An intriguing generic characteristic of complex solids such as doped manganites, cuprates, and cobaltates seems to be the coexistence of patches of metallic and insulating regions (often dubbed as ‘phases’). This ‘electronic inhomogeneity’ can vary from nanometers to microns and can be static or dynamic. Questions as to whether this can be characterized as ‘electronic softness’ and is a defining feature of these materials, its origins and role in determining their electronic properties, etc., are among the most actively explored issues. However, proposed mechanisms are either at the level of scenarios or toy models, and do not adequately address the specifics of the scale and nature of inhomogeneities in the actual systems.

Motivated by the fact that electronic inhomogeneities came to prominent attention first in manganites, in this letter we address these issues using extensive simulations on a new model with Coulomb interactions amongst two electronic fluids, one localized (polaronic), the other extended (band-like), and dopant ions. The long range Coulomb interactions frustrate phase separation induced by the strong on site repulsion between the fluids. A single quantum phase ensues which is intrinsically and strongly inhomogeneous at a nano-scale, but homogeneous on meso-scales, with many characteristics (including colossal responses) that agree with experiments. This, we argue, is the origin of nanoscale inhomogeneities in manganites, rather than phase competition and disorder related effects as often proposed.

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The above model (2) on finite 3d periodic lattices of sizes \( H \) Hartree approximation, using the notes regions with separates the lightest region, where no \( b \) states are occupied, from the intermediate shade region, which has \( b \) states occupied in puddles. For larger doping, percolating clumps are obtained and the system is a metal (right). The inset is results in the absence of long range Coulomb interaction \( (V_0 = 0.0) \) and shows ‘macroscopic phase separation’. All energy scales are in the units of \( t \).

![FIG. 1: (color online) Real space electronic distribution obtained from simulations on a 16³ cube. Magenta (darkest) denotes hole clumps with occupied \( b \) electrons, white (lightest) denotes hole clumps with no \( b \) electrons, cyan (2nd lightest) denote singleton holes, and light blue (2nd darkest) represents regions with \( \ell \) polarons. The configuration on the left shows isolated clumps with occupied \( b \)-electrons (\( b \)-electron puddles). For larger doping, percolating clumps are obtained and the system is a metal (right). The inset is results in the absence of long range Coulomb interaction \( (V_0 = 0.0) \) and shows ‘macroscopic phase separation’. All energy scales are in the units of \( t \).](image)

![FIG. 2: (color online) Critical doping levels \( x_{c1} \) and \( x_{c2} \). \( x_{c1} \) separates the lightest region, where no \( b \) states are occupied, from the intermediate shade region, which has \( b \) states occupied in puddles. The darkest shaded region, with \( x > x_{c2} \), has some occupied \( b \) states that percolate through the 10³ box. The solid line corresponds to the DMFT result \( E_{JT} \) for \( x_{c1} \).](image)

The Coulomb term \( H_C \) has two parts; the charge at site \( i \) has energy \( q_i \Phi_i \), where \( \Phi_i \) is the electrostatic potential there due to \( \cal{A} \) \( K^{2+} \) ions, and the interaction between the charges at site \( i \) and \( j \) leads to an energy \( V_0 \frac{q_i q_j}{|r_{ij}|} \).

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Furthermore, since \( U \) is large, for simplicity we take the \( U \to \infty \) limit; i.e., assume that \( b \) electrons do not hop to sites occupied by \( \ell \), so the two kinds of electrons form disjoint clusters. On a hole-cluster, which has two or more \( h \)-sites each accessible to the other by electron hoppings, which we will refer to as a “clump”, the \( b \) electron energy levels are determined exactly by solving the intersite Hamiltonian \( H_b = -t \sum_{ij} \langle b_i^\dagger b_j \rangle + \text{h.c.} \). The ground state (i.e., at zero temperature) is obtained by starting from an overall charge neutral trial configuration random of \( \ell \) polaronic sites, and performing electron transfers that lower energy, till none such exist. Some of these are of the type \( \ell - h \) as in a classical Coulomb glass \([12]\), in which an \( \ell \) electron is moved to a hole site. In addition there are \( \ell - b \) and \( b - b \) transfers involving one or two \( b \)-states whose energy is quantum mechanically obtained for a particular clump structure. At each iteration then, we find the best possible transfer, i.e., the one which lowers most the (occupied) single particle levels whose energies include Coulomb interactions. Then we perform the transfer, and update the clump structure, if necessary, i.e., redistribute the \( b \) electrons in the new clumps. The process is repeated till the final \( b \)-clump/\( \ell \)-polaron structure is stable against all further electron transfers. This is a new generalization of the common Coulomb glass simulation \([13]\) which includes the quantum mechanically obtained \( b \) states within their clump or puddle. The electrostatic energy is calculated accurately using the Ewald technique and fast Fourier transform routines.

