PHYSICAL REVIEW B **81**, 235135 (2010)

Nonconservation of fermionic degrees of freedom at low energy in doped Mott insulators

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(Received 25 February 2010; published 29 June 2010)

Hall and optical conductivity experiments on the cuprates indicate that the low-energy fermionic degrees of freedom in a doped Mott insulator possess a component that is dynamically generated and hence determined by the temperature. We show explicitly how the spectrum in the lower Hubbard band should be partitioned to describe such dynamically generated charge degrees of freedom and corroborate this picture with the results from the exact low-energy theory of the Hubbard model. A consequence of such dynamics is that the Landau one-to-one correspondence between bare electrons and the effective fermionic degrees of freedom at low energies breaks down explicitly. This state of affairs obtains because the total hole number is not conserved as it contains a dynamical contribution. We propose that any experimental probe that couples to the low-energy dynamics of a doped Mott insulator, quantum oscillation experiments included, should be interpreted in terms of the total dynamically generated hole number rather than the bare value.

DOI: 10.1103/PhysRevB.81.235135 PACS number(s): 71.27.+a

In 1993, Meinders, Eskes, and Sawatzky[1] concluded based on an exact diagonalization study that because the effective number of low-energy degrees of freedom in a doped Mott insulator is a function of the hybridization and therefore the volume and temperature, "…it is not possible to define a Hamiltonian that describes the low-energy-scale physics unless one accepts an effective nonparticle conservation." Particle nonconservation as used here refers to the fact that the number of low-energy degrees of freedom is not strictly determined by the electron filling or equivalently the doping level but rather by dynamical degrees of freedom generated from the hybridization and hence the temperature. Thus far, an explicit construction demonstrating this has not been advanced. Given the obvious importance of this result, it is surprising how little attention it has attracted. In this paper, we directly address the question of how particle conservation breaks down in a low-energy theory of a doped Mott insulator. We first show that experiments on the Hall[2–5] and optical[6–8] conductivities and general theoretical considerations support this claim. Finally, we propose a simple partitioning of the spectral weight in the lower Hubbard band (LHB) which isolates the explicit hybridization-dependent degrees of freedom that are responsible for the dynamical generation of charge carriers and hence effective particle nonconservation as defined above. We show that these degrees of freedom can be understood within the recently[9–13] derived exact low-energy theory of the Hubbard model.

I. EXPERIMENTAL MOTIVATION

Is there any experimental indication in doped Mott systems that the number of charge carriers is dynamically generated? It would suffice to show that either (1) the carrier density is temperature dependent or (2) the number of charge carriers exceeds the nominal doping level, hereafter referred to as x. Consider first the experiments on the Hall coefficient in $La_{2-x}Sr_xCuO_4$ (LSCO). In the underdoped regime, the inverse Hall number is strongly temperature dependent.[2–4] Gor'kov and Teitel'baum[5] observed that a two-component empirical formula,

$$n_{\mathrm{Hall}}(x,T) = n_0(x) + n_1(x)\exp[-\Delta(x)/T], \tag{1}$$

accurately describes the inverse Hall coefficient in LSCO in the underdoped regime. One of the components is independent of temperature, $n_0(x)$, given by the static doping level, while the other is strongly temperature dependent, $n_1(x)\exp[-\Delta(x)/T]$. The key observation here is that the temperature dependence in n_{Hall} is carried entirely within $\Delta(x,T)$ which defines a characteristic activation energy scale for the system. Gor'kov and Teitel'baum's[5] analysis suggests that the activation energy is set by the pseudogap energy scale. Consequently, the bound component should be liberated beyond the T^* scale for the onset of the pseudogap.

Additionally, optical conductivity experiments indicate that the effective number of charge carriers exceeds the nominal count provided by the doping. In the experiments[7,8] the integrated optical conductivity,

$$N_{\mathrm{eff}}(\Omega) = \frac{2mV_{\mathrm{cell}}}{\pi e^2}\int_0^{\Omega}\sigma(\omega)d\omega \tag{2}$$

is generally plotted in which the cutoff is set by the optical gap, which for LSCO is $\Omega \approx 1.2$ eV. Here $\sigma(\omega)$ is the optical conductivity, V_{cell} the unit-cell volume per formula unit, m the free electron mass, and e the electron charge. In a rigid-band semiconductor model in which spectral weight transfer is absent, $N_{\mathrm{eff}}=x$. However, in all cuprates, regardless of whether they are electron[7] or hole doped,[6,8] N_{eff} exceeds x in the underdoped regime.

