Density of States of Doped Hubbard Clusters

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Numerical results for the density of states to remove or add electrons to a finite Hubbard cluster are presented for both positive and negative on-site interactions. The behavior of the filling versus the chemical potential is discussed. At half filling there is a gap in the density of states. When the repulsive-U Hubbard model is doped spectral weight is transferred into the gap, and the chemical potential is found to move across the gap as one goes from electron to hole doping. We discuss the relationship of these results to experiment. For the negative-U Hubbard model, the chemical potential shifts continuously and lies in the middle of the superconducting gap.

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It is widely agreed that the insulating antiferromagnetic state of the high-temperature superconducting Cu-O materials is characterized by a charge-transfer gap [1]. However, the nature of the metallic state which arises from the doping of the insulating state remains a puzzle. On the one hand, photoemission spectroscopy (PES) and inverse photoemission spectroscopy (IPES) indicate that as the insulating state is doped, the gap region appears to be gradually filled in, with spectral weight transferred from both the lower valence band and the upper conduction bands of the undoped insulator [2,3]. Furthermore, the chemical potential μ of the doped metal was found to lie in the insulating gap, with μ roughly the same, relative to the valence- and conduction-band peaks, for *both* the electron- and hole-doped materials. That is, as the antiferromagnetic insulating state is doped, the chemical potential does *not* move across the gap if the doping is changed from holes to electrons [2]. Alternatively, oxygen soft-x-ray absorption spectra measured on $La_{2-x}Sr_xCuO_4$, using fluorescence-yield detection, have been interpreted in terms of a picture in which hole doping introduces carriers into the lower band [4]. Here the observed two-peak absorption structure is identified as an upper peak arising from transitions into the upper band, renormalized by core-hole excitonic correlations, while the lower peak is associated with holes doped into the top edge of the lower band. Similar experiments have been carried out [5] for electron-doped $Nd_{2-x}Ce_xCuO_4$. In this picture, the chemical potential would be expected to move across the gap when the doping is changed from holes to electrons, at variance with PES experiments [2].

Here, using Lanczos results, we examine the behavior of the chemical potential μ with doping and the structure of the spectral weight for the one-band Hubbard model. Results for both the positive-U and negative-U Hubbard models will be contrasted, and the relationship of the positive-U Hubbard model results to experiment will be discussed. We will use a particle-hole symmetric form for the interaction so that

$$H=-t\sum_{\langle ij\rangle s}(c_{is}^{\dagger}c_{js}+c_{js}^{\dagger}c_{is})+U\sum_{i}(n_{i\uparrow}-\tfrac{1}{2})(n_{i\downarrow}-\tfrac{1}{2})-\mu\sum_{is}(n_{is}-\tfrac{1}{2}), \quad (1)$$

with the chemical potential $\mu=0$ at half filling. The operator $c_{is}^{\dagger}$ creates an electron of spin s on the ith site, t is the one-electron transfer-matrix element which leads to a bandwidth $8t$, and U is the on-site Coulomb interaction. Using standard Lanczos techniques [6], we have obtained results for the chemical potentials and the spectral weight on finite clusters for positive and negative values of U.

Figure 1(a) shows the average site occupation $\langle n\rangle$ versus the chemical potential μ for a $\sqrt{10}\times\sqrt{10}$ lattice [7] with $U/t=8$ and 20. The steplike structure reflects the finite size of the cluster. As U increases, the shift in μ required to change $\langle n\rangle$ from one at half filling also increases. A similar behavior is found from Lanczos results on 4×4 clusters [8] and Monte Carlo results [9] on up to 8×8 clusters as shown in Figs. 1(b) and 1(c). The agreement between the different lattice sizes and techniques is remarkable, showing that finite-size effects are small. Thus, at zero temperature, we observe that a *finite* negative or positive shift of μ (approximately half the gap) is required to add holes or electrons to a half-filled Hubbard model. Once these values are exceeded, $\langle n\rangle$ appears to vary continuously with μ as the cluster size is scaled up, showing the absence of phase separation in this model [9].