In the discussion below, all length scales are normalized by the lattice parameter \( a \), and we use dimensionless energy parameters \( E_{JT} \) and \( V_0 \), scaled by the hopping amplitude \( t \). Realistic values \([11]\) for manganites are \( 2.0 \leq E_{JT} \leq 3.0 \) and \( 0.01 \leq V_0 \leq 0.1 \) with \( t \approx 0.2 \text{eV} \). In the absence of \( V_0 \), the system “phase separates”, i.e., holes move to one side of the box and several \( \ell \) polarons are converted to \( b \) electrons that occupy the low energy band states with energies below \(-E_{JT} \) in this large clump (FIG. 1 see inset). This phase separation is due to strong local correlations (large \( U \) between \( \ell \) and \( b \) states), and is in agreement with known results for the Falicov-Kimball model \([11]\). The presence of \( V_0 \) renders this phase separation energetically unfavorable, and intermixes the phases as expected \([14]\). The favored electronic configurations strongly depend on the JT energy \( E_{JT} \) and the doping \( x \). Two examples are shown in FIG. 1. For a given \( E_{JT} \), there are two critical values of doping, \( x_{c1} \) and \( x_{c2} \), as shown in FIG. 2. For \( x < x_{c1} \), there are no occupied \( b \)-electron states in the system, and the holes form a Coulomb glass \([12]\). For doping larger than \( x_{c1} \), some occupied \( b \)-electron puddles appear (FIG. 2(a)). On further increase of doping, clumps interconnect and percolate through the simulation box (FIG. 2(b)), giving rise to the possibility of occupancy of \( b \)-states extended throughout the system, and hence metallicity. The results for \( x_{c1} \) and \( x_{c2} \) are insensitive to \( V_0 \) for \( V_0 \leq 0.5 \).
The physics behind these results is uncovered by a study of the density of states (DOS) of the $\ell$-polarons and $b$-electrons shown in FIG. 3. We find that the chemical potential $\mu$ (FIG. 3) goes essentially as $\mu = -E_{JT} + V_0$. The polarons form a Coulomb glass with a soft gap (FIG. 3 inset) at the chemical potential, as in the classical Coulomb glass. Furthermore, the polarons have their energies spread over an energy scale of order $V_0$, $\ell$. The $b$ electrons have a band-like DOS with a bandwidth that depends on the doping $x$. In fact, we find that the effective half bandwidth of the $b$ electrons scales as $D_{eff} = D_0 \sqrt{x}$ ($D_0$, the bare half-bandwidth, = 6$t$ for the cubic lattice), in remarkable agreement with an earlier single site dynamical mean field theory (DMFT) result obtained for $\ell$! Furthermore, the simulation results show that the $b$ band center (marked by $E_b$ in the figure) is not affected by doping, and scales as $E_b \approx V_0$ (for small $V_0 \leq 0.5$).

The behavior of the chemical potential and the band center are due to the fact that the random distribution of Ak ions plays an important role in determining the real space structure. From a study of the positional correlation function between two holes, and a hole and an Ak ion, we find that the holes tend to cluster around the Ak ions. Furthermore, the electrostatic screening is quite strong in this system in that these correlation functions (both hole-hole and hole-Ak ion) reach a plateau within a few lattice spacings. Since the holes cluster around the (oppositely charged) Ak ions, it is clear that a typical $\ell$ polaron will have a larger number of $\ell$-polaron neighbors than hole sites. Thus, the $\ell$ polaron sites see, on average, an electrostatic potential of order $V_0$, and the average energy of an $\ell$ polaron is increased from $-E_{JT}$ to $-E_{JT} + V_0$. Since the chemical potential of a Coulomb glass is the average energy of the states, it follows that the chemical potential is $-E_{JT} + V_0$. Similarly every hole site sees an electrostatic potential of order $V_0$, implying that the $b$ band center is placed at $V_0$. These observations and the fact that the $b$ bandwidth scales as $\sqrt{x}$, suggests that the minimum doping for the the $b$-band bottom to touch the chemical potential (i.e., for $b$-state occupancy) is $x_c = \frac{(E_{JT}/V_0)²}{\sqrt{x}}$, exactly the DMFT prediction for $\ell$!

