Dimensional cross-over and charge order in half-doped Manganites and Cobaltites

Oron Zachar\textsuperscript{1,2} and Igor Zaliznyak\textsuperscript{1}

\textsuperscript{1}Brookhaven National Laboratory, Upton, New York 11973-5000 USA, \textsuperscript{2}UCLA Dep. of Physics, Los Angeles, CA 90095, USA.

We propose a generic model for understanding the effect of quenched disorder on charge ordering in half-doped Manganese- and Cobalt-Oxides with different crystal structures. Current experimental observations (Table I) are explained in the light of the global phase diagram of the model (Fig. 2).

In the past decade much theoretical work was devoted to elucidating the nature and origin of charge ordering (CO) in the doped transition metal oxides, particularly in manganites, which are of great experimental and practical interest \textsuperscript{[1,2,3]}. Much attention was focused on studying the interplay of charge correlations with orbital and spin degrees of freedom which are often important for the CO \textsuperscript{[4]}. Surprisingly, however, no coherent approach to understanding the effect of quenched disorder, such as introduced by a random distribution of dopant ions, on charge order (distinguished from orbital order) has been proposed so far.

Dopant disorder is a source of an unavoidable random electrostatic potential which couples linearly to charge density fluctuations \textsuperscript{[2]}. In a strongly correlated electron system close to criticality such perturbation is of crucial importance. We propose that essential account for this random potential can be formulated in terms of a generic model \textsuperscript{[5]}, mapping the CO interaction on an effective Ising problem. We construct the phase diagram of this model, Fig. 2, and find that it adequately describes the essential features of the CO structure in half-doped manganese and cobalt oxides. In particular, a dimensional cross-over in our model gives a natural explanation to the puzzling experimental observation that CO \textsuperscript{[5-7,8,9,10]} in the layered perovskites is short-range \textsuperscript{[5,6,7]}, while it is long-range in the pseudo-cubic materials \textsuperscript{[8,9,10]}.

At half-doping, the ground state of Co oxides and of many isostructural Mn oxides manifests a checkerboard-like planar charge modulation accompanied with a Jahn-Teller (JT) distortion of the MO\textsubscript{6} (M = Mn, Co, etc.) octahedra shown in Fig. 1 \textsuperscript{[11,12]}. The electrons are highly localized in a charge-ordered state with alternating Co\textsuperscript{2+} and Co\textsuperscript{3+} valence (Mn\textsuperscript{4+} and Mn\textsuperscript{3+} in manganites), and the in-plane O\textsuperscript{2−} ions move towards the higher-valence M ions. To be explicit, we present the model construction in terms of a La\textsubscript{1.5}Sr\textsubscript{0.5}CoO\textsubscript{4} \textsuperscript{[12]} which has no superimposed further symmetry breaking due to orbital order.

As a general framework, we argue that hopping dynamics, orbital, magnetic, local Coulomb and Jahn-Teller interactions can all be integrated out into an effective charge-ordering interaction which then competes with the random charge potential introduced by the dopant ions. A perfectly ordered state has a two-fold degeneracy associated with the choice of sublattice occupied by the higher/lower valence ions. We define a local Ising variable, \(\tau(r) = \pm 1\), to denote the two alternative configurations, Fig. 2(c). A single-domain CO state corresponds to a ferromagnetic order of \(\tau(r)\). Topological disorder is introduced by domain walls as shown in Fig. 2(d).

Because of the Jahn-Teller distortion, there is an elastic strain energy cost for nearest neighbor cells with opposite \(\tau\) values, Fig. 2(d). This strain energy can be described by an effective short-range ferromagnetic interaction \(J(r-r')\tau(r)\tau(r')\textsuperscript{[12]}\). Without a loss of generality we may approximate \(J(r-r')\tau(r)\) by a nearest-neighbor coupling. The potential introduced by dopant ions randomly favors one or the other configuration of a two-ion cell, and therefore effectively acts as a random field \(h(r)\tau(r)\). Importantly, depending on the crystal structure the effective coupling may be anisotropic, \(J_{ab} \neq J_{ac}\).