By contrast, for the negative-U Hubbard model, small incremental reductions in μ from 0 produce a series of states in which successive numbers of electron pairs are removed. The variation of $\langle n\rangle$ vs μ shown in Fig. 1(d) and Monte Carlo simulations on up to 8×8 lattices suggest that $\partial\langle n\rangle/\partial\mu$ is nonvanishing for the infinite system at half filling, while for the positive-U model it is zero as shown in Figs. 1(a)–1(c). This is equivalent [10], after a

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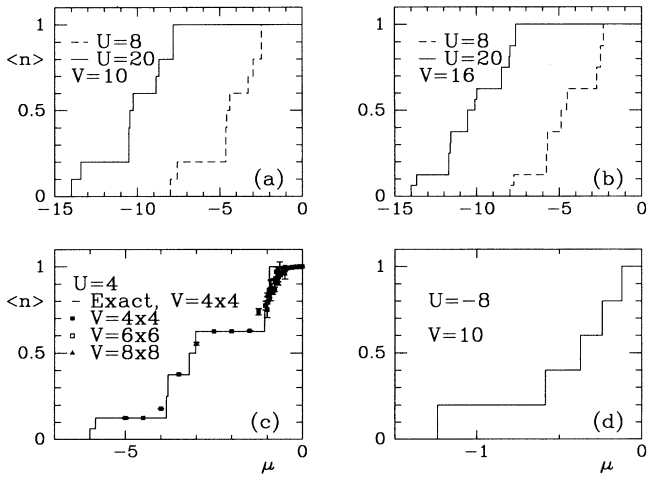

FIG. 1. (a) $\langle n\rangle$ vs μ at zero temperature for the ten-site Hubbard cluster with $U/t=20$ (continuous line) and $U/t=8$ (dashed line) using Lanczos techniques; (b) same as (a) for a sixteen-site cluster; (c) $\langle n\rangle$ vs μ at $U=4$ ($t=1$). The continuous line is the Lanczos result for the sixteen-site cluster. Solid squares denote Monte Carlo results on a 4×4 lattice at temperature $T=t/12$, while the open squares and the solid triangles are Monte Carlo results for the 6×6 and 8×8 clusters, respectively, with $T=t/8$; (d) $\langle n\rangle$ vs μ at zero temperature for the ten-site Hubbard cluster with $U/t=-8$ using Lanczos techniques.

particle-hole transformation, to the statement that the magnetic spin susceptibility of the positive-U Hubbard model at half filling remains finite as the temperature T goes to zero. Likewise the result that $\partial\langle n\rangle/\partial\mu|_{\mu=0}$ vanishes for the positive-U Hubbard model simply reflects the vanishing of the magnetic spin susceptibility of the negative-U Hubbard model. Thus, upon doping, the chemical potential of the negative-U Hubbard model will slowly move away from its half-filled $\mu=0$ value in agreement with the numerical results.

In order to obtain information regarding the distribution of spectral weight, we have calculated

$$N_s^{(+)}(\omega)=\frac{1}{V}\sum_{\mathbf{k},n}|\langle\psi_n^{N+1}|c_{\mathbf{k}s}^{\dagger}|\psi_0^N\rangle|^2\delta(\omega-(E_n^{N+1}-E_0^N)), \tag{2}$$

$$N_s^{(-)}(\omega)=\frac{1}{V}\sum_{\mathbf{k},n}|\langle\psi_n^{N-1}|c_{\mathbf{k}s}|\psi_0^N\rangle|^2\delta(\omega+(E_n^{N-1}-E_0^N)). \tag{3}$$

