Interplay of charge, spin, orbital and lattice correlations in colossal magneto resistance manganites

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We derive a realistic microscopic model for doped colossal magnetoresistance manganites, which includes the dynamics of charge, spin, orbital and lattice degrees of freedom on a quantum mechanical level. The model respects the SU(2) spin symmetry and the full multiplet structure of the manganese ions within the cubic lattice. Concentrating on the hole doped domain (0 ≤ $x$ ≤ 0.5) we study the influence of the electron-lattice interaction on spin and orbital correlations by means of exact diagonalisation techniques. We find that the lattice can cause a considerable suppression of the coupling between spin and orbital degrees of freedom and show how changes in the magnetic correlations are reflected in dynamic phonon correlations. In addition, our calculation gives detailed insights into orbital correlations and demonstrates the possibility of complex orbital states.

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I. INTRODUCTION

The observation of the colossal magnetoresistance effect (CMR) in doped manganese oxides with perovskite structure, R$_{1-x}$A$_x$MnO$_3$ (R = rare-earth, A = alkaline-earth metal), moved these materials into the focus of intense research activity. $^{1}$ It turned out soon that the complex electronic and magnetic properties of the manganites depend on a close interplay of almost all degrees of freedom known in solid state physics, namely itinerant charges, localised spins, orbitals, and lattice vibrations. On the one hand, the strong Coulomb interaction $U$ and the Hund’s rule coupling $J_h$ introduce a spin background and affect the charge mobility via double-exchange. $^{2}$ On the other hand, the cubic environment of the Mn sites within the perovskite lattice results in a crystal field splitting of Mn-$d$-orbitals into $e_g$ and $t_{2g}$ and gives rise to an orbital degeneracy in the ground-state of Mn$^{3+}$ ions. This orbital degeneracy, in turn, connects the electronic system to the lattice, making it sensible to Jahn-Teller distortion and polaronic effects. $^{3}$

There are numerous attempts to describe the electronic (i.e., charge, spin, and orbital) interactions of the manganites on a microscopic level (see, e.g., Refs. 4, 5, 6, 7). However, most of these models do not reflect the full multiplet structure of the Mn ions caused by the cubic site symmetry or violate the spin rotational invariance. To our knowledge, Feiner and Oleś $^8$ were the first who derived a consistent spin-orbital model for the undoped compounds. In the following we extend their derivation to the case of finite doping and complete the resulting electronic model with dynamic electron-lattice interactions of Jahn-Teller and breathing type. Up to now models of similar complexity have only been discussed on the basis of rather extensive approximations, where mean-field approaches are probably the most popular. In particular, the lattice was always treated within the adiabatic approximation which is questionable in view of the comparable energy scales of spins and phonons. For a better understanding of this complex many body system we prefer to consider all interactions on an equal footing. Using exact diagonalisation techniques we study the ground-state properties of the derived model on a four site cluster.

For the hole doped region (0 ≤ $x$ ≤ 0.5) our calculations show how the lattice can effectively control the spin and orbital correlations and the charge mobility. In the undoped compounds the electron-phonon interaction suppresses the coupling of spin and orbital degrees of freedom and is most effective in determining orbital order. At doping $x = 0.25$, where ferromagnetism is stabilised by the double-exchange mechanism, the coupling to the lattice can cause self-trapping of the charge carriers, which immediately switches the spin order. In turn, this change is reflected in dynamic lattice correlations, namely the bond length fluctuation. For the half-doped manganites we find that the electron-phonon coupling enhances the susceptibility for charge ordering which is present already due to the strong Coulomb interaction. In addition, moderate variations in the strength of the electron-lattice coupling trigger pronounced changes in spin and orbital correlations. This relates to the complicated patterns of spin, charge and orbital ordering observed experimentally for different manganites at $x = 0.5$.