Hence, for the charge order, the effect of elastic crystal strain and random dopant distribution is captured by an effective \textit{anisotropic} random-field-Ising-model (RFIM),

\[
H = -J_{ab} \sum_{\langle rr'\rangle_{ab}} \tau_r \tau_{r'} - J_c \sum_{\langle rr'\rangle_c} \tau_r \tau_{r'} + \sum_r h_r \tau_r ,
\]

where \(\langle rr'\rangle_{ab}\) and \(\langle rr'\rangle_c\) denote nearest-neighbor cells in the \(ab\)-plane and along the \(c\)-axis respectively, see Fig. 2(e), (f). To our knowledge, all previous analytical and numerical studies focused on the isotropic \(3D\) RFIM. Here
we make the first attempt aimed at a qualitative and quantitative understanding of the anisotropic RFIM 11.

Random field alters both lower critical dimension 13 and critical exponents 14 of an Ising system. In particular, for weak random field, $\Delta/J < 1$, the isotropic 3D RFIM orders at a finite temperature $T_c > 0$, while the 2D RFIM has no long range order (LRO) even at $T = 0$ ($J$ and $\Delta = \{\text{runs of } h_i\}$ parameterize the Ising coupling and disorder potential, respectively). This prompts an interesting question: what is the fate of a 3D anisotropic random field Ising model (RFIM) of 11? The simplest way of connecting the renormalization group (RG) flows leads us to the phase diagram shown in Fig. 2.

To connect our model analysis with the experiment, we summarize the charge order correlation lengths in the $ab$-plane, $\xi_{ab}$, and along the $c$-axis, $\xi_c$, measured in several representative perovskite oxides in Table I. Correlations in the pseudo-cubic systems I-III (Fig. 1 (e)) are resolution-limited (typically this means $\xi \gtrsim 2000 \, \AA$ 8 3), and CO is apparently long-range. On the other hand, in the layered systems IV-V (Fig. 1 (f)) the CO is finite-range, with highly anisotropic 3D correlations $\xi_{ab} \gg \xi_c$, which implies $J_{ab} \gg J_c$. Large in-plane correlation length $\xi_{ab} \gg 1$ indicates weak planar disorder, $\Delta \ll J_{ab}$. Therefore, these materials probe the most interesting and complicated region of the phase diagram.

Because the isotropic 3D RFIM is long-range ordered for $\Delta < \Delta^{3D} \approx 2.27 J$ 12 13, it is clear that for $\Delta < J_c < J_{ab}$ the system is in the regime of weak 3D disorder and has a 3D LRO. Thus, we examine an intriguing case,

$$J_c \ll \Delta < J_{ab},$$

where the $c$-axis coupling is in the regime of strong disorder, while the $ab$-planes are in the weak disorder limit.

For $J_c = 0$ the system decouples into independent 2D RFIM planes, and the ground state is disordered. This form of disordered ground state is perturbatively stable. For an infinitesimal inter-plane coupling $J_c/\Delta \ll 1$, the $ab$-planes remain uncorrelated on all length scales ($c$-axis correlation length $\xi_c < 1$), and each plane may still be treated as an effectively independent 2D RFIM. Therefore, a critical line of phase transitions exists in the $(J_{ab}/\Delta, J_c/J_{ab})$ phase diagram, separating the disordered and 3D-LRO phases (solid line in Fig. 2). With increasing $J_c/J_{ab}$ on the disordered side we enter a critical regime, where the ground state is still disordered but with significant and highly anisotropic correlations in all spatial directions. In the isotropic 2D and 3D RFIM 16 the ground state correlation length scales with $\kappa = \Delta/J$ as $\xi^{2D} \approx \exp \left(\frac{-\Delta}{J^{2D}}\right)$, and $\xi^{3D} \approx (\kappa - \kappa_c)^{-\nu}$ (for $\kappa \geq \kappa_c$), respectively. What are the correlation lengths in a highly anisotropic 3D RFIM 11, with $J_c \ll \Delta < J_{ab}$?

