Manganese oxides have been intensively studied in recent years due to their colossal magnetoresistance (CMR) effect [1], where small changes in magnetic field induce large changes in their resistivity $\rho_{dc}$. The CMR behavior may originate in the nontrivial interplay of the charge, spin, orbital, and lattice degrees of freedom in these materials. Among the theories proposed for the CMR effect are those based on the recently observed intrinsic tendencies of models for manganites toward mixed-phase formation [2]. In this context, clusters of one phase embedded into the other, typically involving ferromagnetic (FM) metallic and FM or antiferromagnetic (AF) insulating phases, are believed to substantially increase the density of states and the compressibility $\kappa$ of the system [1]. The effect occurs even in stable metallic regimes at low-temperatures, as long as the density is close to those where phase-separation occurs [2]. A variety of experimental results are in qualitative agreement with these ideas and evidence is accumulating that manganites have important microscopic inhomogeneities, particularly in regimes of relevance for the CMR effect [3].

Recently, high-energy resolution angle-resolved photoemission (ARPES) measurements for the bilayer compound $\text{La}_{1.2}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7$ (hole density $x=0.4$) have been reported [4]. The most remarkable result was the existence of a pseudogap at the chemical potential $\mu$, substantially larger than the analog feature reported for the cuprates. The main purpose of this paper is to analyze whether similar PG characteristics appear in the DOS of manganite models when analyzed using unbiased computational many-body methods [5]. The results are surprisingly robust, showing that indeed a prominent pseudogap exists in the DOS, as long as the regime explored has mixed-phase characteristics. The conclusions are general and they should apply to other compounds with similar microscopic phase-separation tendencies as well.

The one-orbital Hamiltonian is given by $H = H_{\text{DE}} + H_{\text{AF}}$, with the Double-Exchange (DE) term defined as [6]

$$H_{\text{DE}} = -t \sum_{\langle \mathbf{i}, \mathbf{j} \rangle, \sigma} (c_{\mathbf{i}\sigma}^\dagger c_{\mathbf{j}\sigma} + \text{h.c.}) - J_H \sum_{\mathbf{i}\alpha\beta} S_i \cdot c_{\mathbf{i}\alpha}^\dagger \sigma_{\alpha\beta} c_{\mathbf{i}\beta}, \quad (1)$$

where $c_{\mathbf{i}\sigma}$ creates an $e_g$-fermion at site $\mathbf{i}$ with spin $\sigma$, and $S_i$ is the total $t_{2g}$-spin, assumed localized and classical ($S_i=1$). $J_H>0$ is the Hund coupling, and the rest of the notation is standard [6]. The AF coupling between localized spins. The two-orbitals Hamiltonian is $H_{\text{KJT}} = H_K + H_{\text{JT}} + H_{\text{AF}}$. The first term containing the kinetic energy and Hund coupling is

$$H_K = - \sum_{\mathbf{i}ab} \epsilon_i^a (c_{\mathbf{i}a\sigma}^\dagger c_{\mathbf{i}b\sigma} + \text{h.c.}) - J_H \sum_{\mathbf{i}} S_{\mathbf{i}} \cdot S_{\mathbf{i}}, \quad (2a)$$

where $a,b=1,2$ are the two $e_g$-orbitals, $\mathbf{u}$ are unit vectors along the main axes, $\mathbf{u}$ labels those axes, $S_{\mathbf{i}} = \sum_{\alpha\beta} c_{\mathbf{i}a\alpha}^\dagger \sigma_{\alpha\beta} c_{\mathbf{i}a\beta}$ is the spin of the mobile fermions, and the rest of the notation is standard [6]. In this paper, the unit of energy will be $t=1$ in Eq.(1), $t_{11}^1 = 1$ in Eq.(2a), and $J'$ will be fixed to 0.05 [6]. The coupling with JT-phonons ($Q_i^{11} = -Q_i^{22} = Q_i^{33}$, and $Q_i^{12} = Q_i^{21} = Q_i^{22}$) is [6]

$$H_{\text{JT}} = \lambda \sum_{\mathbf{i}ab} \epsilon_i^a Q_{\mathbf{i}}^a b Q_{\mathbf{i}}^b + \frac{1}{2} \sum_{\mathbf{i}} (Q_{\mathbf{i}}^{22})^2 + Q_{\mathbf{i}}^{33} \frac{1}{2}. \quad (2b)$$

