Two Hubbard Bands: Weight Transfer in Optical and One-Particle Spectra

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We identify local kinetic-energy contributions which are responsible for the rapid weight transfer from the high- to the low-energy scale observed upon doping in strongly correlated materials. Using strong-coupling perturbation theory for the Hubbard model we derive expressions for the weights of the upper and lower Hubbard bands separately and evaluate them explicitly in one dimension. The optical conductivity is found to be a direct probe of the local spin order.

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The high-energy excitation spectra of the Mott-Hubbard (charge-transfer) insulators show a dramatic dependence upon doping. Perhaps the best known examples are the cuprate materials, where the weight around 2–3 eV in the optical conductivity disappears very rapidly [1,2] as soon as holes or electrons are doped to an insulating La_2CuO_4, and reappears in the low-infrared and midinfrared region, with a roughly constant total sum rule. In contrast to these observations, in a conventional semiconductor only a minor reduction of the intensity would occur due to the reduced number of interband transitions.

A similar phenomenon is found in x-ray absorption and electron energy-loss experiments on the oxygen $1s$ edge. There an electron is promoted to the empty O $2p$ states, and therefore this experiment is closely related to the one-particle electron addition spectrum. As holes are introduced in the insulator one observes a valence-band prepeak as expected. However, at the same time the upper band quickly loses its weight [3–5]. Similar effects are observed in Li-doped NiO [6], suggesting that this is a general feature of doped Mott insulators.

It is well known that the strongly correlated multiband models describing the charge-transfer insulators can be mapped on an effective single-band Hubbard model which reproduces the relevant states near the Fermi energy [7]. Numerous numerical diagonalization studies have shown that it indeed describes qualitatively or even quantitatively the features observed in both experiments [4,5,8]. Therefore, the complete theoretical understanding of these rapid changes of spectral weight in the Hubbard model is of fundamental importance.

In this Letter we derive sum rules for the lower (LHB) and upper (UHB) Hubbard bands *separately*. We explain the physical origin of the observed weight changes and show that the fast disappearance of the UHB has a different interpretation for the optical and for the one-particle spectrum. Our results explain why the t-J model does not represent the spectral properties of the large-U Hubbard model, except precisely at half filling.

We start from the Hubbard Hamiltonian with $t \ll U$,

$$H = V + T = U \sum_i n_{i,\uparrow} n_{i,\downarrow} - t \sum_{i,\delta,\sigma} a^\dagger_{i,\sigma} a_{i+\delta,\sigma}, \qquad (1)$$

where $n_{i,\sigma} = a^\dagger_{i,\sigma} a_{i,\sigma}$, $i = 1, \ldots, N_a$ is the site index, σ the spin, and δ labels the nearest neighbors of site i. Sum rules are derived by identifying the Hubbard bands to which each particular eigenstate of H belongs. This can be achieved by a canonical transformation to new fermions, $c_{i,\sigma} = e^{-S} a_{i,\sigma} e^{S}$, which satisfies the requirement that the number of doubly occupied sites (in $c_{i,\sigma}$) is conserved [9]. This transformation is well known and its first-order term (in t/U) transforms the Hubbard Hamiltonian into what we will call the strong-coupling Hamiltonian (t-J model plus three-site hopping).

Following Harris and Lange [9] the creation operator is decomposed into a part which conserves the number of doubly occupied sites, and a part which increases it by one, $c^\dagger_{i,\sigma} = c^\dagger_{i,\sigma;0} + c^\dagger_{i,\sigma;U}$, where

$$c^\dagger_{i,\sigma;0} = c^\dagger_{i,\sigma}(1 - \tilde{n}_{i,\bar{\sigma}}), \qquad c^\dagger_{i,\sigma;U} = c^\dagger_{i,\sigma} \tilde{n}_{i,\bar{\sigma}}, \qquad (2)$$

and $\tilde{n}_{i,\bar{\sigma}} = c^\dagger_{i,\bar{\sigma}} c_{i,\bar{\sigma}}$. This can be used to decompose *any* operator. For instance, the kinetic energy of the new fermions becomes $\tilde{T} = \tilde{T}_0 + \tilde{T}_U + \tilde{T}_{-U}$, with

