Particle-hole bound states in Mott-Hubbard insulators

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I present results that indicate the existence of strongly bound particle-hole excitations below the edge of the upper Hubbard band in a single-band, large-U Hubbard model in two dimensions, at half-filling. The appearance of these exciton states is due to correlation effects between a vacancy and a doubly occupied site induced by the antiferromagnetic background, in contrast to the conventional mechanism of exciton formation via long-range Coulomb attraction. I argue that in the insulating parent compounds of the high-temperature superconducting cuprates these correlation effects are of the same order of magnitude as particle-hole attraction due to longer-range Coulomb interactions.

Anderson[1] has proposed that much of the physics of high-temperature superconducting cuprates can be understood within the framework of a single-band, large-U Hubbard model in two dimensions:

$$H=-t\sum_{\langle i,j\rangle}(c_{i\sigma}^{\dagger}c_{j\sigma}+c_{j\sigma}^{\dagger}c_{i\sigma})+U\sum_{j}n_{i\uparrow}n_{i\downarrow}\ , \tag{1}$$

where $\langle i,j\rangle$ are nearest-neighbor sites on a $2d$ square lattice, $c_{i\sigma}^{\dagger}$ creates an electron of spin σ on (Cu) site i, $n_{i\sigma}\equiv c_{i\sigma}^{\dagger}c_{i\sigma}$ is the number of electrons of spin σ on site i, t is the hopping energy and U is the effective on-site repulsion. This Hamiltonian does not include all the details of the high-energy [O(eV)] physics of real cuprate materials, as it does not include oxygen orbitals. It has, however, been argued[2] that (1) does faithfully describe the low-energy sector, and confronts the essential problem of strong correlations induced by local electron-electron repulsion.

The Hilbert space of the large-U Hubbard model can be divided into a *lower Hubbard band* (LHB) *and an upper Hubbard band* (UHB). The LHB we define to be the manifold of states which have no doubly occupied sites, while the UHB is the Hilbert-space complement of the LHB, i.e., the manifold of states containing at least one site with double occupancy.

Near half-filling, the two "bands" are separated by an energy $O(U)$. At exactly half-filling there are no charge carriers in the LHB and the system is a Mott-Hubbard insulator. The zero-temperature ground state in this case has long-range antiferromagnetic (AF) order. At low doping a finite density of charge carriers appears, but the ground state is believed to retain at least short-range antiferromagnetic correlations. While the low-energy excitations are confined to the LHB, understanding effects such as optical conductivity or Raman scattering at energy transfers of O(eV) (Ref. 3) requires the study of the UHB as well. However, as in the case of the LHB, this will involve nontrivial effects due to the presence of strong antiferromagnetic correlations.

It has long been known that in conventional [i.e., nearly free-electron (NFE)] insulators and semiconductors the absence of low-energy particle-hole excitations prevents screening of the long-range Coulomb interaction, leading to the formation of *bound* particle-hole states: *excitons*. The presence of excitonic bound states or resonances can quite dramatically alter physical properties, e.g., the shape of the optical edge. In the case of a Mott-Hubbard insulator however, there is the further complication of strong correlations, so that the conventional theory needs to be revised.

While it is true that in order to make full contact with experiments we would need to use a model more complicated than (1), our purpose here is to investigate the effects of strong correlations on the particle-hole excitations of a Mott-Hubbard system at half-filling as described by (1). We shall argue that a band of bound particle-hole states appears (below the continuum of the UHB), as a consequence of many-body correlations induced by on-site repulsion and antiferromagnetic short-range order. Later, we briefly discuss the effect of long-range Coulomb interactions.

Thus motivated, we have studied the nature of particle-hole excitations in the UHB of the large-U half-filled single-band Hubbard model (1). This UHB excitation consists of a vacancy ("hole"), a doubly occupied site ("double"), and a background spin configuration. We shall show that there exist strongly bound hole-double states which, by analogy with the case of bound particle-hole states in NFE insulators, we call "magnetic" excitons. The term *magnetic* is used to emphasize that the hole-double interaction is mediated by the antiferromagnetic background and that the phenomenon is fundamentally different from that of excitons bound by Coulomb attraction.

Below we shall proceed to (i) derive an effective t-J Hamiltonian governing the hole-double dynamics, (ii) discuss the physics of hole-double correlations in the Ising limit, (iii) present the results of an exact diagonalization of the effective Hamiltonian on an 18-site cluster, and (iv) briefly discuss the relevance of the results obtained to the parent compounds of the high-temperature superconducting cuprates.