Classical phonons are assumed for simplicity. The $e_g$-fermionic density $\langle n \rangle$ is adjusted with $\mu$, for both models Eqs.(1,2) (the hole-density is $x = 1 - \langle n \rangle$). To study their properties, a Monte Carlo (MC) algorithm for the classical spins and phonons and an exact diagonalization of a one-electron problem for a given spin-phonon background were used. The method allows dynamical calculations in real-time without uncontrolled analytical continuations, and it has been described in detail in previous publications [6]. The DOS $N(\omega)$ was obtained adding the one-particle spectral functions $A(\mathbf{k}, \omega)$, and it is directly related with the DOS of photoemission experiments.

Fig.1a contains $N(\omega)$ of the one-dimensional (1D) one-orbital model Eq.(1) at large Hund coupling ($J_H=\infty$ for
separated regimes. \(\kappa \rightarrow \infty\) for \(0.77<\langle n \rangle<1.0\). As a consequence, the finite-T state investigated in Fig.1a presents a large compressibility. Size effects are small, and the results are expected to be representative of the bulk limit. The most noticeable feature of Fig.1a is the deep minimum at \(\omega=\mu\), which clearly develops as T is reduced. Similar behavior has been observed for other parameters \((J_H, J', T\) and \(\langle n \rangle)\) investigated here, as long as \(\kappa\) is large. Fig.1b shows two-dimensional (2D) results, still for the one-orbital model. Here phase-separation at \(T=0\) was found in the interval \(0<\langle n \rangle<1.0\). Fixing \(T\) to a low value, and varying \(\langle n \rangle\), PG behavior is once again observed where \(T=0\) phase-separation occurs. In Figs.1a-b the PG is always centered at \(\mu\), i.e. the DOS is not rigid but adjusts with \(\langle n \rangle\) and \(T\). Also note that at densities with marginally stable \(T=0\) ground-states, PG remnants are observed (insets of Figs.1a-b), as long as \(\kappa\) remains large. This occurs in a narrow \(\langle n \rangle\)-range near phase-separated regimes.

**FIG. 1.** Pseudogap formation in the one-orbital model, as a function of \(\langle n \rangle\) and \(T\). (a) Density of states \(N(\omega)\) vs \(\omega-\mu\) at \(J_H=\infty\) and 1D. The overall density is \(\langle n \rangle \sim 0.88\) \((x \sim 0.12)\). Starting from the top at \(\omega-\mu=0\), the temperatures \(T\) of the first four lines are \(1/10, 1/17, 1/25, 1/40\), and the lattice size is \(L=30\). The dotted line with crosses was obtained at \(T=1/40\), but with \(L=100\) showing the absence of strong size effects. The inset contains the DOS for \(L=100\), \(T=1/40\), and \(\langle n \rangle \sim 0.77\), at a marginally stable density as \(T\rightarrow 0\); (b) \(N(\omega)\) vs \(\omega-\mu\) in 2D at \(J_H=\infty\), using a \(10 \times 10\) cluster and \(T=1/30\). The four lines from the top at \(\omega-\mu=0\) correspond to \(\langle n \rangle \sim 0.90, 0.92, 0.94\), and 0.97. The inset has results for \(\langle n \rangle=0.86\), a marginally stable density at \(T=0\).

Pseudogap features appear also in the two-orbital model with JT-phonons, again as long as \(\kappa\) is robust. For instance, working at large \(J_H\) and \(\lambda\), Fig.2a shows results at \(\langle n \rangle \sim 0.7\), varying \(T\). For the parameters of Fig.2a, MC studies showed that \(T=0\) phase-separation occurs in the interval \(0.5<\langle n \rangle<1.0\) between spin FM-phases that differ in the orbital-order pattern (uniform vs staggered) \[3\]. Fig.2a illustrates the PG development as \(T\) is reduced, similarly as for the one-orbital model. An analogous phenomenon also occurs at low-density (Fig.2b) where \(T=0\) phase-separation between spin FM- and AF-phases was numerically observed for \(0.0<\langle n \rangle<0.3\) \[3\]. At low \(\langle n \rangle\), results without electron-phonon coupling (Fig.3a) are similar to those with strong lattice coupling \(\lambda = 1.5\). Vestiges of the PG also appear at \(\langle n \rangle \sim 0.4\) (Fig.2c), which has a stable \(T=0\) ground-state based on previous studies \[3\]. A similar situation occurs at \(\lambda = 1\) and \(\langle n \rangle \sim 0.7\), near the phase-separation boundary (Fig.3b).