$$\begin{aligned} \tilde{T}_0 &= -t \sum_{i,\delta,\sigma} [(1 - \tilde{n}_{i,\bar{\sigma}}) c^\dagger_{i,\sigma} c_{i+\delta,\sigma} (1 - \tilde{n}_{i+\delta,\bar{\sigma}}) \\ &\qquad + \tilde{n}_{i,\bar{\sigma}} c^\dagger_{i,\sigma} c_{i+\delta,\sigma} \tilde{n}_{i+\delta,\bar{\sigma}}], \\ \tilde{T}_U &= -t \sum_{i,\delta,\sigma} \tilde{n}_{i,\bar{\sigma}} c^\dagger_{i,\sigma} c_{i+\delta,\sigma} (1 - \tilde{n}_{i+\delta,\bar{\sigma}}), \end{aligned} \qquad (3)$$

and $\tilde{T}_{-U} = \tilde{T}^\dagger_U$. The transformation S can be expressed by commutators of the terms in $\tilde{T}$ [9,10]. To second order, $S = (\tilde{T}_U - \tilde{T}_{-U})/U + [\tilde{T}_U + \tilde{T}_{-U}, \tilde{T}_0]/U^2$.

The above transformation enables us to derive sum-rule expressions for the optical conductivity for the UHB and LHB separately. Consider an electric field in the x direction, and $R_{i,x}$ is the x component of the position vector of atom i. The corresponding paramagnetic current [11,12] is the time derivative of the polarization

$P_x = \sum_{i,\sigma} R_{i,x} n_{i,\sigma}$, $j_x = i[H, P_x] = it \sum_{i,\delta,\sigma} \delta_x a^\dagger_{i+\delta,\sigma} a_{i,\sigma}$. Using linear response the real part of the optical conductivity per site for the UHB is

$$\sigma_x^{\mathrm{UHB}}(\omega) = \frac{1}{\omega N_a} \sum_{f \in \mathrm{UHB}} |\langle f, N | j_x | 0, N \rangle|^2 \delta(\omega - E_f + E_0), \quad (4)$$

where $|0, N\rangle$ denotes the N-particle ground state and $|f, N\rangle$ is the set of final states belonging to the UHB. We can assure that only UHB states are reached by restricting j_x to $j_{x;U}$, which creates one doubly occupied site. Then the summation can be extended to the complete set of final states $|f, N\rangle$. Replacing one of the j_x's [13] by $i[H, P_x]$, integrating over ω, transforming to $c_{i,\sigma}$ fermions, and using (2) one finds the sum rule for the UHB to order t^2/U [14],

$$W^{\mathrm{UHB}} = \frac{i}{2N_a} \langle j_{x;-U} P_{x;U} - P_{x;-U} j_{x;U} \rangle = -\frac{2t^2}{UN_a} \sum_i \Big[2 \Big\langle \tilde{\vec{S}}_i \cdot \tilde{\vec{S}}_{i+\hat{x}} - \frac{1}{4} \tilde{n}_i \tilde{n}_{i+\hat{x}} \Big\rangle + \sum_\sigma \Big\langle c^\dagger_{i-\hat{x},\sigma} \tilde{n}_{i\bar{\sigma}} c_{i+\hat{x},\sigma} - c^\dagger_{i-\hat{x},\sigma} c^\dagger_{i\bar{\sigma}} c_{i\sigma} c_{i+\hat{x},\bar{\sigma}} \Big\rangle \Big]. \quad (5)$$

Here $\hat{x}$ is the unit vector in the x direction and $\langle \cdots \rangle$ denotes the expectation value in the ground state of the strong-coupling Hamiltonian.

The total sum rule W is proportional to the kinetic energy [11–13]. Transforming to $c_{i,\sigma}$ fermions we find

$$W = -\frac{1}{zN_a} \langle T \rangle = -\frac{1}{zN_a} \langle \tilde{T} \rangle + \frac{2t^2}{zUN_a} \times \sum_{i,\delta,\delta',\sigma} \Big\langle c^\dagger_{i+\delta,\sigma} \tilde{n}_{i\bar{\sigma}} c_{i+\delta',\sigma} - c^\dagger_{i+\delta,\sigma} c^\dagger_{i\bar{\sigma}} c_{i\sigma} c_{i+\delta',\bar{\sigma}} \Big\rangle, \quad (6)$$

with z being the number of nearest neighbors. The sum rule for the LHB to order t^2/U is simply the difference of the above expressions [15].