II. MICROSCOPIC MODEL

A. Electronic interactions

The electronic and magnetic properties of the mixed-valence manganites are governed by the manganese $d$-electrons, which are divided into $t_{2g}$ and $e_g$ according to the cubic symmetry within the crystal field. Due to the large Coulomb and Hund’s rule interactions each of the
and the spins of several \( d \)-electrons are aligned in parallel at every site (see Figure 1). Starting from the undoped compounds with only Mn\(^{3+} \) ions doping will remove \( e_g \)-electrons. Hence, the local electronic Hilbert space can be restricted to the large Hund’s rule ionic ground-states of manganese ions in a cubic crystal field (see Ref. 5). For Mn\(^{3+} \) \((d^3)\) this corresponds to the spin-2 orbital doublet \( ^3E \ [t_2^3 (A_2) c] \),

\[
|\theta, 2, m \rangle = + \sqrt{\frac{(2m-3)}{3(2m+1)}} (S^+)^{(2m+1)} c^\dagger_{\xi_+} c^\dagger_{\eta_+} c^\dagger_{\zeta_+} |0\rangle
\]

\[
|\epsilon, 2, m \rangle = - \sqrt{\frac{(2m-3)}{3(2m+1)}} (S^+)^{(2m+1)} c^\dagger_{\kappa_+} c^\dagger_{\eta_+} c^\dagger_{\zeta_+} |0\rangle ,
\]

and for Mn\(^{4+} \) \((d^3)\) to the spin-\( \frac{3}{2} \) orbital singlet \( ^4A_2 \ [t_2^3] \),

\[
|a_2, \frac{3}{2}, m \rangle = \sqrt{\frac{(2m-3)}{3(2m+1)}} (S^+)^{(2m+1)} c^\dagger_{\xi_+} c^\dagger_{\eta_+} c^\dagger_{\zeta_+} |0\rangle .
\]

Here the operators \( c^\dagger_{\alpha \sigma} \) create spin \( \sigma \) electrons in the \( e_g \) \((\alpha = \theta, \xi)\) or \( t_{2g} \) \((\alpha = \xi, \eta, \zeta)\) orbitals and \( S^+ = \sum \alpha c^\dagger_{\alpha \uparrow} c_{\alpha \downarrow} \) raises the on-site spin. The overlap of neighbouring manganese \( d \) and oxygen \( p \) orbitals allows for a tunneling of the manganese electrons between adjacent sites. Due to the specific symmetry of the involved orbitals this hopping acquires an anisotropy \( [3, 13] \).

\[
H_t = - \sum_{i, \delta, \sigma} R_\delta \left[ t c^\dagger_{i, \delta \sigma} c_{i+\delta, \theta \sigma} + t_\theta (c^\dagger_{i, \xi \sigma} c_{i+\delta, \xi \sigma} + c^\dagger_{i, \eta \sigma} c_{i+\delta, \eta \sigma}) \right] + \text{H.c.} ,
\]

which can be expressed in terms of the spatial rotations

\[
R_\epsilon = (C^d_3)^1 , R_\eta = (C^d_3)^2 = (C^d_3)^{-1} , R_\zeta = (C^d_3)^3 = 1
\]

\[
C^d_3 : c_{\theta/\epsilon} \rightarrow -\frac{1}{\sqrt{3}} c_{\theta/\epsilon} \mp \frac{1}{\sqrt{3}} c_{\zeta/\eta} ;
\]

\[
c_{\xi/\eta} \rightarrow c_{\eta/\zeta}.
\]

The transfer amplitudes \( t \) and \( t_{2g} \) of \( e_g \) and \( t_{2g} \) electrons are small compared to the local energies \( U \) and \( J_h \). Therefore the mobility of the \( e_g \) electrons depends on the correlations of the spin background formed by the \( t_{2g} \) electrons. Their kinetic energy is maximal, if neighbouring spins are aligned ferromagnetically, which is the essence of the well known double-exchange interaction \( [3] \). Second order processes in \( t \) and \( t_{2g} \) are responsible for numerous superexchange interactions between the localised spins, which inherit the orbital anisotropy of the hopping.