**FIG. 2.** Pseudogap behavior of the 1D two-orbitals model at large electron-phonon coupling, as a function of \(\langle n \rangle\) and \(T\). (a) \(N(\omega)\) vs \(\omega-\mu\) with \(J_H=8\), \(\lambda = 1.5\), \(L=20\), and \(\langle n \rangle \sim 0.7\) \((x \sim 0.3)\). The thin-dashed, thick-dashed, and solid lines correspond to \(T=1/5, 1/10\) and \(1/20\), respectively; (b) Same as (a) but at \(J_H = \infty\), \(L=22\) and \(\langle n \rangle \sim 0.5\). The thin-dashed, thick-dashed, and solid lines correspond to \(T=1/5, 1/10\) and \(1/25\), respectively; (c) Same as (a) but for \(T=1/15\) and \(\langle n \rangle \sim 0.4\), a stable density as \(T\rightarrow 0\).

To investigate the origin of the PG, in Fig.3c the charge-charge correlation in momentum-space \(N(q)\) is shown in the region of interest. At low hole-density, \(N(q)\) is enhanced at small-q in the PG regime, indicating the existence of extended charged structures. Similar conclusions are reached by noticing that the average FM-cluster size increases as \(T\) is reduced (Fig.3d). The overall study of the data accumulated here lead us to conclude that the PG formation is correlated with large charge fluctuations, which occur both with and without phonons in regions that phase-separate as \(T\rightarrow 0\), and in their vicinity.

Although the relation between PG formation and mixed-phase tendencies is clear, the sharpness of the PG feature and its location following \(\mu\) as \(\langle n \rangle\) is varied requires additional analysis. For this purpose, several \(t_{2g}\)-spin MC-configurations (snapshots) of the 1D one-orbital model (Fig.1a) were individually studied (the reasoning and results described below apply with minor changes to the two-orbital model with JT-phonons and in any dimension). The analysis of the one-electron energies re-
revealed that most of the MC spin configurations produce the PG feature, and, thus, a single MC-generated snapshot should be enough to understand the phenomenon. Visual analysis indicated that the configurations with PG contain FM clusters of an intermediate size between 2 to 6 lattice spacings, immersed in a background with AF-correlations (short-ranged since here T is comparable to J'). To understand why this spin arrangement produces a PG, consider for simplicity the low-⟨n⟩ limit. Since at J_H = ∞, the high and low density limits are identical, while at finite but large J_H they are very similar, our analysis applies to ⟨n⟩ close to 1 simply changing electrons by holes. A cartoon-like representation of a typical MC t_{2g}. spin snapshot is in Fig.4a. The electrons are correlated with the classical spins such that they mostly populate the FM regions (Fig.4b), and, thus, aligned t_{2g}-spins provide to the electrons an attractive effective potential (Fig.4c). When extra electrons are added, FM clusters (with one or more electrons) are created, and new (occupied) levels appear below µ. Then, the one-particle spectrum at ω < µ for one MC classical spin configuration contains energies corresponding to quasi-localized electrons in each FM-well, with a finite bandwidth (“cluster” band) caused by the different shapes of those wells, plus tunneling among them. The AF-regions contribute to ω > µ. As a consequence, the resulting DOS (Fig.4d) contains a pseudogap at ω = µ.