Here $N_s^{(+)}(\omega)$ is the density of states for adding an electron with spin s and energy ω and $N_s^{(-)}(\omega)$ is the density of states for removing an electron with spin s from a ground state with N electrons. V denotes the number of lattice sites, and the operator $c_{\mathbf{k}s}^{\dagger}=(1/\sqrt{V})\sum_{\mathbf{j}}e^{i\mathbf{k}\cdot\mathbf{j}}c_{\mathbf{j},s}^{\dagger}$ creates a state with momentum $\mathbf{k}$ and spin s. ψ_0^N is the ground state of the subspace with N electrons and E_0^N is its energy. $\{\psi_n^{N\pm1}\}$ denote states in the subspace with $N\pm1$ electrons and energies $\{E_n^{N\pm1}\}$. Figure 2 shows $N_{\uparrow}^{(+)}(\omega)$ (solid lines) and $N_{\uparrow}^{(-)}(\omega)$ (dashed lines) versus ω for $U/t=20$. The chemical potential μ (obtained in Fig. 1) is shown as a vertical line and moves from zero at half filling (ten electrons), Fig. 2(a), to the top edge of the lower Hubbard band, Fig. 2(b), when two electrons are removed (results for $s=\downarrow$ are identical to Fig. 2). Comparing Figs. 2(a) and 2(b), we see that the effects of

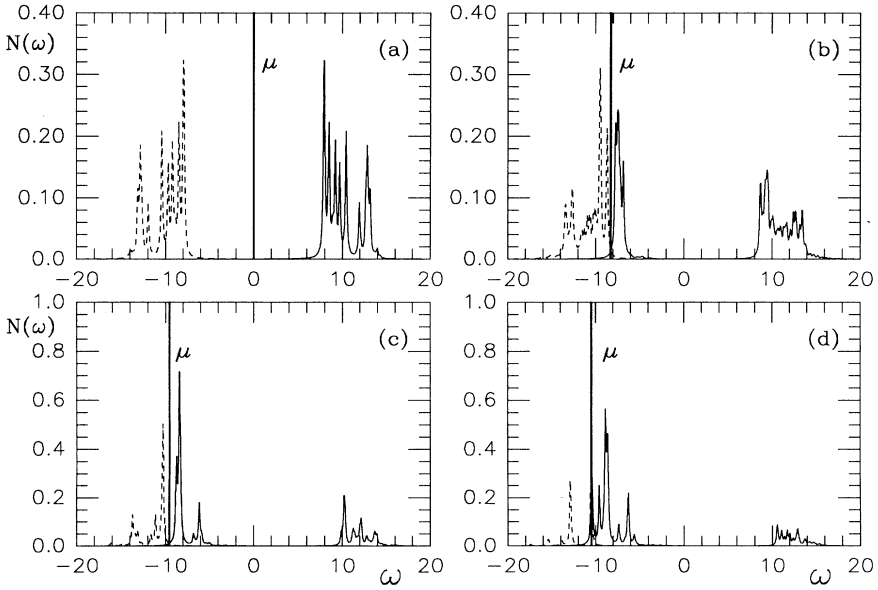

FIG. 2. Electronic spectral density of the Hubbard model $[N(\omega)=N_{\uparrow}^{(\pm)}(\omega)]$ on a $\sqrt{10}\times\sqrt{10}$ cluster. The solid lines correspond to $N_{\uparrow}^{(+)}(\omega)$, while the dashed lines denote $N_{\uparrow}^{(-)}(\omega)$. The hole doping $x=1-N/V$. (a) Results at half filling, corresponding to $x=0$; (b) at $x=0.2$; (c) at $x=0.4$; and (d) at $x=0.6$. The solid thick vertical line indicates the position of μ taken from Figs. 1(a)–1(c) (for these plots the mean value of μ in each plateau of Fig. 1 was used).