The electronic part of our model is derived by second order degenerate perturbation theory \( [3] \) i.e., we calculated the matrix elements of \( H_t \) between the basis states \( (1) \) or \( (2) \) and all admissible excited states \( (6) \). In Figure 2 the spin part of the fifteen different virtual excitations contributing to the superexchange is summarised graphically. We neglect terms which involve three different lattice sites.

A compact expression for the resulting electronic Hamiltonian,

\[
H_{el} = \sum_{i, \delta} R_\delta (H^z_{i, i+\delta} + \text{H.c.}) ,
\]

is obtained by rewriting the orbital and charge degrees of freedom of the basis states \( (1) \) and \( (2) \) in terms of new Fermi operators \( d^\dagger_\alpha \) and projectors \( P^\alpha \),

\[
|\theta \rangle = d^\dagger_\theta |0\rangle , \quad |\epsilon \rangle = d^\dagger_\epsilon |0\rangle , \quad |a_2 \rangle = d^\dagger_0 d^\dagger_\epsilon |0\rangle ,
\]

\[
P^\theta = n_\theta (1 - n_\epsilon) , \quad P^\epsilon = n_\epsilon (1 - n_\theta) , \quad P^{a_2} = n_\epsilon n_\theta .
\]

Note that these projectors are related to the common pseudo-spin operators

\[
\tau^z = \frac{1}{2} \sigma^z , \quad \tau^{x/y} = R_x/y (\tau^z) = \frac{1}{2} (\sigma^z \pm \sqrt{3} \sigma^x) ,
\]

by the equation \( R_\delta (P^\theta/\epsilon) = \frac{1}{2} \pm \delta \). Since the amplitude of the on-site spin is different for the basis states \( (1) \) and \( (2) \), it is convenient to represent the spin degree of

![Fig. 1: Local electronic structure of Mn-\( d \)-electrons in a cubic environment. Here \( \Delta_{cl} \), \( J_h \), and \( E_{Jr} \) denote the crystal field splitting, the Hund’s rule coupling, and the Jahn-Teller splitting, respectively.](image)

![Fig. 2: Second order virtual excitations contributing to the model Hamiltonian \( H_{el} \). The shaded region corresponds to \( t^2 \) terms, the other terms are proportional to \( t_2 \). \( n \) denotes different nonnegative prefactors of \( J_h \).](image)
freedom by Schwinger bosons \( a_s \) and \( a_l \) which allow for a uniform description. Then, the spin operator is defined by
\[
2S = a_μ^† σ_μ a_ν,
\]
but the amplitude is subject to the constraint
\[
2 |S| = a_s^† a_s + a_l^† a_l = 4 - n_θ n_ε.
\]
Using this notation in \( z \)-direction the interaction between nearest-neighbour sites is given by,

\[
H_{ij}^z = - \frac{t}{5} (a_i^† a_j^† + a_i a_j) \left( d_{ij}^0 n_{i,\theta} d_{j,\theta} n_{j,\epsilon} \right) + \frac{t^2}{32 J_h} S_i S_j - \frac{3 P_{ij}^a n_{i,\theta} - \frac{1}{2} P_{ij}^a P_{ij}^\theta}{10(U - 5 J_h)} P_{ij}^\theta
\]

The first order in \( t \) corresponds to the well known double exchange interaction, which the authors discussed in detail recently. The second order in \( t \) and \( t_π \) appears to be more involved, since the number of accessible virtual excitations is rather large (see Figure 2). However, in all cases it is basically the product of a Heisenberg-type spin interaction and two orbital projectors. The energies of the virtual excitations depend, in general, on all three Racah parameters, \( A, B, \) and \( C \). Applying the convention of Ref. 14 we define \( U \) and \( J_h \) in terms of \( d^4 d^3 = d^2(4 A_1) d^3 \) and \( d^4 d^1 = d^1(3 A_1) d^3 \) excitations. Together with the approximation \( C \approx 4B \) this yields \( U = A + 22B \) and \( J_h = 6B \). Typical estimates for these energies are \( U \approx 6 \ldots 9 \) eV and \( J_h \approx 0.6 \ldots 0.8 \) eV [15,12,23,24].