The different bands are related to different parts (forward, backward, and sideward) of the three-site hopping term of order t^2/U in the kinetic energy [16], as shown in Table I. The sum rule for the LHB does not contain the exchange part, and therefore its weight vanishes at half filling. It is easy to understand why the sum rule of the UHB (5) contains the spin-spin correlation function. The current transfers an electron to a neighboring site. In order to reach the UHB (i.e., create a doubly occupied site) first of all there has to be an electron present, and second the electrons have to form a singlet. Both conditions are included in the spin-spin correlation function. This also explains why the UHB loses its weight so rapidly upon doping. If one introduces a hole in the half-filled spin system, two spin-spin bonds are removed in the x direction, reducing $W^{\mathrm{UHB}}(x)$ to $(1 - 2x)W^{\mathrm{UHB}}(0)$, where $x = 1 - n$ is the doping percentage. Apart from this static weight transfer, the dynamics due to three-site hopping to neighbors at a distance 2 *reduces* $W^{\mathrm{UHB}}(x)$ down to $(1 - 3x)W^{\mathrm{UHB}}(0)$ in one dimension (1D) (see Table I and below). In more than 1D the holes weaken the spin order and this reduces the UHB even more.

TABLE I. Contributions of the different three-site hopping terms to the total conductivity and the optical intensity of the two Hubbard bands. $\vec{\delta}$ and $\vec{\delta}'$ point to the nearest neighbors of a common site i.

Hopping term	Total	LHB	UHB
$\vec{\delta} = \vec{\delta}'$ or $(\vec{S}_i \cdot \vec{S}_{i+\delta} - n_i n_{i+\delta}/4)$	1	0	1
$\vec{\delta} \perp \vec{\delta}'$	1	1	0
$\vec{\delta} = -\vec{\delta}'$	1	2	−1

In 1D one may evaluate W^{UHB} (5) and W (6) analytically using the Bethe-ansatz wave function which for large U reduces [17] to a product of a spinless fermion and a Heisenberg model spin wave function,

$$W^{\mathrm{UHB;1D}} = 4(\ln 2)\frac{t^2}{U}\left(n^2 + n\frac{\sin(2\pi n)}{2\pi} - 2\frac{\sin^2(\pi n)}{\pi^2}\right),$$

$$W^{\mathrm{1D}} = t\frac{\sin(\pi n)}{\pi} + 4(\ln 2)\frac{t^2}{U}\left(n^2 - n\frac{\sin(2\pi n)}{2\pi}\right). \quad (7)$$

The spectral weights for the infinite chain are plotted in Fig. 1 for $U/t = 10$ and compared with numerical data for a ring of 10 sites. If U is decreased, the weight around half filling ($n = 1$) increases relative to the quarter-filled case, and the total weight for U of the order of the bandwidth becomes almost independent of the density n close to half filling. These qualitative features are described well by Eq. (7), but quantitative deviations from the numerical data become larger due to the neglect of higher-order terms. These higher-order terms even enhance the weight transfer from the UHB to the LHB.

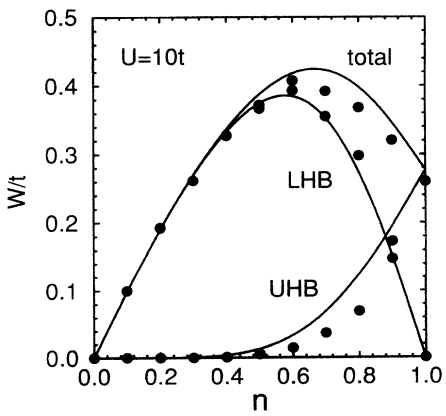

FIG. 1. The integrated conductivities: total, for the upper (UHB) and lower (LHB) Hubbard bands versus occupation n in 1D. The lines are the perturbation theory results (7) and the dots are numerical results for a 10-site ring.