The phase diagram, Fig. 2 instructs that upon a RG transformation model 11 scales either to a 3D LRO fixed point, or towards a 3D strong disorder fixed point. Currently, there is no generally satisfying analytical derivation of the RG equations for the RFIM for $d < 4$. However, because $d = 2$ is the lower critical dimension of the RFIM, 3D random field critical fixed point should be accessible perturbatively in $2 + \varepsilon$ expansion, 17. Therefore, we derive perturbative RG equations for the anisotropic 3D RFIM 11 starting from a semi-phenomenological scaling "ansatz" for the 2D RFIM 16 17. These equations, which naturally account for the RG flow of the coupling anisotropy, allow us to derive and analyze all qualitative features of the phase diagram of Fig. 2.

For the isotropic $d$-dimensional RFIM a simple scaling analysis of Imry-Ma, \(\{J(l) \sim l^{d-1}; \Delta(l) \sim \sqrt{l^d}\Delta\}\), 12 17, predicts the relative disorder $\kappa = \frac{\Delta}{J}$ to scale as

$$\kappa(l) = \frac{\Delta(l)}{J(l)} \approx \sqrt{\frac{l^d}{l^{d-1}}},$$

The marginality of $\kappa(l)$ in 2D is associated with $d = 2$ being the lower critical dimension. It is broken by subdominant contributions from the interface roughening at the domain boundaries which lead to a disordered ground state 17 17. For weak disorder, $\kappa \ll 1$, the correlation length is $\xi = A \exp \left(\frac{-1}{\sigma \kappa^2}\right)$, with $\sigma \approx 1$ (clearly, this expression fails for strong disorder, where $\xi \leq 1$). By demanding that $\kappa(l)$ scales consistently with the correlation length, $\xi(l) = \xi/l$, and with the expression

\[
\begin{array}{llll}
\text{#} & \text{Material} & \text{Ref.} & \xi_{ab} \quad \xi_c \\
I. & \text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3 & 8 & \infty \quad \infty \\
II. & \text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3 & 9 & \infty \quad \infty \\
III. & \text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3 & 10 & \infty \quad \infty \\
IV. & \text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_4 & 6 & \approx 35 \approx 3.3 \\
V. & \text{La}_{1.5}\text{Sr}_{0.5}\text{CoO}_4 & 5 & \approx 4 \leq 1 \\
\end{array}
\]
\[ \xi(l) = A \exp \left\{ +\frac{1}{2} \kappa^{-2}(l) \right\}, \]

i.e. scaling of the relative disorder in 2D is governed by the logarithmic corrections to Eq. (4). The disordered ground state is manifested by the flow of \( \kappa^{-2}(l) = \frac{J(l)}{\Delta(l)} \to 0 \). In the context of the model (11), equation (14) describes the RG flow for \( J_c = 0 \).

To proceed with deriving the qualitative features of the RG flows shown in Fig. 4 for \( 0 < J_c \leq J_{ab} \), we note that the anisotropy of interactions is, in fact, an irrelevant perturbation at the 3D critical point. This is reflected by the fact that all stable fixed points are located on the isotropic line in the phase diagram, at \( J_c/J_{ab} = 1 \). At the mean-field level (and without random fields) the anisotropy of the interactions can be removed by appropriate anisotropic re-scaling of the coordinates. Here, we implement this idea by devising an anisotropic real-space RG transformation, effectively coarse-graining to anisotropic blocks \( l_{ab} \times l_{ab} \times l_c \). We derive the RG equations along the lines of 2 + \( \varepsilon \) expansion, and demonstrate explicitly the flow towards the isotropic fixed points.

We parametrize the anisotropy as \( \alpha = \left( \frac{\kappa_c}{\kappa_{ab}} \right)^x, \ x > 0 \), and define the RG transformation through an infinitesimal anisotropic re-scaling, \( d(l_{ab}) = d(l_{ab}) \); \( d(l_c) = \alpha(l) d(l_{ab}) \). This determines the shape of the coarse-grained blocks to be gradually varying, flowing from the anisotropic towards isotropic. We use the Imry-Ma approximation (17) to derive the lowest order contributions to the flow equations for the model parameters, and supplement it by the appropriate higher order correction to reproduce the scaling equation (14) for the 2D RFIM case, \( \alpha = 0 \). The resulting perturbative RG equations are

\[
\frac{d}{dl} \left( \kappa^2 \right) = -\alpha \kappa^2 + \sigma \kappa^4 + O(\alpha^2) \quad (5)
\]

\[
\frac{d\alpha}{dl} = x (1 - \alpha) \alpha \quad (6)
\]