### B. Electron-phonon interaction

Concerning the electron-lattice interaction we concentrate on the local environment of the manganese ions, namely the surrounding oxygen octahedra (see Figure 3). At every site two optical phonon modes of \( E_g \) symmetry, \( q_θ \) and \( q_ε \), couple to the orbital degree of freedom of the \( e_g \) electrons. In addition, a breathing-mode \( q_{a_1} \) is sensitive to the electronic density. To lowest order in the elongation \( q_{a_1} = b_{i,a_1}^0 + b_{i,a_1} \) \((\alpha \in \{θ, ε, a_1\})\), this is modelled by the \( E \otimes e \) Jahn-Teller Hamiltonian [24]

\[
H_{JT} = g \sum_i \left[ (n_{i,θ} - n_{i,ε}) (b_{i,θ}^0 + b_{i,θ}) + (d_{i,θ}^0 d_{i,ε} + d_{i,ε}^0 d_{i,θ}) (b_{i,θ}^0 + b_{i,ε}) \right]
\]

and a Holstein-type [25] interaction

\[
H_{br} = \tilde{g} \sum_i \left( n_{i,θ} + n_{i,ε} - 2n_{i,θ} n_{i,ε} \right) (b_{i,a_1}^0 + b_{i,a_1})
\]

![FIG. 3: Distortions of the oxygen octahedra for the Jahn-Teller \((q_{θ}, q_{ε})\) and the breathing-type \((q_{a_1})\) phonon modes.](image)

The bosonic operators \( b_{i,α} \) describe the usual harmonic lattice dynamics. To a good approximation we can assume these phonons to be dispersion-less,

\[
H_{ph} = \omega \sum_i \left[ b_{i,θ}^1 b_{i,θ}^0 + b_{i,ε}^1 b_{i,ε}^0 \right] + \tilde{ω} \sum_i b_{i,a_1}^1 b_{i,a_1}^0.
\]

In experiments [26,27] the vibrational modes involving distortions of the oxygen octahedra are found to have frequencies of the order of 50 to 80 meV. In the following we assume \( g = \tilde{g} \) and \( \omega = \tilde{ω} \). This approximation reduces the number of model parameters and is reasonable, since we are mainly interested in qualitative features of the electron-lattice interaction.

### III. NUMERICAL RESULTS

The Hilbert space of the complete microscopic model,

\[
H = H_{el} + H_{JT} + H_{br} + H_{ph},
\]

is large and grows rapidly with the system size. However, using a density matrix based optimisation procedure for the phonon subsystem we are able to retain the full
quantum dynamics of the lattice and the electronic subsystem in our numerical calculation of the ground-state properties. In addition we take into account some of the discrete symmetries of the model, namely the conservation of the $S^z$ component of the spin, the particle number conservation, and the mirror symmetries orthogonal to the $x$ and $y$ axes (dot-dashed in Figure 5(b)). Nevertheless, the typical dimension of the eigenvalue problem is of the order of $10^6$ and its repeated solution during the phonon optimisation requires the use of large scale computers.

![Spatial symmetries of the considered cluster.](image)

**FIG. 4: Spatial symmetries of the considered cluster.**

### A. Undoped manganites

The undoped manganese oxides (LaMnO$_3$, PrMnO$_3$) usually exhibit A-type anti-ferromagnetic order and strong Jahn-Teller distortion of the ideal perovskite structure.[4,5] The origin of the observed magnetic order has been subject to discussions. While different band structure calculations underscore the importance of lattice distortions for the stability of anti-ferromagnetism, Feiner and Oles[4] favoured a purely electronic mechanism.