The zero-frequency Drude weight D in 1D can be expressed in terms of the charge velocity and the second derivative of the ground-state energy with respect to the density [18]. Using published large-U Bethe-ansatz expressions for the velocity [19] and energy [17], and comparing with Eq. (7) we find $W^{\mathrm{LHB;1D}} = D + \mathcal{O}(t^3/U^2)$. So up to second order [20] all the weight in the LHB goes into the Drude peak. This is a direct consequence of the spin-charge decoupling for large U, in contrast to the two-dimensional case where the motion of the holes disrupts the spin system resulting in a large finite-frequency optical response in the LHB. The spin-charge decoupling in 1D involves *all* states in the LHB, but hardly affects the states in the UHB. As long as the spin correlations are antiferromagnetic one can always make excitations to the UHB, independent of dimension.

As shown by Eq. (5), the weight of the UHB in optical conductivity effectively measures $\vec{S}_i \cdot \vec{S}_{i+\delta}$. Therefore, the doping dependence of this intensity contains information about the influence of the holes on the local spin order. If, for instance, the holes form ferromagnetic polarons this will lead to an extremely rapid disappearance of the UHB with increasing x, depending on the size of the polarons. Using the ground-state energy of the 1D Heisenberg model as a function of the magnetization [21], we calculated the weight of the UHB as a function of the magnetic field (Fig. 2). The surprising result is that *a small magnetic field* $\sim J$ *causes a complete disappearance of the conductivity at a frequency* $\sim U$, a few eV. Experimentally the above effect should be measurable by optical spectroscopy for substances with exchange couplings $J = 4t^2/U \simeq 10$ K. Unfortunately, the intensity of the upper band will be small when J is small [see Eq. (5)].

The local (site independent) one-particle spectrum is the sum of electron removal and addition,

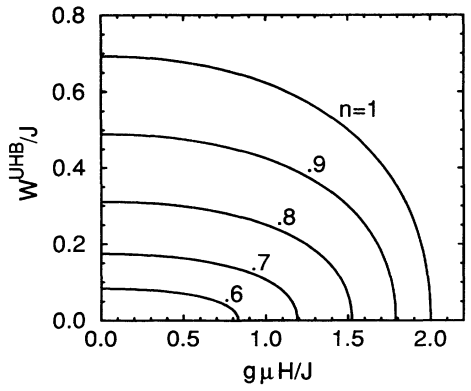

FIG. 2. Spectral weight of the upper Hubbard band in units of $J = 4t^2/U$ as a function of the applied magnetic field $g\mu H/J$ for different fillings n in 1D.

$$A(\omega) = \sum_{f,\sigma} \left|\langle f, N+1 \left| a_{i,\sigma}^{\dagger} \right| 0, N\rangle\right|^2 \delta\left(\omega - E_f^{N+1} + E_0 N\right) + \sum_{f,\sigma} \left|\langle f, N-1 \left| a_{i,\sigma} \right| 0, N\rangle\right|^2 \times \delta\left(\omega - E_0^N + E_f^{N-1}\right). \tag{8}$$

The total sum rule or zeroth moment is $m^{(0)} = \sum_\sigma \langle\{a_{i,\sigma}, a_{i,\sigma}^{\dagger}\}\rangle = 2$. To second order in t/U this is the sum of the weights of the UHB and LHB. As before, if one restricts $a_{i,\sigma}^{\dagger}$ to the part which does not create a doubly occupied site for the $c_{i,\sigma}^{\dagger}$ fermions, then only states in the LHB are reached. The zeroth moment of the LHB is therefore $m_0^{(0)} = \sum_\sigma \langle\{a_{i,\sigma;0}, a_{i,\sigma;0}^{\dagger}\}\rangle$. In analogy with the calculation of the sum rules for the optical conductivity one finds for the electron addition or inverse photoemission (IPES) part of the LHB ($x = 1 - n$),

$$m_0^{(0),\mathrm{IPES}} = m_0^{(0)} - n = 2x + 2\frac{t}{U}\frac{1}{N_a}\sum_{i,\delta,\sigma}\langle c_{i,\sigma}^{\dagger} c_{i+\delta,\sigma}\rangle + \left(\frac{t}{U}\right)^2 \frac{1}{N_a}\sum_{i,\delta,\delta',\sigma;\delta'\neq\delta}\Big\langle -3c_{i+\delta,\sigma}^{\dagger} c_{i+\delta',\sigma} + 6\left(c_{i+\delta,\sigma}^{\dagger}\tilde{n}_{i,\bar{\sigma}} c_{i+\delta',\sigma} - c_{i+\delta,\sigma}^{\dagger} c_{i,\bar{\sigma}}^{\dagger} c_{i,\sigma} c_{i+\delta',\bar{\sigma}}\right)\Big\rangle. \tag{9}$$

The first-order term [22] was derived before by Harris and Lange [9]. Again the expectation values have to be calculated using the strong-coupling model.