Consequently, experimental probes which couple to the current reveal that the number of charge carriers in the cuprates is (1) temperature dependent and (2) exceeds the nominal doping level, consistent with Meinders, *et al.*[1] An interesting question is how does one define the chemical potential for such dynamically generated charge degrees of freedom. Clearly it is not equal to that of the bare electrons as the effective number of charge degrees of freedom exceeds the bare charge count. We argue below that the effective doping level that captures the dynamical generation of the charge degrees of freedom as in Eq. (1) is given by

$$x' = x + \alpha, \tag{3}$$

where α is a dynamical correction determined by the hybridization. This redefinition of the doping level naturally arises from the exact[9–12] low-energy theory of the Hubbard model which has been shown to explain[9,11,13] both Eqs. (1) and (2).

II. REDEFINITION OF CHEMICAL POTENTIAL

The goal in this section is to redefine the chemical potential so that the effective number of fermionic charge carriers is consistent with dynamical generation of charge degrees of freedom discussed in the previous section. In the standard theory of metals, the intensity or spectral weight of a band is completely exhausted by counting the number of electrons it can hold. That is, it is a constant given by one per unit cell and per spin direction. Essential to this view is the robustness of electron quasiparticles even in the presence of interactions. Because of the one-to-one correspondence between electrons and quasiparticles, the chemical potential, μ, can be defined either by counting electrons

$$n = \int_{-\infty}^{\mu} N(\omega) d\omega, \tag{4}$$

or by integrating,

$$y = \int_{\mu}^{\Lambda_g} N(\omega) d\omega, \tag{5}$$

the unoccupied part of the spectrum. Here $N(\omega)$ is the single-particle electron density of states and Λ_g is a cutoff demarcating the low-energy physics. As a result of the electron-quasiparticle correspondence, y is identical to the number of doped holes, x, and the electron filling is given by $n=2-x$ (for a single band).

In stark contrast, the empty part of the spectrum per spin at low energies, Eq. (5), exceeds the doping level in strongly correlated systems such as doped Mott insulators. The inherent problem with strongly correlated systems is that the energy bands are not the traditional static bands that typify band insulator systems. This can be illustrated simply by considering the Hubbard model

$$H_{\text{Hubb}} = -t \sum_{\langle i,j\rangle,\sigma} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum_{i} c_{i\uparrow}^{\dagger} c_{i\downarrow}^{\dagger} c_{i\downarrow} c_{i\uparrow}, \tag{6}$$

in which electrons hop among a set of lattice sites, but pay an energy cost U whenever they doubly occupy the same site. Here i,j label lattice sites, $\langle i,j\rangle$ indicates nearest neighbors, $c_{i\sigma}$ annihilates an electron with spin σ on site i and t is the nearest-neighbor hopping matrix element. When $t=0$, the Hamiltonian is diagonal

$$H_U = U \sum_{i} n_{i\uparrow} n_{i\downarrow} = \frac{U}{2} \sum_{i\sigma} \eta_{i\sigma}^{\dagger} \eta_{i\sigma}, \tag{7}$$

where $\eta_{i\sigma}=c_{i\sigma}n_{i\bar{\sigma}}$ creates the excitations above the gap in the upper Hubbard band (UHB) on sites occupied by a single electron. Its complement, $\xi_{i\sigma}=c_{i\sigma}(1-n_{i\bar{\sigma}})$ creates excitations strictly on empty sites and hence describes particle motion below the gap. Here $\bar{\sigma}=-\sigma$. Consequently, the anticommutator

$$\text{m}_{\text{LHB}}^{0} = \frac{1}{N} \sum_{i,\sigma} \langle \{\xi_{i\sigma}, \xi_{i\sigma}^{\dagger}\} \rangle = 2 - n, \tag{8}$$

determines the spectral weight in the lower Hubbard band (LHB). Since each hole in a half-filled band decreases the single occupancy by one, the weight of the UHB is $1-x$. Because the total weight of the UHB and LHB must be 2, we find that $2-n+1-x=2$ or $n=1-x$ and $\text{m}_{\text{LHB}}^{0}=1+x$ in the atomic limit. The weights $1+x$ and $1-x$ also determine the total ways electrons can occupy each of the bands. Thus, in the atomic limit, electrons alone exhaust the total degrees of freedom of each band. Further, since each hole leaves behind an empty site that can be occupied by either a spin up or a spin down electron, the electron addition spectrum in the LHB has weight $y=2x$.[1,15,16] Hence, the occupied part of the LHB and UHB both have identical weights of $1-x$ in the atomic limit.

