Pseudogap in Doped Mott Insulators is the Near-Neighbor Analogue of the Mott Gap

Tudor D. Stanescu
Department of Physics and Astronomy, Rutgers University, 136 Frelinghuysen Road, Piscataway, New Jersey 08854-8019, USA

Philip Phillips
Loomis Laboratory of Physics, University of Illinois at Urbana-Champaign, 1100 W. Green Street, Urbana, Illinois 61801-3080, USA

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We show that the strong-coupling physics inherent to the insulating Mott state in 2D leads to a jump in the chemical potential upon doping and the emergence of a pseudogap in the single-particle spectrum below a characteristic temperature. The pseudogap arises because any singly occupied site not immediately neighboring a hole experiences a maximum energy barrier for transport equal to t^2/U, t the nearest-neighbor hopping integral and U the on-site repulsion. The resultant pseudogap cannot vanish before each lattice site, on average, has at least one hole as a near neighbor. The ubiquity of this effect in all doped Mott insulators suggests that the pseudogap in the cuprates has a simple origin.

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In a Mott insulator with one orbital per site, each unit cell contains an odd number of particles but the Fermi energy lies in the middle of a gap. In contrast, band insulators contain an even number of electrons per unit cell and the Fermi energy lies atop a full band. Consequently, Mott and band insulators are *not* adiabatically connected. Nonetheless, most Mott insulators tend to order antiferromagnetically below some temperature, T_N. As a consequence, it is standard to view the Mott state simply as an antiferromagnet in which the unit cell has doubled. On this view, the insulating properties of a Mott insulator are equivalent to those of a band insulator. Band insulators possess rigid bands, and hence doping creates quasiparticles only at the Fermi level. That this picture fails fundamentally in the parent cuprates, which are all antiferromagnetic Mott insulators, is immediately evident from optical conductivity experiments [1,2] which reveal that even for $T \gg T_N$, a gap of order 2 eV exists and doping leads to a massive reshuffling of spectral weight from 2 eV to the Fermi energy. These experiments lay plain that what is missing in the antiferromagnetic reduction is Mottness itself: (1) in the absence of magnetic ordering ($T > T_N$), a charge gap exists in the single-particle spectrum, (2) each electronic state in the first Brillouin zone has spectral weight both above and below the charge gap, and (3) the sum rule that each single-particle state carries unit weight is satisfied [3] only when the spectral function is integrated across the charge gap not simply up to the chemical potential as in a band insulator. A consequence of (3) is that in the Mott state, the traditional notion that the chemical potential demarcates the boundary between empty and occupied states fails fundamentally. This failure is central to Mottness.

The breakdown of the traditional band insulator sum rule in the cuprates is well described [3] by the Hubbard model in which the on-site energy for double occupancy leads to a charge gap at half-filling. Such local on-site physics dominates the insulating behavior at half-filling. In the lightly doped regime, $\delta \approx 0$, effective interactions of longer range come into play as neighboring sites now become correlated. If on-site correlation leads to a charge gap at half-filling, it is certainly a possibility that nearest-neighbor correlations for $\delta \approx 0$, for example, might lead to a suppression of the density of states at the chemical potential as well. In fact, it is well documented that all the underdoped cuprates possess a pseudogap [4] in the single-particle spectrum. However, the origin of this phenomenon is unknown. As the pseudogap does not appear to be a true $T = 0$ phase and the pseudogap line joins continuously to the Mott insulator, proposals which require broken symmetry [5–8] are difficult to reconcile with the Mott state.

Without any assumption as to the nature of the ground state, we show that the electron spectral function for the 2D Hubbard model contains a dip at the Fermi energy which results in a pseudogap in the single-particle density of states (DOS). The pseudogap remains pinned at the Fermi level in the underdoped regime but moves above it at an intermediate doping level, as is seen experimentally [4]. The pseudogap is fundamentally tied to *local* correlations on neighboring sites much the way the Mott gap arises from on-site physics.