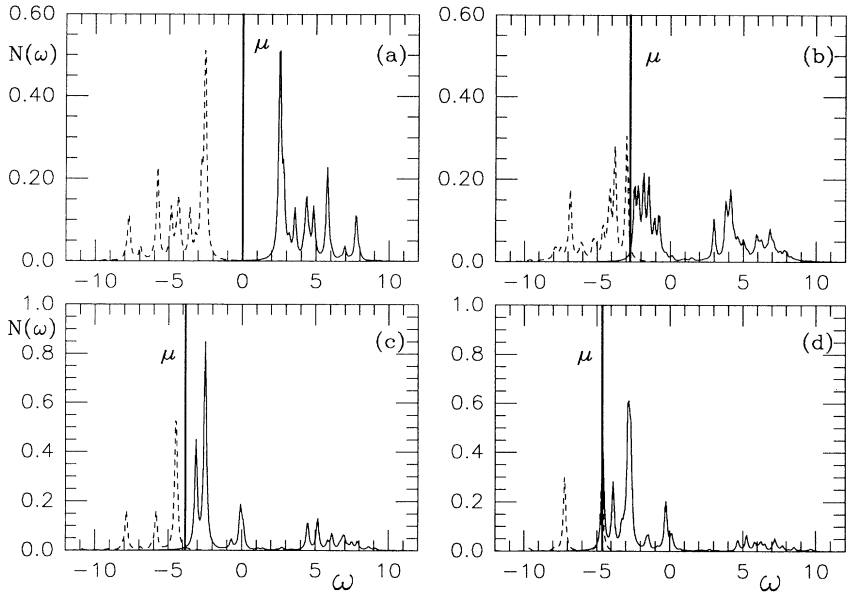

FIG. 3. Same as Fig. 2 with $U/t=8$.

hole doping are to remove spectral weight from both the upper and lower Hubbard bands and to create additional density of states in the gap beginning at the edge of the lower Hubbard band. Results for further dopings are shown in Figs. 2(c) and 2(d). With this additional doping, the chemical potential moves slowly down from its value with eight electrons, and further spectral weight is removed from both the upper and lower Hubbard bands, creating states in the gap region above the lower Hubbard band [11].

Similar results for $U/t=8$ are shown in Fig. 3. In this case, U is equal to the bandwidth $8t$, and the spectral weight created in the gap region by doping fills the gap (although with small spectral weight), as shown in Fig. 3(b) (for $U/t=4$ and $x=0.2$ the gap is filled with an appreciable amount of spectral weight). Just as for $U/t=20$, the chemical potential initially shifts from zero down to the upper edge of the lower Hubbard band when holes are added. Because of particle-hole symmetry, the results for adding electrons to the half-filled cluster are given by simply reflecting these figures about the $\omega=0$ axis. In this case the solid curves represent $N^{(-)}(\omega)$ and the dashed curves $N^{(+)}(\omega)$.

The spectral weight of $N_{\uparrow}^{(-)}(\omega)$ is *exactly* $(1-x)/2$ (where x is the hole doping, i.e., $x=1-N/V$). The reason is that $\int_{-\infty}^{+\infty}d\omega N_{\uparrow}^{(-)}(\omega)$ is equal to the number of particles in the state ψ_0^N with spin $\uparrow$ per site. Since the sum rule states that $\int_{-\infty}^{+\infty}d\omega[N_{\uparrow}^{(-)}(\omega)+N_{\uparrow}^{(+)}(\omega)]=1$, the spectral weight in the IPES region of the spectrum is also identically equal to $(1+x)/2$. In addition, we observed that for large U, the spectral weight per spin of the new states created in the gap (I_{new}) increases approximately linearly with x. The slope of the curve I_{new} vs x converges to one as U/t is increased in agreement with predictions in the atomic limit [12]. For finite U/t the slope is *larger* than one (at small x) and increases when U/t is reduced, at least for U/t large where a genuine gap exists in the IPES spectrum.

Thus we find, as seen in PES experiments [2], that doping a half-filled Hubbard cluster does not simply produce a rigid shift of the density of states relative to the Fermi level, but rather, new states are created in the gap. However, in apparent contrast to the same PES experiments [2] and in agreement with x-ray-absorption experiments [4,5], the chemical potential for the Hubbard model moves across the gap when the doping is changed from holes to electrons, lying near the top of the lower Hubbard band when holes are added and near the lower part of the upper Hubbard band when electrons are added.