Our calculation points out that both parameters, $U/J_h$ and $g$, can drive a transition from ferromagnetic (FM) to anti-ferromagnetic (AFM) order. Figure 5(c) shows the phase diagram of the electronic model without electron-phonon interaction, i.e., $g = 0$. We assume $t = 0.4$ eV and $t/t_z = 3$ for the hopping integrals and characterise the magnetic phases according to the total spin $S^{tot}$ of the ground-state of the four site cluster. Parameters in the range $U \leq 5J_h$ implicate that for two neighbouring sites a $d^8d^3$ configuration becomes the ground-state in favour of the $d^4(\pi E)d^4(\pi E)$ configuration. This is incompatible with the situation in the manganites and consequently the electronic Hamiltonian $H_{el}$ is not applicable for these values of $U$ and $J_h$. Starting from the FM phase with $5J_h \leq U \leq 9.2J_h$ both, increasing $U$ or $g$, change the magnetic order of the ground-state to AFM [Figure 5(a) and (b)]. The magnetic transition is accompanied by a change in the corresponding orbital order, which we identify by the local expectation value $\langle n_\theta - n_\varepsilon \rangle$ and the orbital exchange correlation $(\sigma_i^\mu \sigma_{i+\delta}^\nu)$ between neighbouring sites. Here the Pauli matrices $(\sigma_i^\mu)_{\mu\nu}$ operate on the orbital degree of freedom, $\mu, \nu \in \{\theta, \varepsilon\}$, at the site $i$. The panels (a) and (b) of the Figures 5(c) and 6 illustrate the transition from staggered to uniform orbital order.

In view of the distinct driving interactions ($U$ or $g$) both transitions appear to be very similar. However, we observe a significant difference, if we study the (de)coupling of spin and orbital degrees of freedom. The latter has been a rather controversial issue in the case of...
the Kugel-Khomskii model\textsuperscript{[26]}

\[
H = J \sum_{\langle ij \rangle} \left[ 4(S_i S_j)(\tau_i^\alpha + \frac{1}{2})(\tau_j^\alpha + \frac{1}{2}) + (\tau_i^\alpha - \frac{1}{2})(\tau_j^\alpha - \frac{1}{2}) - 1 \right],
\]

which contains the same kind of spin-orbital interactions \( S_i S_j \tau_i^\alpha \tau_j^\alpha \) as our Hamiltonian. Using equation of motion approaches for the approximate solution of the model\textsuperscript{[13]} Khaliullin and Oudovenko\textsuperscript{[27]} decoupled spin and orbital degrees of freedom, while Feiner, Oléš and Zaanen\textsuperscript{[28]} emphasised the role of mixed spin-and-orbital excitations (dot-dot-dashed in Figure 4). However, since this approximation is not taken into account in the calculations\textsuperscript{[e.g., the spin-orbital fluctuation in Figure 6 (c)].} As the distortions develop, which is also reflected in spin and orbital correlations with increasing \( q \).

At intermediate values of \( g / \omega \approx 1 \) a finite \( x-y \)-distortion develops, which is also reflected in spin and orbital correlations e.g., the spin-orbital fluctuation in Figure 3(c)]. Such type of asymmetry seems surprising in view of the invariance of the cluster with respect to diagonal reflections (dot-dot-dashed in Figure 3). However, since this symmetry is not taken into account in the calculation the system is trapped in one particular linear combination of two degenerate ground-states and different expectation values in \( x \) and \( y \) direction can evolve. At larger electron-phonon coupling \( g \) only a finite \( q_x \) distortion remains and the \( q_x \) modes are uncorrelated.