Because the operators ξ and η do not diagonalize the hopping term, the total intensity of the LHB

$$\text{m}_{\text{LHB}} = 1 + x + \frac{2t}{U} \sum_{ij\sigma} g_{ij} \langle \tilde{c}_{i\sigma}^{\dagger} \tilde{c}_{j\sigma} \rangle + \cdots = 1 + x + \alpha, \tag{9}$$

has t/U corrections as shown by Harris and Lange.[9] Here $\tilde{c}_{i\sigma}$ are related to the original bare fermion operators via a canonical transformation that brings the Hubbard model into block diagonal form in which the energy of each block is nU. In fact, all orders of perturbation theory[15] increase the intensity of the LHB beyond its atomic limit of $1+x$. It is these dynamical corrections that α denotes. While the intensity of the LHB increases away from the atomic limit, the total number of ways of assigning electrons to the LHB still remains fixed at $1+x$. Consequently, the total weight of the LHB exceeds the fermionic phase space and additional degrees of freedom are needed.

Nonetheless, the sum of the spectral weights in the LHB and UHB must be 2 by charge conservation. Consequently, the weight in the UHB, $\text{m}_{\text{UHB}}=1-x-\alpha$, decreases faster than $1-x$. How should the spectrum in the LHB be partitioned? Harris and Lange[9] did not address this issue possibly because when the total weight of a band exceeds the electron count, the chemical potential for the effective low-energy degrees of freedom is not simply understood within a conventional picture of adding or removing electron quasiparticles. Despite this difficulty, it is common in the strongly correlated community[1,15–17] to assign the spectral weight for the bare electrons assuming the doping level is not renormalized by the dynamics. Hence, the weight below the chemical potential, n, remains[1,15–17] at $1-x$ and the weight immediately above the chemical potential becomes $y=2x+\alpha$ as depicted in Fig. 1(a). On this account,[1,15–17] only the states above the chemical potential acquire doubly occupied character dynamically.

This assignment of the chemical potential is valid for the bare electrons alone and does not include the dynamically generated charges as distinct low-energy entities. To address this problem, consider the Lehmann representation,

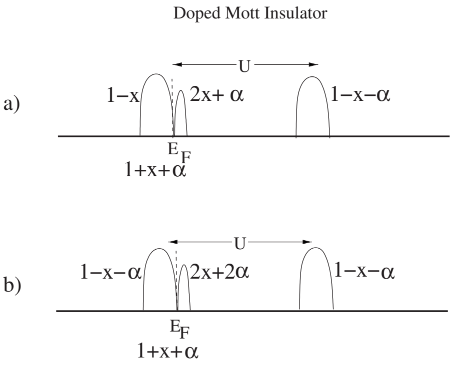

FIG. 1. Redistribution of spectral weight in the Hubbard model upon doping the insulating state with x holes. α is the dynamical correction mediated by the doubly occupied sector. To order t/U, this correction worked out by Harris and Lange.[14] (a) The traditional approach (Refs. 1, 16, and 17) in which the occupied part of the lower band is fixed to the electron filling $1-x$. (b) New assignment of the spectral weight in terms of dynamically generated charge carriers. In this picture, the weight of the empty part of the LHB per spin is the effective doping level, $x'=x+\alpha$.

$$m_{\rm LHB}=\sum_{k,m}\int_{-\infty}^{\mu}d\omega|\langle\psi_m^{N-1}|c_k|\psi_g^N\rangle|^2\delta(\omega-E_m^{N-1}+E_g^N)+\sum_{k,m}\int_{\mu}^{\Lambda_g}d\omega|\langle\psi_m^{N+1}|c_k^\dagger|\psi_g^N\rangle|^2\delta(\omega-E_m^{N+1}+E_g^N) \tag{10}$$

of the spectral function. In these expressions, E_m^N is the mth eigenenergy of the N particle system with ground state E_g^N with associated many-body states $|\psi_m^N\rangle$ and $|\psi_g^N\rangle$, respectively. The filled (first term) and empty (second term) parts of the spectrum do not have traditional definitions in terms of n and $1-n$, respectively, if one insists on setting $n=1-x$. That is, x is not a fundamental property of the spectral function as it is for a rigid-band system such as a band insulator in which it equals the empty part of the spectrum. The disconnect between x and the empty part of the spectrum in the LHB [the second term in Eq. (10)] obtains because holes are generated either by doping or mixing with the doubly occupied sector. This is not an option for a band insulator. As a result, the number of holes is *not* determined strictly by the doping. That is, although n is well defined, $1-n$ does not have any fundamental meaning in terms of the integrated spectral weight of the empty part of the LHB. In fact, the empty part of the spectrum has no obvious relation to anything.