The starting point for our analysis is the Hubbard model with nearest-neighbor hopping matrix element t and on-site Coulomb repulsion U. We base our strong-coupling analysis on a two-component composite basis ψ with $\psi_{1\sigma}(i) = \xi_{i\sigma}$ and $\psi_{2\sigma}(i) = \eta_{i\sigma}$ and its associated Green function $S(i, j, t, t') = \langle\langle \psi_{i\sigma}; \psi^\dagger_{j\sigma}\rangle\rangle = \theta(t - t')\langle\{\psi_{i\sigma}(t), \psi^\dagger_{j\sigma}(t')\}\rangle$, where $\xi_{i\sigma} = c_{i\sigma}(1 - n_{i-\sigma})$ and $\eta_{i\sigma} = c_{i\sigma}n_{i-\sigma}$. Here, $c_{i\sigma} = \eta_{i\sigma} + \xi_{i\sigma}$ annihilates an electron on-site i and n_i is the number operator for site i. The basis ψ exactly diagonalizes the on-site interaction and hence serves as a natural starting point for a strong-coupling analysis. To overcome the standard

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truncation problems inherent in the use of Hubbard operators, we adopt the following procedure. First, project [9] all new operators that arise from the Heisenberg equations of motion of the Hubbard operators onto the Hubbard basis. Second, write the self-energy exactly in terms of the remaining operators which are now orthogonal to the Hubbard basis. Third, use local methods in the spirit of dynamical mean-field theory (DMFT) [10] to calculate the resultant electron self-energy. To go beyond the single-site treatment indicative of dynamical mean-field theories [10], we adopt the two-site expansion proposed by Matsumoto and Mancini [11] in which the self-energy is determined self-consistently from a two-site Hubbard cluster embedded self-consistently in an interacting bath. As all orientations of the two sites are considered, the electron spectral function will be momentum dependent. Self-consistent cluster methods which are exact as the limit of an infinite cluster appear to be rapidly convergent, providing accurate results for the thermodynamics of the 1D and 2D half-filled bands [9] and, in fact, constitute the accepted methodology [10] for treating strongly correlated systems. Hence, an implementation of the Hubbard operators coupled with DMFT-type technology places the limitations not on truncation in the equations of motion but on the accuracy of the impurity solver and the size of the finite cluster. As the complete procedure is detailed elsewhere [9,12], we mention only that in contrast to the work of Matsumoto and Mancini [11], we required that for a fixed filling in the lattice, the chemical potential of the cluster equal that of the lattice.

Using this procedure [9,12], we report first the doping dependence of the chemical potential. Two distinct possibilities arise: (1) the chemical potential remains pinned and midgap states are generated or (2) the chemical potential jumps across the Mott gap. Our results shown in Fig. 1 demonstrate that the chemical potential jumps upon hole or electron doping, indicating an absence of midgap states. The magnitude of the jump is set by the Mott gap which is fully developed at $T = 0$. Even for $U = 4t$, the inset on the right shows that the chemical potential resides in the lower Hubbard band (LHB) for $n = 0.95$. While at some finite temperature, the chemical potential may appear to evolve smoothly, $\Delta\mu \neq 0$ as the doping increases, and hence no midgap states are present. Exact results in the 1D Hubbard model [13] as well as quantum Monte Carlo simulations [14] in 2D also reveal a chemical potential jump upon doping and hence no midgap states. However, unlike 1D and 2D, in $d = \infty$, the chemical potential remains pinned [10] upon doping as midgap states emerge. A chemical potential jump requires a large imaginary part of the self-energy at the Fermi energy. From the inset of Fig. 1, we find that $\mathrm{Im}\Sigma$ is initially large in the underdoped regime and acquires the characteristic ω^2 dependence in the overdoped regime indicative of a Fermi liquid. Consequently, the method we use here is capable of recovering Fermi liquid theory in the overdoped regime. Experimentally, whether $\Delta\mu$ vanishes or not appears to be cuprate dependent. For example, in $La_{2-x}Sr_xCuO_4$ (LSCO) [15], the chemical potential remains pinned roughly at 0.4 eV above the top of the LHB, while for $Nd_{2-x}Ce_xCuO_4$ (NDCO) [16] and $Bi_2Sr_2Ca_{1-x}R_xCu_2O_{8+y}$ (BSCO) [17–20], the chemical potential jumps upon doping and scales roughly as δ^2 as obtained here. Because stripes require $\Delta\mu = 0$, they have been invoked [21] to explain the origin of the midgap states in LSCO. The pseudogap in the underdoped cuprates has also been attributed [21] to stripes. However, because $\Delta\mu \neq 0$ for both NDCO and BSCO, a requirement for stripe formation, if the pseudogap has a universal origin in the cuprates, stripes are not its cause.