In Figure 7 the change of orbital, spin and phonon correlations with increasing \( g \) is shown schematically. This, however, is a rather suggestive picture, which can not cover all the details of the various correlations. To select the appropriate graphical representation for the orbital arrangement we study the \( E \otimes E \)-eigenstates of reduced orbital density matrices. On a bond \( \langle ij \rangle \) these states are either anti-symmetric,

\[
|a\rangle_{ij} \propto (|\theta_i\rangle \otimes |\epsilon_j\rangle - |\epsilon_i\rangle \otimes |\theta_j\rangle),
\]

or symmetric,

\[
|s(\varphi, \psi)\rangle_{ij} \propto (|\varphi_i\rangle \otimes |\psi_j\rangle + |\psi_i\rangle \otimes |\varphi_j\rangle),
\]

with respect to the permutation of two sites. The angles \( \varphi, \psi \in \mathbb{C} \) parameterise two generalised orbital states of the form \(|\varphi\rangle = \cos(\varphi)|\theta\rangle + \sin(\varphi)|\epsilon\rangle\). At small \( g \) the staggered orbital order implies that the anti-symmetric orbital state is the most probable for each bond, i.e., it belongs to the largest eigenvalue of the density matrix. Since an anti-symmetric combination of two arbitrary generalised states \(|\varphi\rangle \) and \(|\psi\rangle \) is always proportional to \(|a\rangle_{ij} \) the representation is undefined. Therefore we took into account also the second eigenstate of the density matrix, which is slightly less probable but necessarily symmetric. The associated angles \( \varphi \) and \( \psi \) define the orbital pattern shown in the left hand panel of Figure 3. For larger \( g \) the uniform orbital order allows for a calculation of the depicted orbital pattern directly from the most probable eigenstate of the density matrix. The spin arrangements are chosen such as to reflect the expectation.
of the bond length in the FM planes of the A-type AFM structure. Nevertheless, the orbital arrangement within the FM spin background closely resembles the data measured within the FM planes of the A-type AFM structure.

values of the Heisenberg interaction, $\langle S_i S_j \rangle$, for nearest neighbour bonds $\langle ij \rangle$. Of course, the calculated ordering patterns can not be directly compared to experimental observations for the three dimensional compounds. Nevertheless, the orbital arrangement within the FM spin background closely resembles the data measured within the FM planes of the A-type AFM structure.

**B. Doping $x = 0.25$**

In view of the CMR effect the domain of low to intermediate doping ($0.15 \leq x \leq 0.5$) is certainly the most interesting one. Here ferromagnetism is stabilised by the double exchange interaction, which depends on the mobility of the charge carriers. However, if too strong electron-phonon coupling causes localisation of the holes, the spin order breaks down. For $x = 0.25$ our calculation of the ground-state properties clearly illustrates this coincidence. Figure 4(a) shows the total spin $S_{tot}$ of the cluster together with the expectation value of the kinetic energy in the ground state,

$$E_{kin} = \left\langle -\frac{t}{2} \sum_{i,\delta,\sigma} R_{\delta} \left[ a_{i,\sigma} a_{j,\sigma}^\dagger d_{i,\delta,\epsilon} n_{i,\epsilon} d_{j,\sigma}^\dagger n_{j,\delta}^\dagger \right] + \text{H.c.} \right\rangle.$$  

(17)

Obviously, the change from FM to AFM spin correlations with increasing electron-phonon interaction $g$ is directly related to the charge mobility. In addition the transition is accompanied by the appearance of a lattice distortion in the $x$-$y$-plane [see Figure 11(c)], which is interesting also in view of the lattice dynamics. Whereas one of the elongations, $\langle q_y \rangle$, grows linearly in $g$, the associated fluctuation $\langle q_x^2 \rangle - \langle q_x \rangle^2$ shows a kink near the FM to AFM transition point. This behaviour closely reminds the data collected by Booth et al. using x-ray-absorption fine-structure measurements (XAFS). For La$_{1-x}$Ca$_x$MnO$_3$ these authors observed a similar rise in the Mn-O bond length variance $\sigma^2$ near the critical temperature of the transition from the ferromagnetic metallic to the paramagnetic insulating phase.