We begin by applying perturbation theory about the point $t/U=0$ at which the LHB and the UHB are separated in energy by U. Apart from generating superexchange interactions within each of the bands, turn-

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ing on t has two other effects: (i) the hole and double are able to move, and (ii) the LHB and the UHB are no longer orthogonal because the hopping term connects them. We reorthogonalize the LHB and the UHB by a canonical transformation

$$H \rightarrow e^{iS} H e^{-iS} ,$$

where

$$S = \frac{it}{U} \sum_{\langle i,j \rangle} [P_i^{(2)} T_{ij} (P_j^{(0)} + P_j^{(1)}) - (P_i^{(0)} + P_i^{(1)}) T_{ij} P_j^{(2)} + (i \leftrightarrow j)]$$

is chosen so as to remove the mixing between the LHB and the UHB to $O(t/U)$. $P_i^{(n)}$ projects onto the n-particle subspace on site i (e.g., $P_i^{(2)}$ is unity if there are two electrons on site i, and vanishes otherwise). Retaining terms to $O(t^2/U)$ only we obtain an effective Hamiltonian at half-filling:[4]

$$\begin{aligned}
&H_{\text{eff}} = H_t^h + H_t^d + H_J + H_{\text{ph}} + H_U, \quad H_t^h = -t \sum_{\langle i,j \rangle} P_i^{(1)} P_j^{(0)} c_{i\sigma}^\dagger c_{j\sigma} P_i^{(0)} P_j^{(1)} + \text{H.c.} , \\
&H_t^d = -t \sum_{\langle i,j \rangle} P_i^{(2)} P_j^{(1)} c_{i\sigma}^\dagger c_{j\sigma} P_i^{(1)} P_j^{(2)} + \text{H.c.}, \quad H_J = J \sum_{\langle i,j \rangle} P_i^{(1)} P_j^{(1)} \mathbf{S}_i \cdot \mathbf{S}_j P_i^{(1)} P_j^{(1)} , \\
&H_{\text{ph}} = \frac{J}{4} \sum_{\langle i,j \rangle} [P_i^{(0)} P_j^{(2)} + (i \leftrightarrow j)] + \frac{J}{2} \sum_{\langle i,j \rangle} [P_i^{(2)} P_j^{(0)} c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger c_{j\downarrow} c_{j\uparrow} P_i^{(0)} P_j^{(2)} + (i \leftrightarrow j)] \\
&\quad + \frac{J}{8} \sum_{\langle i,j,k \rangle} [P_i^{(2)} \overline{P_j P_k} T_{ij} \overline{P_i P_j P_k} T_{jk} \overline{P_i P_k} P_j^{(2)} + P_j^{(2)} \overline{P_i P_k} T_{ij} \overline{P_i P_j P_k} T_{jk} \overline{P_i P_j} P_k^{(2)} \\
&\quad + P_j^{(2)} \overline{P_i P_k} T_{ij} \overline{P_i P_j P_k} T_{jk} \overline{P_i P_k} P_j^{(2)} + P_i^{(2)} \overline{P_j P_k} T_{ij} \overline{P_i P_j P_k} T_{jk} \overline{P_i P_j} P_k^{(2)} + (i \leftrightarrow j) + \text{H.c.}] , \\
&H_U = U \sum_i P_i^{(2)} .
\end{aligned} \tag{2}$$

Here $T_{ij} = c_{i\sigma}^\dagger c_{j\sigma}$, $\mathbf{S}_i = c_{i\sigma}^\dagger \tau^{\sigma\sigma'} c_{i\sigma'}/2$, the brackets $\langle i,j \rangle$ and $\langle i,j,k \rangle$ refer to *nearest neighbors* (in the latter i, j, and k are *distinct* sites), and $\bar{P}_i \equiv (P_i^{(0)} + P_i^{(1)})$ projects out double occupancy.