It is interesting to compare these results with the corresponding density of states for the negative-U Hubbard model. Figure 4 shows $N_{\uparrow}^{(\pm)}(\omega)$ vs ω for the same set of dopings with $U/t=-8$ (results for $U/t=-4$ and -20 are qualitatively similar). Monte Carlo simulations [13] of the negative-U Hubbard model support a simple phase diagram at zero temperature in which the doped system is in a superconducting state with long-range order for all values of $U/t<0$ and $x\neq 0$ (while at half filling the superconducting state is degenerate with a charge-density-wave state).

At half filling [Fig. 4(a)], $N_{\uparrow}^{(\pm)}(\omega)$ is the same negative and positive U [10], but for negative U the gap in the electronic density of states reflects the underlying pairing and charge-density-wave order. Now, as previously shown in Fig. 1(d), when the negative-U model is doped away from half filling, μ moves continuously away from 0. This behavior is clearly seen in Figs. 4(b)–4(d). The gap follows μ as we expect for a superconductor. As electrons are removed, spectral weight is redistributed from the lower band $(1-x)/2$ to the upper band $(1+x)/2$, but

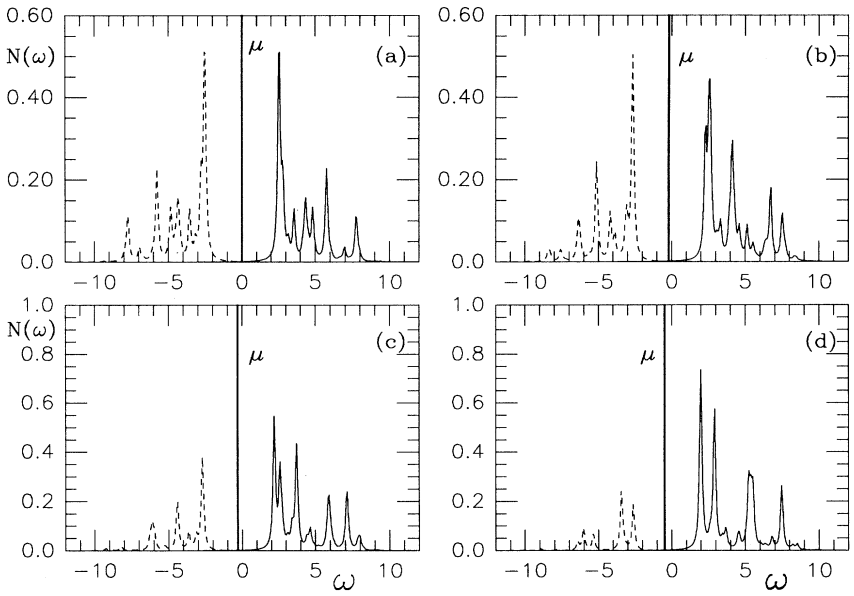

FIG. 4. Same as Fig. 2 with $U/t=-8$.

the gap remains.

It is difficult to know in detail how these results will extrapolate to the bulk limit. It seems likely that the redistribution of spectral weight from the upper and lower bands to form states in the gap is a general feature of a strongly interacting system [14]. In addition, Monte Carlo simulations on a 4×4 CuO_2 three-band Hubbard model [15] and cluster and slave-boson calculations [16] show the same $\langle n\rangle$ vs μ behavior as observed in Figs. 1(a)–1(c) when the gap is a charge-transfer gap. Thus, we believe that the jump of μ from the top of the lower band to the bottom of the upper band as holes or electrons are added to a half-filled band is a general property of strongly correlated electronic models. This disagreement with PES experiments [2] could mean that the n-type materials are actually producing hole doping on the Cu-O planes or that Hubbard-like models are missing some essential feature needed to describe properly the physics of the new superconductors.

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