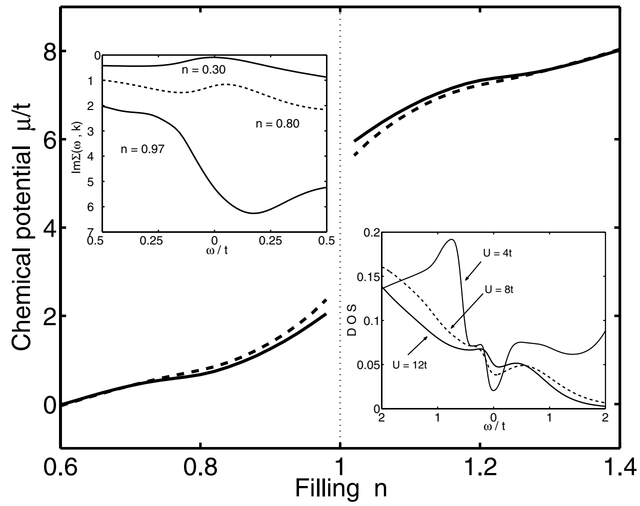

FIG. 1. Doping dependence of the chemical potential in the 2D Hubbard model computed using the local cluster approach for $T = 0.15t$ (dashed line) and $T = 0.07t$ (solid line). The inset on the left shows the imaginary part of the self-energy evaluated at a Fermi momentum $(0.3, 2.10)$ for $n = 0.97$, $(0.3, 1.84)$ for $n = 0.8$, and $(0.3, 1.06)$ for $n = 0.3$, whereas the inset on the right contains the density of states for $n = 0.95$ for $U = 4t$, $U = 8t$, and $U = 12t$.

To address the origin of the pseudogap, we focus on the doping dependence of the electron spectral function, $-\mathrm{Im}(S_{11} + 2S_{12} + S_{22})/\pi$, shown in Fig. 2. Six features are evident: (1) the chemical potential moves farther into the LHB as the filling decreases, (2) no coherent peaks exist near the chemical potential for $n = 0.97$, (3) each state in the first Brillouin zone (FBZ) has spectral weight both above and below the chemical potential as dictated by Mottness, (4) the Mott gap remains intact but moves to higher energy as the doping increases, (5) at (π, π), the upper Hubbard band (UHB) carries most of the spectral weight regardless of the filling, and (6) a dip exists in the spectral function at the chemical potential for $n = 0.97$ but is absent for $n = 0.60$. In the underdoped regime, the characteristic width of each $\mathbf{k}$ state is of order t and even much larger near $(\pi, 0)$. Such broad spectral features in the underdoped regime are seen experimentally [18] and

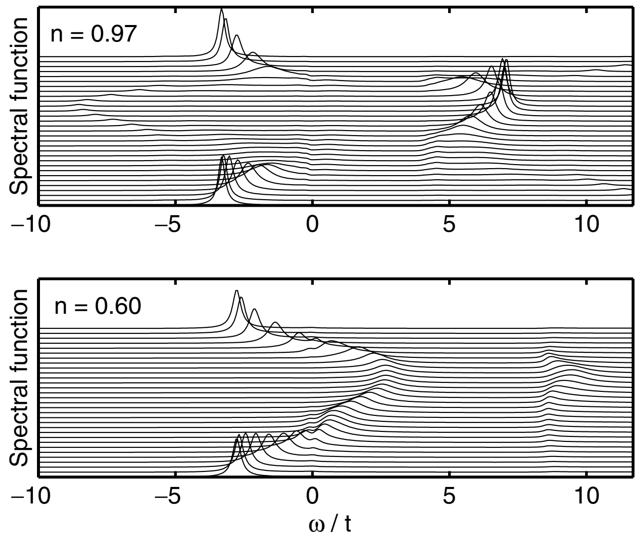

FIG. 2. Electron spectral function for $U = 8t$, $T = 0.07t$, and $n = 0.97$ and 0.60 along a path in the first Brillouin zone from top to bottom: $(k_x, k_y) = (0, 0) \rightarrow (\pi, \pi) \rightarrow (\pi, 0) \rightarrow (0, 0)$.

arise in this context because $\mathrm{Im}\Sigma \neq 0$ as shown in Fig. 1. As a consequence, there is no sharp criterion for unit occupancy of each state in the FBZ. Because the spectral weight at each momentum is unity, however, and each state lives both below and above the chemical potential, the charge carried by the piece of the state lying below the chemical potential is less than unity.