We obtain an unstable 2D fixed point at \( \{ \alpha^* = 0, \kappa^2 = 0 \} \), and stable isotropic 3D fixed points at \( \alpha^* = 1 \). The 3D critical point is at \( \{ \alpha^* = 1, \kappa^2 = \kappa_c^2 = \kappa_0^2/\alpha \} \). With \( \kappa_c \approx 2.27 \) from numerical simulations (16), the extrapolation of our perturbative equations to \( \alpha = 1 \) will imply \( \sigma \approx 0.2 \), which is too small. This indicates that while qualitatively correct, and valid in the perturbative region of parameters, RG equations (5), (6) are inadequate for quantitative estimates.

It is straightforward to integrate Eq. (14) and obtain \( \alpha(l)^{-1} = 1 + (\alpha(l)^{-1} - 1)l^{-2} \), the scaling flow of the anisotropy. In principle, the phase transition line can be determined by a further numerical integration. The important asymptotes can be derived analytically,

\[
\alpha(\kappa^2) \approx C_0 e^{-\frac{\sigma \kappa^2}{\kappa_{ab}^2}}, \quad \alpha \ll 1, \quad (7)
\]

\[
\alpha(\kappa^2) \approx 1 + C_1 (\kappa^2 - \kappa_c^2), \quad 1 - \alpha \ll 1, \quad (8)
\]

and show that neglecting \( \mathcal{O}(\alpha^2) \) term in Eq. (5) is indeed justified for \( \alpha \ll 1 \), but not for \( \alpha \approx 1 \).

To obtain quantitative estimates of the anisotropic correlation lengths as a function of \( J_c, J_{ab} \) and \( \Delta \), we use an alternative approach. In the absence of the exact RG equations for the 3D ARFIM, we resort to a real-space rescaling procedure where we employ the Migdal-Kadanoff bond-moving technique (18) combined with the scaling ansatz (14). We adopt conventional approximations in ignoring the generation of longer range interactions (16) and using the “majority rule” (18) for block spin variable. Hence, we follow only the transformation of effective nearest neighbor block interactions \( J_{ab}(l) \), \( J_c(l) \) and effective disorder parameter \( \Delta(l) \).

Our strategy is to map the anisotropic RFIM onto a “solvable” isotropic 3D RFIM, whose known properties enable us to derive the estimates for \( \xi_{ab}(J_{ab}, J_c, \Delta) \) and \( \xi_c(J_{ab}, J_c, \Delta) \). Because the correlation length scales as \( \xi(l) = \xi/l \), a bare anisotropic 3D RFIM model with finite \( \xi_{ab} > \xi_c \geq 1 \) is mapped onto an effective isotropic 3D model with \( \xi_{ab} = \xi_c \) when the coarse-grained block size \( l_{ab} \times l_{ab} \times l_c \) is chosen with \( l_{ab} = \xi_{ab}/\xi_c \) and \( l_c = 1 \). In other words, the cross-over from quasi-2D to isotropic 3D scaling is intuitively implemented in two steps. First, a 2D transformation to blocks of size \( l_{ab} \times l_{ab} \times 1 \), and then a usual isotropic 3D scaling (see Fig. 3).

Performing the Migdal-Kadanoff transformation to \( l_{ab} \times l_{ab} \times 1 \) blocks we first move the \( J_c \) bonds (18) and thus create an effective block interaction \( J_{ab}(l_{ab}) = J_{c,ab}^2 \), Fig. 3B. Now that the c-axis coupling is removed, each planar \( l_{ab} \times l_{ab} \) section can be integrated out in a 2D fashion using (14). This approximation is equivalent to assuming that fluctuations on length scale \( l_{ab} < \xi_{ab}/\xi_c \) in \( ab \)-planes are uncorrelated in the c-axis direction. In addition, we keep the Imry-Ma approximation for scaling of the disorder \( \Delta(l_{ab}) \) under 2D block transformation (17).