Orbital correlations are again extracted from the eigenstates of the reduced density matrices for a single bond. We concentrate on the environment of a hole, which is characterised by the $E \otimes A_2$ eigenstates, i.e., by states describing a Mn$^{3+}$ site neighbouring a Mn$^{4+}$ site. Each of these states can be understood as the product $|\varphi \rangle_i \otimes |a_2 \rangle_j$ of a generalised orbital state $|\varphi \rangle$ and the basis state $|a_2 \rangle$ [see Eq. (2)]. In Figure 8(b) the corresponding angle $\varphi$ of the most probable eigenstate of the density matrix is given for bonds in $x$ and $y$ direction. The data for weak and strong electron-phonon coupling $g$ is translated into the orbital arrangement presented in Figure 11. Increasing $g$ reduces nearest-neighbour correlations and forces the orbital order to depend locally on the dominant electron-phonon interaction. The orbital polaron pattern disappears, if the charge carrier is trapped completely.

**C. Doping $x = 0.5$**

At doping $x = 0.5$ the picture is more involved. Again we consider model parameters, which yield ferromagnetic spin correlations for vanishing electron-phonon coupling, $g = 0$. The strong Coulomb interaction $U$ then leads to the formation of a charge density wave with ordering vector $(\pi, \pi)$ in reciprocal space. Increasing electron-phonon coupling $g$ amplifies this trend, and at large $g$ the trapping of the carriers gives rise to charge order. In Figure 11(c) this is illustrated with the density-density correlation between different lattice sites $i$ and $j$. From the model Hamiltonian (8) it is obvious that if charges tend to maximise their mutual distance the particular antiferromagnetic component of the Heisenberg spin interaction, which is proportional to $t^2 / J_h$, gains importance. Consequently FM order is unstable at much lower $g$, if compared to the case $x = 0.25$. The FM to AFM transition is not connected to charge localisation and causes...
Further increase of $g$ causes the carriers to localise and the distortion to disappear. The spin background undergoes a change from FM order to different types of AFM order. Of course, within the four site system we are not able to detect more complicated spin arrangements like the CE-type order observed experimentally.

However, the sensitivity of the system to small changes in the model parameters is clearly visible. Note that the depicted orbitals represent the amplitude of the underlying complex states $|\psi\rangle$.

### IV. CONCLUSION

In the present work we have derived a microscopic model for doped CMR manganites which includes the dynamics of charge, spin, orbital, and lattice degrees of freedom on a quantum mechanical level. Using exact diagonalisation techniques we have studied how the electron-lattice interaction affects short range spin and orbital correlations. An observation, which is important for the understanding of the undoped compounds, is the suppression of the spin-orbital coupling with increasing electron-phonon interaction. For the weakly doped compounds we have demonstrated the direct relationship between the trapping of charge carriers and the breakdown of ferromagnetism. In addition, we have shown that changes in the spin correlations are reflected in dynamic lattice correlations. At intermediate doping we find that the system depends on a subtle balance of double-exchange, superexchange and electron-lattice interaction. The latter enhances the tendency for charge ordering which in turn affects spin and orbital order. Besides the calculation proves that complex orbital states can be a suitable approximation for the description of orbital correlations. Although the calculated data is not quantitatively comparable to real, three dimensional materials, the exact diagonalisation of even a small system provides detailed insight into correlations and driving interactions behind the rich phase diagram of the manganites. In addition, the exact results may support the development of approximate theories.

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1. R. M. Kusters, J. Singleton, D. A. Keen, R. McGreevy, and W. Hayes, Physica B 155, 362 (1989).
2. R. von Helmolt, J. Wecker, B. Holzapfel, L. Schultz, and K. Samwer, Phys. Rev. Lett. 71, 2331 (1993).
3. S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnach, R. Ramesh, and L. H. Chen, Science 264, 413 (1994).
4. J. M. D. Coey, M. Viret, and S. von Molnár, Adv. Phys. 48, 167 (1999).
5. Y. Tokura and Y. Tomioka, J. Magn. Magn. Mater. 200, 1 (1999).
6. E. Dagotto, T. Hotta, and A. Moreo, Physics Reports 344, 1 (2001).