Is the dip in the spectral function shown in Fig. 2 for $n = 0.97$ real? Displayed in Fig. 3 is the DOS for $T = 0.25t$ and $T = 0.07t$ for several fillings. As is evident, no local minimum of DOS exists at the chemical potential at high temperature, $T = 0.25t$. Features which emerge even at high temperature are the reshuffling of spectral weight from above the charge gap to below as the filling is changed and also a movement of the Mott gap to higher energies. Note that even at $n = 0.30$ the Mott gap is still present, though almost all of the spectral weight now resides in the LHB which closely resembles the noninteracting density of states. This is further evidence that we correctly recover Fermi liquid theory as $n \rightarrow 0$. At low temperature, the lower panel of Fig. 3 demonstrates that a pseudogap forms in the DOS for $\delta \approx 0$. The vertical line at 0 indicates that the pseudogap occurs precisely at the chemical potential. Similar qualitative results based on a cluster method have been obtained by Maier *et al.* [22], except their pseudogap is slightly displaced above E_F. In contrast, in the analysis of Haule *et al.* [23], the DOS has a negative slope through E_F (as dictated by the proximity to the Mott gap) but never acquires a local minimum at E_F indicative of a true pseudogap. Because the pseudogap exists below some characteristic temperature and vanishes at higher doping, the result obtained here is nontrivial and highly reminiscent of experimental trends [4]. What is its origin? The inset of Fig. 1 indicates that for a fixed filling, the pseudogap vanishes as U increases and

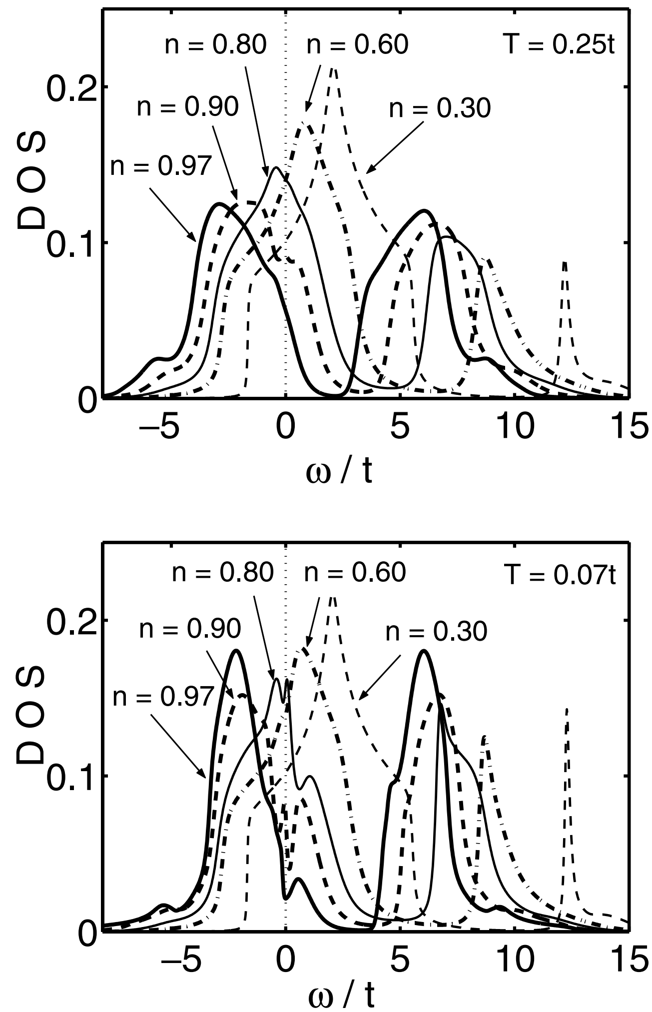

FIG. 3. Density of single particle states for $T = 0.25t$ and $T = 0.07t$, $U = 8t$ for the fillings shown. No pseudogap exists at high temperature. At low T, a pseudogap emerges and remains pinned at the Fermi level but moves above at an intermediate doping level. In the overdoped regime, the pseudogap vanishes entirely and a noninteracting system is recovered.