The present study also included the individual one-particle spectral functions A(k, ω). Figs.5a-b show the 2D one-orbital model at large J_H, low-T, and low hole-density. The data displays the PG feature, which appears at all values of the momenta which have appreciable weight at ω=µ. Similar behavior is obtained for the two-orbitals model at large λ (Fig.5c). These results are compatible with the explanation of PG behavior based on mixed-phase tendencies, since the clusters formed in such a state are not distributed forming a regular pattern. Then, there is no preferred momentum for the PG to occur, unlike in a charge-density-wave state. The results of Fig.5 are in excellent qualitative agreement with ARPES measurements for La_{1.2}Sr_{1.8}Mn_2O_7 [8], where the PG was observed all along the “Fermi surface”. Note also the dispersive character of the results of Fig.5, in qualitative agreement with the ARPES data [8]. They originate mainly from the AF-regions that induce an effective DE-hopping t_{eff}=t⟨cos(θ/2)⟩ which for the temperatures in Figs.5a-b is approximately t/2.

\[ \text{AF} \quad \text{FM} \quad \text{AF} \quad \text{FM} \quad \text{AF} \]

\[ \langle n \rangle \]

\[ V_{\text{eff}} \]

\[ \text{N}(\omega) \]

\[ \omega - \mu \]

\[ \text{Cluster Band} \]

FIG. 3. (a) N(ω) vs ω-µ for the 2D two-orbitals model without phonons (λ = 0) at low-density ⟨n⟩ ∼ 0.1, J_H = ∞, and T=1/20. Shown are sizes L=40 and 22, denoted by solid and dashed lines, respectively; (b) Same as (a) but at λ = 1.0, J_H=∞, 1/25 and ⟨n⟩ ∼ 0.7. This is at the boundary of the T=0 phase-separated region [3]. The solid and dashed lines correspond to L=40 and 50, respectively; (c) N(q) for the 1D one-orbital model with J_H=∞, T=1/30, and L=30. Open circles, full diamonds, open squares, full triangles, and open triangles, denote ⟨n⟩ = 0.75, 0.84, 0.87, 0.90, and 0.94, respectively; (d) Size of typical FM clusters for the 1D one-orbital model. S_F(j) (normalized to S_F(0)) is the classical spin-spin correlation (S_i • S_{i+j}) (j > i) measured only when S_F(1) is positive as a way to isolate FM clusters.

The results of Fig.5 are presented. A (populated) “cluster band” is formed (thick line). In (d) the resulting DOS is shown.

FIG. 4. Schematic explanation of the PG formation in the low-⟨n⟩ limit and for the one-orbital model. In (a) a typical MC t_{2g}-spin configuration is sketched. In (b), the corresponding n_{eff}-density is shown, with electrons mostly located in the FM regions. In (c), the effective potential felt by the electrons is presented. A (populated) “cluster band” is formed (thick line). In (d) the resulting DOS is shown.

Other unexpected effects in ARPES data are also reproduced in Fig.5. For instance, the width of the peaks found in Ref. [9] were as large as 0.5-1.0 eV, similar to the widths in Fig.5 which are of order 1-2t, with t estimated to be 0.2-0.4 eV [9]. The large width is caused by the disordering influence of localized spins and phonons, particularly within the AF-clusters which have only partially ordered spins at the T’s analyzed here. The peaks...
do not sharpen as $\mu$ is reached, as in experiments, and the concept of quasiparticle seems not applicable in the models studied here.

Motivated by the similarities of our results with those of ARPES studies for the bilayer manganites, here it is conjectured that the $x=0.4$ state of La$_{1.2}$Sr$_{1.8}$MnO$_7$ should present microscopic inhomogeneities, both above and below $T_C$. This is compatible with a recently reported neutron-scattering-based phase diagram, where coexistence of FM (metal) and AF (insulator) features were observed at $x=0.4$ and low-$T$. Our prediction is also compatible with other neutron scattering results that reported mixed AF/FM characteristics and short-range charge ordering due to cluster formation, for the same compound above $T_C$.

FIG. 5. $A(k,\omega)$ at $J_H=\infty$. (a) is for a 2D one-orbital case with $T=1/30$, $\epsilon=0.92$ ($\epsilon \sim 0.08$), and on a $12 \times 12$ cluster with periodic boundary conditions (PBC). Shown are results along $(0,0)$ to $(\pi,0)$; (b) Same as (a) but along $(0,0)$ to $(\pi,\pi)$; (c) Results for the 2D two-orbitals model with $T=1/10$, $\lambda=1.5$, $\epsilon=0.7$ ($\epsilon \sim 0.3$), on a $10 \times 10$ cluster, along $(0,0)$ to $(\pi,0)$, with PBC.