Indeed, a perturbative expression for the surface tension per unit length on the scale \( l \) in 2D RFIM (16), \( \Sigma(l) = J \left[ 1 - \sigma \kappa^2 \ln(l) \right] \), indicates that logarithmic corrections to \( \kappa(l) \) come from the effective weakening of the inter-block interaction \( J(l) \) as domain boundaries adjust to the random field. We thus arrive at the final form of our approximate scaling equations for block \( l_{ab} \times l_{ab} \times 1 \),

\[
J_c(l_{ab}) = J_c^2 \quad (9)
\]

\[
J_{ab}(l_{ab}) = J_{ab} l_{ab} \sqrt{1 - \sigma \kappa^2 \ln(l_{ab})} \quad (10)
\]

\[
\Delta(l_{ab}) = \Delta_{ab} \quad (11)
\]

For \( l_{ab} = \xi_{ab}/\xi_c \), the correlation lengths in the transformed model are isotropic \( \xi_{ab}(l_{ab} = \xi_{ab}/\xi_c) = \xi_c \). Since
the disorder remains isotropic, it must be that the rescaled interactions are also isotropic. Thus, by imposing a self-consistency condition, $J_{ab}(l_{ab} = \xi_{ab}/\xi_c) = J_c(\xi_{ab}/\xi_c)$, we obtain a mapping of the bare anisotropic RFIM \(1\) onto isotropic 3D RFIM with effective interactions

$$J_{ab}(l_{ab} = \xi_{ab}/\xi_c) = J_c(\xi_{ab}/\xi_c) = \left(\frac{\xi_{ab}}{\xi_c}\right)^2 J_c,$$

and $\Delta(\xi_{ab}/\xi_c) = \left(\frac{\xi_{ab}}{\xi_c}\right) \Delta$. Using \(10\), \(11\), \(12\) and expression for the correlation length of the 3D RFIM, $\xi_{3D} = \Lambda(\kappa_c^{-2} - \kappa^{-2})^{-\nu} = \xi_c$, we obtain

$$\left(\frac{J_{ab}}{\Delta}\right)^2 = \kappa_c^{-2} - \left(\frac{\Lambda}{\xi_c}\right)^{1/\nu} + \sigma \ln \left(\frac{\xi_{ab}}{\xi_c}\right),$$

$$\left(\frac{J_c}{\Delta}\right)^2 = \left(\frac{\xi_c}{\xi_{ab}}\right)^2 \left[\kappa_c^{-2} - \left(\frac{\Lambda}{\xi_c}\right)^{1/\nu}\right].$$

Substituting $\xi_c = 1$, we obtain the crossover line $\xi_c \gtrsim 1$ in the phase diagram,

$$\frac{J_c}{J_{ab}} = \frac{\Delta}{J_{ab}} \sqrt{\kappa_c^{-2} - \Lambda^{1/\nu} e^{-\frac{\xi_{ab}}{\xi_c} + \frac{\xi_{ab}}{\xi_c} - \kappa_c^{-2}}}.$$

With $\kappa_c \approx 2.27$, $\Lambda \approx 0.1$ and $\nu \approx 1.37$ from numerical simulations, \(13\), \(14\) allow us to retrodict the effective model parameters. For $La_{0.5}Sr_{1.5}CoO_4$ (Table 1, V) we find $\frac{\Delta}{J_{ab}} \approx 0.85$, and $\frac{\Delta}{J_c} > 53$, indeed satisfying the relations $J_c \ll \Delta < J_{ab}$ under which our approximations are valid. For $La_{0.5}Sr_{1.5}MnO_4$ (Table 1, IV) $\frac{\Delta}{J_{ab}} \approx 0.64$ and $\frac{\Delta}{J_c} \approx 49$. In both cases the anisotropy of the bare coupling parameters is remarkably high! Yet, we think it is quite realistic for the layered systems. Because neighbor $ab$-planes are shifted so that MOG octahedra in one plane fit in-between those in the other (see Fig. 1(f)), and the inter-plane spacing is rather large, breathing-type distortions of the octahedra accompanying the CO are weakly coupled between the planes. On the other hand, octahedra within each plane form a corner-sharing network, so the coupling of in-plane distortions is strong. In the pseudocubic perovskite structure, the MOG octahedra share apical oxygens along the $c$-axis as well (see Fig. 1(c)), and crystal fields are more isotropic. Note that, for a system to have a LRO groundstate, interactions need not be isotropic. In fact, the phase diagram instructs us that only very strongly anisotropic systems will be disordered if the disorder potential is weak (as exemplified by Eq. \(4\), and the above estimates of $J_{ab}/J_c$ in the layered compounds).