scales as t^2/U. This suggests that the pseudogap is tied to short-range correlations and hence explains why it is absent in $d = \infty$ [10]. While $J_{\mathrm{eff}} \propto O(t^2/U)$ corresponds to the energy scale for antiferromagnetic spin fluctuations, such fluctuations cannot inhibit hole transport. In fact, Maier *et al.* [22] have shown that even if antiferromagnetism is killed, the pseudogap still persists. Further, we have found that J_{eff} is only weakly doping dependent for $0 < x < 0.25$ and, in fact, vanishes at $x \approx 0.8$. Hence, the resolution of the pseudogap problem lies elsewhere. The energy scale, t^2/U also describes any transport process in which the intermediate state is doubly occupied. Such processes are captured by our approach as a result of the coupling of the two-site cluster to the interacting bath. Consider placing a single hole in a Mott insulator. Unlike a site neighboring the hole, a singly occupied site two

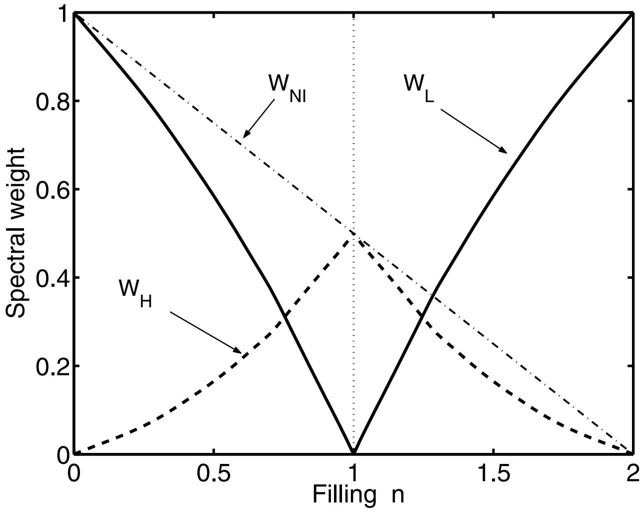

FIG. 4. High (W_H) and low (W_L) spectral weight as a function of filling. $W_{\rm NI}$ is the spectral weight in the noninteracting system.

lattice sites away must temporarily doubly occupy one of its neighbors if it is to move to the hole. The energy for this two-step process is t^2/U. Sites farther away experience an energy barrier with a higher power of t^2/U. Hence, t^2/U is the largest energy barrier for hole transport once a Mott insulator is doped. Because some sites experience no energy barrier, the single-particle density of states exhibits only a suppression, a signal that hole transport involves virtual excitations to the UHB. This pseudogap cannot vanish before each site has on average one hole as its immediate neighbor, roughly $x = 0.25$ for a square lattice. Hence, the pseudogap is of the form $(t^2/U)P(x)$, where $P(x)$ determines the probability that hole transport involves double occupancy and, consequently, is a steadily decreasing function of x.

Additionally, it is precisely two-step (or three-site) hopping that makes the single-particle low-energy spectral weight increase faster [3,24] than $2x$. To show that we recover this result, we compute the high and low spectral weights by integrating the DOS from the energy which minimizes the DOS to ∞ ($-\infty$ for electron doping) and from μ to that fixed energy, respectively. The results shown in Fig. 4 (which have been normalized per spin) demonstrate that the initial spectral weight in the UHB which is $1/2$ at $n = 1$ all moves to low energies as the filling decreases as is observed experimentally [1,2]. The same is true for electron doping ($n > 1$). Further, the curvature of the low-energy spectral weight (LESW) is positive, indicating that the LESW grows faster [3,24] than $2x$. The growth in excess of $2x$ arises entirely from virtual excitations between the LHB and UHB and points to an inseparability of the low- and high-energy scales. Such behavior is absent from a band insulator (see W_{NI} in Fig. 4). That Mottness leads to such a drastic deviation from the noninteracting result is a direct consequence of each state having spectral weight both above and below the chemical potential (see Fig. 2). Our finding that the three-site terms lead to an inseparability of low- and high-energy scales resonates with the recent work of Kirkpatrick and Belitz [25] who have shown that three-body terms are ubiquitous in strongly correlated electron systems and lead to breakdown of a true low-energy description.

Without global symmetry breaking, spin-charge separation, or pairing, we have shown that lightly doped Mott insulators possess a pseudogap which arises entirely from local correlations. The pseudogap is ubiquitous because any singly occupied site not immediately neighboring a hole experiences an energy gap for transport equal to t^2/U. This generic phenomenon offers a simple resolution of the pseudogap problem in the cuprates.

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