The general character of our explanation of the computational results suggests that PG behavior should be present in other compounds if they are in mixed-phase regimes. Previous studies suggest that this may occur in the small $x$ region of La$_{1-x}$Sr$_x$MnO$_3$ at low- and intermediate-$T$, as well as within the FM metallic phase at low-$T$ close to the metal-insulator transition where $\kappa$ is expected to be large. On the other hand, at $x=0.3$ this compound behaves more as a regular metal and it should not have a PG. In general, where $T_C$ is the largest where PG tendencies should be the weakest, correlated with weak CMR effects. In La$_{1-x}$Ca$_x$MnO$_3$ at low-$T$, the interesting region is again small $x$, but also $x \sim 0.5$ where phase-separation has been observed. If the insulating character of this compound above $T_C$ is caused by microscopic inhomogeneities, PG features should also appear in a wide range of compositions at intermediate-$T$.

Summarizing, simple models for manganites studied computationally have a DOS with robust pseudogap features in regions of parameter space with large compressibility, particularly low hole-doping. $N(\mu)$ is drastically reduced as $T$ is lowered in this regime. The PG was found for all momenta along the “Fermi surface”, in agreement with experiments. The effect is caused by the formation of FM metallic clusters in an insulating background, precursors of the $T=0$ phase-separation tendencies which occurs both with and without JT-phonons. Mixed-phase tendencies are crucial for PG formation and its associated $\rho_{dc}$ increase. This conclusion seems independent of the details of the models, and may occur even if cluster formation is caused by $1/r$-Coulomb interactions on phase-separated regions. The close relation between $N(\mu)$ and $\rho_{dc}$ expected here differs drastically from theories based on Anderson localization, or in a simple hopping reduction by spin fluctuations $t\rightarrow t(\cos(\theta/2))$, where $N(\mu)$ is approximately unchanged. The mechanism discussed in this paper can also be operative in non Jahn-Teller compounds with tendencies to phase-separation, such as diluted magnetic semiconductors.

Discussions with D. Dessau and M. Kubota are acknowledged. The authors are supported in part by grant NSF-DMR-9814350.

[1] Jin et al., Science 264, 413 (1994).
[2] S. Yunoki et al., Phys. Rev. Lett. 80, 845 (1998); S. Yunoki and A. Moreo, Phys. Rev. B58, 6403 (1998).
[3] S. Yunoki et al., Phys. Rev. Lett. 81, 5612 (1998).
[4] A. Moreo et al., Science 283, 2034 (1999).
[5] See for instance T. Egami, J. of Low Temp. Phys. 105, 791 (1996); M. Hennion et al., Phys. Rev. Lett. 81, 1957 (1998); S. Mori et al., C. H. Chen, and S-W. Cheong, Phys. Rev. Lett. 81, 3972 (1998). For additional references see Ref. [1].
[6] D. Dessau et al., Phys. Rev. Lett. 81, 192 (1998); D. Dessau and Z.X. Shen, in “Colossal Magnetoresistive Oxides”, Ed. Y. Tokura, Gordon and Breach 1999.
[7] C. Zener, Phys. Rev. 82, 403 (1951).
[8] In 1D $t_{11}=t_{22}=2t_{12}=2t_{21}$ was used. In 2D, the sets $t'_{11}=3t'_{22}=\sqrt{3}t'_{12}=\sqrt{3}t'_{21}$ and $t''_{11}=3t''_{22}=-\sqrt{3}t''_{12}=-\sqrt{3}t''_{21}$ were used (S. Ishihara et al., Phys. Rev. B56, 686 (1997)).
[9] This $J'$ reproduces the Néel temperature of the fully doped CaMnO$_3$ material.
[10] A. J. Millis, et al., Phys. Rev. Lett. 74, 5144 (1995).
[11] In 2D it was observed that the FM clusters have a variety of shapes including a large percentage with elongated 1D-like characteristics.
[12] M. Kubota et al., cond-mat/9902288, and private communications.
[13] T. G. Perring et al., Phys. Rev. Lett. 78, 3197 (1997); L. Vasilii-Doloc et al., preprint.
[14] $\rho_{dc}$ measurements on La$_{0.67}$Ca$_{0.33}$MnO$_3$ thin-films also provides evidence against Anderson localization (V. N. Smolyaninova et al., cond-mat/9903238).