In conclusion, we argued on very general grounds that charge order observed in half-doped manganites and their isostructural relatives is described by the anisotropic ($J_c \neq J_{ab}$) 3D random field Ising model \(1\). We constructed the schematic phase diagram of this model, Fig. 2, and supported its intuitive structure by perturbative RG equations \(9\), \(10\). We also obtained the quantitative estimates of the effective model parameters as a function of the measured correlation lengths using a Migdal-Kadanoff block transformation scheme combined with a phenomenological scaling equation \(3\) for a 2D RFIM.

Long-range charge order is observed in the pseudocubic materials at low temperatures. In contrast, a disordered ground state with anisotropic correlations is found in the layered compounds. This disparity is naturally explained in our model: because of the strong anisotropy of the layered materials these systems reside in the different regions of the phase diagram. The anisotropic correlation lengths measured in $La_{0.5}Sr_{1.5}CoO_4$ and $La_{0.5}Sr_{1.5}MnO_4$ corroborate our conclusion. We note that a disordered 3D RFIM has well known features which may be verified experimentally. Furthermore, our analysis may be applied to thin films, where even in pseudocubic compounds correlations in one direction are limited by the film thickness. The latter, together with the model parameters, will then determine the CO finite correlation length within the film. Finally, we note that charge disorder in the form of dislocations, as in Fig. \(1d\), has inevitable effects on the superimposed orbital and spin orders which have thus far not been elaborated.

We thank S. A. Kivelson, A. Tsvelik, F. Essler and J.P. Hill for fruitful discussions, and acknowledge the financial support of DOE#DE-AC02-98CH10886 (O.Z. and I.Z.) and DOE#DE-FG03-00ER45798 (O.Z.).

[1] A. Millis, B. I. Shraiman, P. B. Littlewood, Phys. Rev. Lett. 74, 5144 (1995).
[2] E. Dagotto, T. Hotta, A. Moreo, Physics Reports 344 (2001).
[3] E.L. Nagaev, Physics Reports 346, 387 (2001).
[4] Y. Tokura and N. Nagaosa, Science 288, 462 (2000).
[5] I. A. Zaliznyak et al, Phys. Rev. Lett. 85, 4353 (2000); I. A. Zaliznyak et al, Phys. Rev. B 64 195117 (2001).
[6] B. J. Sternlieb et al, Phys. Rev. Lett. 83, 4872 (1996).
[7] Y. Wakabayashi et al, J. Phys. Soc. Jpn. 70, 1194 (2001).
[8] P. G. Radaelli, D. E. Cox, M. Marezio, S.-W. Cheong, Phys. Rev. B 55, 3015 (1997).
[9] M. v. Zimmermann et al, Phys. Rev. Lett. 76, 2169 (1999).
[10] K. Nakamura et al, Phys. Rev. B 60, 2425 (1999).
[11] Among pseudocubic manganites such CO occurs in some Sr-doped and most of the Ca-doped systems, but not in $La_{0.5}Sr_{0.5}MnO_3$ and $Pr_{0.5}Sr_{0.5}MnO_3$.
[12] G. A. Gehring and K. A. Gehring, Rep. Prog. Phys. 38, 1 (1975).
[13] Y. Imry and S. K. Ma, Phys. Rev. Lett. 35, 1399 (1975).
[14] A. Aharony, Y. Imry, S. K. Ma, Phys. Rev. Lett. 37, 1364 (1976); G. Grinstein, Phys. Rev. Lett. 37, 944 (1976).
[15] A.A. Middleton and D.S. Fisher, cond-mat/0107489.
[16] For a review see T. Nattermann and J. Villain, Phase Trans. 11, 5 (1987).
[17] John Cardy, “Scaling and Renormalization in Statistical Physics”, Cambridge U. Press (1996).
[18] Leo P. Kadanoff, Ann. of Phys., 100, 359 (1976).