The factor $2x$ can be readily understood by state counting: At $U = \infty$ one recovers two low-energy states for every electron removed [5,9]. Equation (9) shows that both the first- and second-order terms ($\vec{\delta} \neq \vec{\delta}'$) are kinetic and *increase* the weight of the LHB for small x. This enhancement can be understood as a positive interference between the ground state and low-energy final states.

In 1D the Ogata-Shiba wave function [17] gives

$$m_0^{(0),\mathrm{IPES,1D}} = 2x + \frac{4t}{\pi U}\sin(\pi x) + 6\left(\frac{t}{U}\right)^2\left\{\frac{\sin(2\pi x)}{2\pi} + 2(\ln 2)\left((1-x)\frac{\sin(2\pi x)}{2\pi} + \frac{\sin^2(\pi x)}{\pi^2}\right)\right\}. \tag{10}$$

The first-order term is just the kinetic energy of spinless fermions and leaves the spins unchanged, while the second-order term involves the spin orientations as well. In Fig. 3 we plot the first- and second-order terms in Eq. (10) and compare them with numerical results for a 10-site ring [5]. The first-order term is symmetric around $n = 0.5$. The second-order term enhances the weight transfer for small doping. For $U = 5$, which is of the order of the bandwidth, the perturbation results deviate from the numerical data due to higher-order corrections.

To conclude, using large-U perturbation theory we derived sum rules for the individual Hubbard bands in the optical and one-particle spectra. To obtain a correct description it was essential to transform the operators,

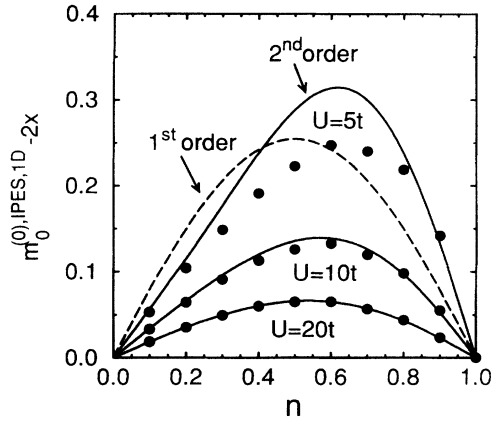

FIG. 3. Electron-addition intensity in the lower Hubbard band as a function of n in 1D. The static part ($= 2x$) is subtracted. The lines and the dots stand for the second-order (10) and numerical results for a 10-site ring, respectively. The dashed line shows the first-order result at $U = 5t$.

such as the current operator for the optical conductivity. The first-order corrections give rise to large intensity changes in general and reasonably reproduce the observed features, even for intermediate values of U. Although we derived explicit expressions only in 1D, we would like to emphasize that due to the local nature of the sum rules the distribution of weight over the two bands is qualitatively very similar also for higher dimensions [14].

Especially for the optical spectrum the lowest order corrections result directly from the three-site hopping terms $\vec{\delta} \neq \vec{\delta}'$ in the strong-coupling Hamiltonian [15]. Therefore, the simpler t-J model *does not describe* accurately the spectral properties of the Hubbard model.

The fast disappearance of the UHB with doping shown in both experiments on doped La_2CuO_4 [1–4] are in qualitative or even quantitative agreement with a two-dimensional Hubbard model for intermediate U. However, the physical interpretation is different. The reduction of the UHB in the optical spectrum has three reasons: (i) a hole breaks two spin bonds in the direction of the electric field, (ii) the spin order is weakened, and (iii) a dynamical weight transfer due to initial-final state interference occurs. In the one-particle spectrum the UHB weight is reduced since there are simply less sites that, after adding a particle, become doubly occupied. The effect is, however, again largely enhanced by the interference effects. As a result, the low-energy peaks are larger than might be expected from the actual level of doping.

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