = 0.8$
for $\Delta = 0$ indicates that the clean system has weak ferro-
distortive/ferro-orbital correlations. Tails going down to
$C_Q \sim -1.4$ arise in the distribution function upon in-
cluding binary disorder.

The distribution function $P(C_Q)$ for random scatterers
looks qualitatively different. We recall that the
strengths of the two types of disorder are related via
$\Delta \sim 0.46D$. The low-temperature distributions $P(C_Q)$
are plotted in Fig. 3(b) for random scatterers. A qualita-
tive change in the shape of the distribution function
occurs for $D = 2$, where a second peak centered around
$C_Q \sim -1.2$ emerges. This is a direct indication that a sig-
nificant fraction of the system becomes orbitally ordered.
This correlates perfectly with the strong rise in $DQ(q_0)$
at low temperatures (see Fig. 1(b)), and the anomalous
increase in the resistivity (see Fig. 2(b)).

A real-space picture for the emergence of orbital or-
dering is presented in Fig. 4, which displays the disorder
potential $\epsilon_i$, the electronic density $n_i$, and the lattice cor-
lations $C_Q(i)$. The top row for binary disorder shows
that the charge density closely follows the disorder po-
tential. The local lattice correlations are centered around
$C_Q = 0$, which is also evident from the peak in the dis-
tribution $P(C_Q)$ shown in Fig. 3(a). The bottom row in
Fig. 4 shows the corresponding results for the disor-
der potential arising from random scatterers. Since the
doping concentration in this case coincides with the con-
centration of the scatterers, the holes are trapped at the
impurity sites. This leaves the surrounding effectively

FIG. 3: (Color online) Low-temperature distribution func-
tions generated from the Monte Carlo data for the nearest-
neighbor correlations $C_Q$ of the lattice distortions for (a) bi-
ary disorder and (b) for random scatterers. The curves for
different $\Delta$ are off-set along the $y$-axis for clarity. $C_Q$ is posi-
tive (negative) for ferro- (antiferro)-distortive patterns of the
lattice variables.

FIG. 4: (Color online) Real-space patterns of the disorder
potential $\epsilon(i)$, charge density $n(i)$, and lattice correlations
$C_Q(i)$. Top row: binary disorder with $\Delta = 1$, bottom row:
random scatterers with $D = 2$. The patterns in both cases
are shown on a $24 \times 24$ lattice for a single disorder realization
at $T = 0.01$ and $x = 0.3$. 

$\sum_1^4$
undoped and thereby induces orbital ordering. This is apparent from the spread of the dark-blue regions and their cross correlation with the charge density distribution in the bottom row of Fig. 4. Such a picture with orbitally ordered regions coexisting with orbitally disordered patches describes perfectly the double peak structure of the distribution function in Fig. 3(b).

Although we are primarily interested in the experimentally relevant case \( x = 0.3 \), it is useful to see how the real-space patterns evolve as one moves from low to high hole densities. The undoped system is an orbitally ordered insulator, which turns into an orbitally disordered metal upon doping \[^{31}\] . We show real-space patterns at three different doping concentrations in Fig. 5. The density of random scatterers is kept equal to the doping fraction \( x \). The charge density distribution is largely controlled by the disorder distribution. At low doping, disconnected orbitally disordered regions are essentially tied to the trapped holes. With increasing \( x \) the orbitally disordered regions begin to connect in one-dimensional snake-like patterns. By further increasing the doping and the concentration of scattering centers the orbitally disordered regions grow. The phenomenon of disorder-induced orbital ordering is likely to be present only in a narrow doping range near and above 0.25, because for \( x < 0.25 \), the system is orbitally ordered even in the clean limit.

It is worthwhile to point to a similarity between the effects of disorder in the present study and in a model analysis for \( d \)-wave superconductors with non-magnetic impurities. In Ref. \[^{32}\] it was found that the impurities nucleate antiferromagnetism in their near vicinity. Upon increasing the impurity concentration static antiferromagnetism is observed. There seems to be a perfect analogy between the two situations, if one interchanges antiferromagnetism by orbital-ordering; both are ordering phenomena with the staggered ordering wavevector \((\pi, \pi)\). The \((\pi, \pi)\) ordering phenomena is partially triggered by the charge inhomogeneities in both cases. An additional complication in the present case arises from the spin degrees of freedom in addition to the orbital variables and from the anisotropy of the hopping parameters.

As inferred above from the results for the temperature dependent magnetization \( m(T) \), the magnetic ground-state appears to be homogeneous for binary disorder, but may be inhomogeneous in the case of doped scatterers (see Figs. 1(a),(c)). Since the magnetism is partially controlled by the antiferromagnetic superexchange coupling \( J_s \), we study the effect of increasing \( J_s \) for a fixed large disorder strength of random scatterers. Fig. 6(a) shows the result for \( m(T) \) and Fig. 6(b) the result for the temperature dependence of \( D_Q(\mathbf{q}_0) \). The saturation value of \( m(T) \) as well as the onset scale for ferromagnetism decrease with increasing \( J_s \). More importantly, \( D_Q(\mathbf{q}_0) \) at low temperatures increases with increasing \( J_s \) indicating an enhancement in the orbital ordering. For a homogeneous system this would mean that orbital ordering and antiferromagnetism are both enhanced with increasing \( J_s \). This is a contradiction to the Goodenough-Kanamori rules, which state that an orbitally antiferromagnetic system should be magnetically ferro. The contradiction is resolved by analyzing the microscopic details of this complicated state providing an example where the real-space structures are essential for a comprehensive understanding.

We show in Fig. 7, the effect of the superexchange coupling on the real-space patterns of lattice and spin variables. The lattice correlations are shown in the top row and the analogously defined spin correlations \( C_S(i) = (1/4) \sum_{s} S_i \cdot S_{i+d} \) in the bottom row. For \( J_s = 0.02 \) the system contains orbitally ordered nanoscale regions, but magnetically it appears homogeneous. For \( J_s = 0.06 \) the area of the orbitally ordered regions is enlarged and magnetic inhomogeneities appear. The orbitally ordered regions remain ferromagnetic, while the orbitally disordered regions become antiferromagnetic upon increasing
ever, upon increasing $J_s$ further to 0.1, the antiferromagnetic regions start to extend also into the orbitally ordered clusters. The charge density patterns (not shown here) are insensitive to the increase in $J_s$.

IV. CONCLUSIONS

Our analysis for a two-band double-exchange model for manganites leads us to conclude that the disorder induced orbital ordering in manganites near $x = 0.3$ is properly described, if the density of scattering centers tracks the hole concentration. Within this specific model of quenched disorder the induced staggered orbital ordering is responsible for the orders of magnitude increase in the low-temperature resistivity as observed in the experiments in Ref. 5.

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