We now show how the spectrum can be partitioned so that the chemical potential accounts for a charge number consistent with Eq. (1). Note we have some degree of freedom in describing the physics in the LHB since it is not a rigid band. If the dynamical contribution can be removed through a redefinition of the chemical potential, then the empty part of the spectrum per spin will be the effective hole number. The justification for this picture is as follows. In a hole-doped system, turning on a finite t/U creates pairs of double occupancies and empty sites (doublon-holon pairs). The weight in the UHB corresponds to adding one electron in the high energy sector, in other words creating double occupancy. Doublon-holon pairs clearly deplete this intensity leading to a loss of spectral in the UHB faster than the atomic limit value of $1-x$. The occupied weight in the LHB corresponds to removing an electron in the low-energy sector. In other words, the occupied part of the spectrum corresponds to removing an electron such that the number of double occupancies remains conserved. Hence the occupied part of the LHB is a measure of single-occupancy whose weight as well must decrease on creation of doublon-holon pairs. In other words, the weights in the occupied part of the LHB and the UHB must be the same, since both provide a measure of the same phase space. Therefore, we propose that the consistent definition of the chemical potential for the low-energy fermionic degrees of freedom can be obtained by demanding that the two weights be equal. Note this says nothing about the nature of the excitations which live in the high-energy scale. Consequently, we arrive at the assignments of the spectral weights in Fig. 1(b). The occupied part of the LHB has weight $(1-x-\alpha)$ and the unoccupied part $2(x+\alpha)$. The fermionic degrees of freedom that are associated with this assignment of the chemical potential reflect the dynamical generation of the charge degrees of freedom. As a result of the dynamics, $x'=x+\alpha$ now denotes the effective number of hole degrees of freedom per spin at low energy. Consequently, we propose that it is with respect to x' that a Luttinger theorem exists not x, the bare hole number.

In the case of electron doping, the chemical potential (μ) lies in the UHB where $2x$ electron removal states are created below μ and the weight above μ is given by $1-x$ in the atomic limit. Turning on a finite t/U creates doublon-holon pairs. In this case, the holes belong to the LHB and represent the high-energy configurations of the system. The weight above μ represents the amplitude for adding an electron to the UHB, or creating a double occupancy, which is depleted upon creation of doublon-holon pairs since neither holons nor doublons can contribute to the creation of double occupancies upon addition of a single electron. This weight is analogous to that of the occupied part of the LHB in the case of hole doping. For charge-transfer systems, such as the cuprates, the same argument applies because of the equivalence[1] with the Hubbard model for realistic values of the hybridization between the bands.

To counter the argument that the dynamical corrections might not affect the physics on all energy scales, it suffices to compute the cross correlator between $\xi_{i\sigma}=c_{i\sigma}(1-n_{i\bar{\sigma}})$ and $\eta_{i\sigma}=c_{i\sigma}n_{i\bar{\sigma}}$. The full electron spectral function, $A(\mathbf{k},\omega)=-\text{Im}\,FT(\theta(t-t')\langle\{c_{i\sigma}(t),c_{j\sigma}^{\dagger}(t')\}\rangle)/\pi=A_{\eta\eta}+A_{\xi\xi}+2A_{\eta\xi}$, contains two diagonal terms $A_{\eta\eta}$ and $A_{\xi\xi}$ and a cross term $A_{\eta\xi}$ which represents the degree to which the high and low energy degrees of freedom are coupled. Here, FT represents the frequency and momentum Fourier transform. We have computed $A_{\eta\xi}$ previously[18] and it is clearly nonzero at all frequencies that bracket the turn-on of the spectral weights in the LHB and UHB at half-filling and at finite doping. This is simply a reflection of the fact that at all frequencies, the states in the LHB all have doubly occupied character. The dynamical contribution reduces the spectral weight. Let us call the reduction q and hence the weight is given by $1-x-q$. The weight in the unoccupied part of the LHB is

$2x+\alpha+q$. For the weight of a hole per spin to be equal to that of an electron, we must have that $q=\alpha$. This results in the assignments in Fig. 1(b).