However, we note that the simulation results include important new physics not contained in the DMFT, namely that $b$ state occupancy does not by itself make the system a metal, as these states are localized inside the clumps. Based on inverse participation ratio, geometric percolation of the clumps and Kubo conductivity calculations, we have good estimates for a second, higher, critical doping level, $x_{c2}$, at which the system actually becomes a metal (FIG. 2), for which the occupied $b$-states should extend across the simulation box. Our results suggest that for $x < x_{c1}$, electrical transport should be that of a classical Coulomb glass ($\sigma \sim e^{-A/\sqrt{x}}$). For $x_{c1} < x < x_{c2}$, the transport should have two contributions - the first a Coulomb glass contribution of the $\ell$ polarons and the second a variable range type inter-puddle hopping of the electrons in the $b$ puddles, as is indeed observed in doped manganites. For $x > x_{c2}$ we find a highly resistive metal.

Another interesting aspect that we have investigated is the clump size $R$, and its dependence on $V_0$. For a (fictitious) uniform distribution of Ak ions, one can show by an approximate analytical calculation that $R \sim \frac{1}{\sqrt{V_0}}$, corresponding, for $E_{JT} = 2.5$ and $x = 0.3$, to clump sizes between 10 and 5 lattice spacings for $V_0$ between 0.01 and 0.1. The clump sizes for the more realistic, random, distribution of Ak ions obtained from our simulations are much smaller; even a very small $V_0$ produces clumps that are four to five lattice spacings, and this spacing is essentially independent of $V_0$ for realistic values of $V_0$ - in stark contrast to the analytical result above. Thus the long range Coulomb interaction is a ‘singular perturbation’ that prevents macroscopic phase separation; but the sizes and the distribution of the clumps are determined by the random distribution of the Ak ions. Thus we conclude that doped manganites as modeled by are necessarily and intrinsically electronically inhomogeneous, on a nanometric scale.

Our results have been obtained for a Hamiltonian and energy parameters that are very realistic especially in low bandwidth manganites with a large ferromagnetic region in their phase diagram. In sharp contrast to some of the earlier scenarios proposed in manganites, the nanoscale inhomogeneities we obtain are not due to ‘phase competition’ induced ‘phase separation’ between ‘insulating’ and ‘metallic’ phases frustrated by dis-
order as suggested by studies on ‘toy models’, such as
spin Hamiltonians\cite{1} or Hamiltonians with two localized
states and electron lattice coupling\cite{2}. Rather, they arise
due to the the long ranged Coulomb interactions frustrat-
ing the phase separation induced by strong local correla-
tions. The mechanism itself has been discussed in a vari-
ety of contexts\cite{3, 4, 5}, but to the best of our knowledge, ours is the first quantitative study on a realistic model
(that includes dopant ions) for any correlated oxide.

We emphasize that the nanoscale inhomogeneities we
obtain are present in both the insulating and metallic
phases of doped manganites between which one has a
transition as a function of doping, at \( x = 2 \); and furthermore, as is clear from Figs.\cite{6} and \( \ell \), each of these con-
stitutes a valid, single, thermodynamic phase that is ho-
mogeneous on meso-scales. These results are in conform-
ity with the electron holography results of Loudon et
al.\cite{7}, where even the ferro-metallic state is seen to have
interspersed in it nanoscale ‘insulating regions’, which
in the context of the present simulation are just the \( \ell \)-
clusters\cite{8}.

Furthermore, our work suggests that mesoscale phase
separation\cite{9}, or proximity to multicriticality\cite{10}, are not essential for explaining CMR in manganites. Given
the correspondence between the simulation results for the
extended \( \ell \)b model and the (homogeneous) DMFT re-
sults for the simple \( \ell \)b model, the single metallic phase
obtained here will show a ferro-metal to para-insulator
transition as well as CMR due to strong Hund’s cou-
bling between the Mn \( t_{2g} \) core spins and the \( b \) ele-
ctrons and thermal fluctuations. As shown elsewhere\cite{11},
the CMR arises from small field induced changes in the
energetics of the extended \( b \) states which are respon-
sible for charge transport, and the consequent exponential
changes in their occupancy. Indeed, by way of experimen-
tal confirmation, we note that there are many mangan-
ites without mesoscale inhomogeneities that show colos-
sal responses\cite{12}. Finally, our work reinforces sug-
gestions that the mesoscale patterns seen in experiments
arise from other sources such as long range elastic strains,
possibly due to defects\cite{13, 14}. In our model, they can be
generated from the strain dependence of the local energy
parameters such as \( E_{JT} \) and \( t \). Investigations along these
lines are in progress.

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