As a result, the bare electrons and the low-energy dynamically generated fermionic charge carriers in the LHB do not stand in a one-to-one correspondence. The efficient cause of this breakdown is dynamical spectral weight transfer. Insertion of an electron affects the spectrum at all energies while only local changes occur in terms of the low-energy degrees of freedom. Such an orthogonality catastrophe is due entirely to the existence of the UHB[18,19] and persists as long as the degrees of freedom transferred from the UHB provide a relevant perturbation to those in the LHB. In fact, Fig. 1(b) provides a possible basis for the Anderson[19] conjecture that the very existence of the UHB (in the form of dynamical spectral weight transfer) leads to a breakdown of Fermi liquid theory.

An experimental prediction of this work is that α should be temperature dependent. Making contact with Eq. (1), α should turn on at T^*. As a result, the dynamical part of the spectral weight signifies an opening of the pseudogap in the single-particle spectrum as pointed out earlier.[9,13] This is reasonable for two reasons. First, if $\alpha \neq 0$, the number of ways of adding a particle exceeds the number of ways of adding an electron to the empty part of the spectrum in the LHB. That is, some of the particle addition states in the LHB are orthogonal to the addition of an electron. Second, there is no reason to separate the UHB and LHB's if there is no gap between them. Consequently, the collapse of the UHB should be coincident with the closing of the pseudogap. Recently, Peets *et al.*[20] have observed that the UHB collapses once the pseudogap closes, consistent with our prediction here.

III. CONFIRMATION FROM EXACT LOW-ENERGY THEORY

Since the weight of the band in which the chemical potential resides in a doped Mott insulator exceeds the electron count, new degrees of freedom are required in any consistent low-energy theory. The extra degrees of freedom are generated from mixing with the doubly occupied sector and hence should emerge upon integration of the states far away from the chemical potential. We have carried[9–12] out this Wilsonian program exactly for the Hubbard model and showed that a charge $|2e|$ bosonic field emerges. The boson which is nonpropagating has charge $2e$ for hole doping and $-2e$ for electron doping, represents the mixing with double occupancy and double holes, respectively. For hole doping, the conserved charge Q, which equals the total electron filling n,[9–12] is a sum of two components,

$$Q=\sum_{i\sigma} a^\dagger_{i\sigma}a_{i\sigma}+2\sum_i \varphi^\dagger_i\varphi_i, \tag{11}$$

immediately implying that the weight of the fermionic part must be less than the conserved charge. Here $a_{i\sigma}$ is the annihilation operator for the fermionic degree of freedom that results when the high-energy scale is integrated out and φ is a charge $2e$ boson. That Q is the conserved charge can be verified by inspection as it trivially commutes with the low-energy effective Hamiltonian. In fact, Eq. (11) gives a prescription for α, namely, the bosonic charge, if we interpret Q as $1-x$ and the fermionic quasiparticle density as $1-x'$. In this theory,[9–11] the quasiparticles are transformed at low energies to

$$c^\dagger_{i,\sigma} \to (1-n_{i,\bar{\sigma}})c^\dagger_{i,\sigma}+V_\sigma\frac{t}{U}b^\dagger_i c_{i,\bar{\sigma}}+V_\sigma\frac{t}{U}\varphi^\dagger_i c_{i,\bar{\sigma}}, \tag{12}$$

to leading order in t/U upon the integration of the high energy scale. Here $b_i=\Sigma_j b_{ij}=\Sigma_{j\sigma}c_{j\sigma}V_\sigma c_{i\bar{\sigma}}$ with $V_\uparrow=-V_\downarrow=1$ and j a nearest neighbor of site i. The first two terms represent the standard electron operator in the lower Hubbard band dressed with spin fluctuations which constitutes the quasiparticles or the effective fermionic degrees of freedom. However, the last term represents the correction due to dynamical spectral weight transfer. Equation (12) lays plain that an electron at low energy contains a propagating part that arises from the charge $2e$ boson. To illustrate that more than just the fermions are needed to satisfy the $1-x$ sum rule, we computed the pure fermionic part of the spectral function by evaluating the Green function: $\int d\varphi^* d\varphi FT(\int[Dc^*_i Dc_i]c_i(t)c^*_j(0)\exp(-\int L_{\rm IR}dt))/Z$. $L_{\rm IR}$ is the low-energy Lagrangian[9–12] obtained by integrating out the UHB and Z the partition function. We computed this quantity assuming that the boson is spatially homogeneous, which is justified[9–12] since there are no gradient terms of the boson in the low-energy action. The results in Fig. 2 demonstrate that the integrated weight in the occupied part of the spectrum is indeed less than $1-x$. That this weight is less than $1-x$ is independent of the approximations used to calculate the spectral function. This follows entirely from the fact that the conserved charge, Q is a sum of a fermionic and a bosonic part. The deficit from $1-x$ is carried in the $\varphi^\dagger c_{i\bar{\sigma}}$ term. The difference between the solid triangles and the solid line approximates α.

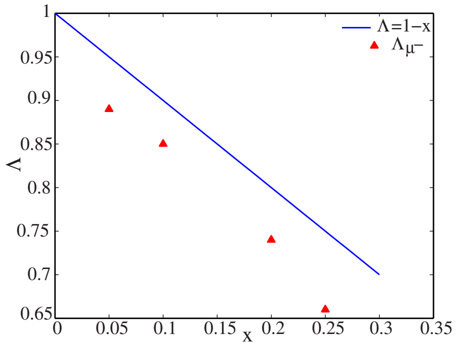

FIG. 2. (Color online) Integrated spectral weight in the occupied part of the lower Hubbard band, Λ_{μ^-}, from the charge $2e$ low-energy theory (Refs. 9–12) with $U/t=8$. Here x is the doping level for the conserved charge, $Q=\Sigma_{i\sigma}c^\dagger_{i\sigma}c_{i\sigma}+2\Sigma_i\varphi^\dagger_i\varphi_i$. Clearly shown is that the occupied part (solid triangles) of the one-particle spectrum has a weight less than $1-x$ (solid blue line).

IV. CONCLUDING REMARKS

Experimentally, any measurement which probes the fermionic low-energy degrees of freedom should be interpreted in terms of the total number of hole degrees of freedom, $x+\alpha$ not x. For example, the superfluid density should exceed x and scale as $x+\alpha$, already confirmed in $\mathrm{YBa_2Cu_3O_{6+x}}$ (Ref. 6) (YBCO). Similarly, Fermi surface volumes, that is the total volume of the hole pockets minus that of the electron pockets, extracted from quantum oscillation experiments,[21] whose origin is still not understood, should be compared with x' not x as the experimental probe is the current. This is particularly germane because the Fermi surface volumes extracted experimentally[21,22] for YBCO are not consistent with any integer multiple of the physically doped holes. Interestingly, the first experiments of this type observed oscillations in the Hall coefficient.[21] Hence, it is perfectly reasonable that the effective doping level should be consistent with the physics that leads to Eq. (1).

Finally, Fermi liquid theory is recovered when the charge $2e$ boson decouples from the electronic spectrum. By decoupling we mean that the UHB collapses and the LHB has a weight of 2. In this limit, there is no true high-energy scale and φ should be an irrelevant degree of freedom. To illustrate, using the appropriate[23] scaling such that the kinetic energy remains constant in the limit $d\rightarrow\infty$, that is, $t\rightarrow O(1/\sqrt{d})$, and averages of the form $\langle c_j^\dagger c_i\rangle \propto 1/\sqrt{d}$ (note as $d\rightarrow\infty$ the scaling of c and $\tilde{c}$ [Eq. (9)] are not trivially related), we find that the boson-dependent terms in the exact low energy theory, $t\Sigma_i\varphi_i^\dagger c_{i\uparrow}c_{i\downarrow}\rightarrow O(1/\sqrt{d})$, $t^2/U\Sigma_i\varphi_i^\dagger\varphi_i\rightarrow O(1/d)$ and $t^2/U\Sigma_{\langle ij\rangle}\varphi_i^\dagger b_{ij}\rightarrow O[d\times(1/\sqrt{d})^3]$, vanish when $d=\infty$. Consequently, no breakdown of Fermi liquid theory obtains as seen numerically[23] for $d=\infty$ and $n\neq 1$. In finite dimensions, the precise value of the coupling constant and doping level at which the bosonic degrees of freedom decouples remains the open problem in Mottness.

ACKNOWLEDGMENTS

This work was funded by the NSF Grant No. DMR-0940992, NSF Grant No. PHY05-51164 (KITP) and Samsung scholarship (S. Hong). We also thank F. Krüger, T. Stanescu, R. Leigh, and G. Sawatzky for critical remarks. In addition, this work is funded in part by the Center for Emergent Superconductivity, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Award No. DE-AC0298CH1088.

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