The physics of this Hamiltonian is quite straightforward: H_t^h hops holes, H_t^d hops doubles, H_J is the Anderson antiferromagnetic superexchange term ($J = 4t^2/U$), and H_{ph} are hole-double pair hopping terms which arise from virtual hole-double recombination processes (and are, therefore, nonvanishing only on states which have the hole and double as near neighbors). The most important physics lies in the first three terms where hopping is in competition with AF order induced by the superexchange term, so we introduce

$$\tilde{H}_{\text{eff}} = H_t^h + H_t^d + H_J + H_U . \tag{3}$$

We have omitted next-nearest-neighbor hopping terms and recombination terms involving a hole and a double on next-nearest-neighbor sites [both being $O(J)$]. The latter would give rise to $O(J^2/U)$ energy shifts ($\ll J$) and, potentially, widths for the UHB eigenstates of H_{eff}. But for sufficiently large U/t the widths will vanish simply because such recombination terms would not produce a sufficient number of magnons to conserve energy in a decay process (a consequence of the large U/t limit). Thus, as written, H_{eff} conserves *separately* the total number of holes and the total number of doubles. There is *no recombination*. This is a restatement of the fact that we have orthogonalized, to $O(t/U)$, the LHB and the UHB.

We shall now attempt to study the structure of the UHB governed by H_{eff}. This is a nontrivial problem involving strong correlations. We apply two complementary methods: (a) the limit of Ising spins, and (b) a finite cluster exact diagonalization. By (a) we mean replacing H_J by $J_z \sum_{\langle i,j \rangle} S_i^z S_j^z$. The value of this limit is that the ground state for the spins is a simple Ising antiferromagnet. As a consequence, a hole or double is unable to move without disrupting the spin order. If the hole or double hops l lattice spacings it gains $\sim t/l^2$ in kinetic energy at a cost of $\sim Jl$ in lost magnetic energy in the "string" of flipped spins left in its wake. These energy scales determine a time scale $\tau_{\text{string}} \sim (J_z^{-2/3} t^{-1/3})$. Now, if one turns on $J_\perp$, the transverse spin fluctuations, there will be a second time scale $\tau_{\text{flip}} \sim 1/J_\perp$ associated with flipping a pair of spins, and possible restoration of spin order. Thus

$$\tau_{\text{string}}/\tau_{\text{flip}} \sim (J_\perp/J_z)(J_z/t)^{1/3}$$

so that, even in the Heisenberg limit, $J_\perp = J_z \equiv J$, there is always a regime in which $\tau_{\text{string}}/\tau_{\text{flip}} \ll 1$. This requires sufficiently small J/t, and in this regime we should, in the spirit of Born-Oppenheimer, solve the string (Ising) problem first and *then* deal with spin flips.

Thus in the Ising limit the physical picture is that whenever a hole or double hops, it leaves in its wake a string of spins which are aligned opposite to the sublattice magnetization of the Ising antiferromagnetic ground state. In the single-hole (or double) case this translates into a confining linear potential.[5,6] However, if a hole and double move in a coherent manner, "chasing" one another, it is possible that they may gain more kinetic energy than a well-separated hole and double would for

there would be only *one* string, with the hole at one end and the double at the other, instead of two. Thus there is the possibility of bound states via correlated hopping of hole and double.

In all of our analysis, we shall first consider $\tilde{H}_{\rm eff}$, either in the Ising or the Heisenberg limits and then determine the effect of the O(J) repulsive pair hopping term $H_{\rm ph}$.

Our approach is to modify a technique used by Shraiman and Siggia for the two-hole problem[5] and based upon the Brinkman-Rice retracing path approximation.[7,8] In this approximation the Hamiltonian is diagonalized in the Hilbert subspace consisting of the states generated by the successive action of the hopping part of the Hamiltonian on some initial "seed" state. The idea behind the scheme is to construct the states which take most advantage of the kinetic energy in the presence of the AFM background.[9]

Let us begin with a "seed" state

$$|\mu,\sigma\rangle \equiv \frac{1}{\sqrt{N}} \sum_{\mathbf{r}} e^{i\mathbf{k}\cdot(\mathbf{r}+\mu/2)} c_\sigma(\mathbf{r}) c_\sigma^\dagger(\mathbf{r}+\mu)|0\rangle \ , \tag{4}$$

where $\mu=\pm x, \pm y$, and $|0\rangle$ is the Ising AF ground state. These are the eight fundamental "bond" states with momentum $\mathbf{k}$.

The (normalized) string states $|l,m;\mu,\sigma\rangle$ are generated by the action of H_t^h and H_t^d upon $|\mu,\sigma\rangle$:

$$\begin{aligned}|l,m;\mu,\sigma\rangle \equiv (-1)^l \frac{1}{\sqrt{N}} & \times \sum_{\mathbf{r}} e^{i\mathbf{k}\cdot(\mathbf{r}+\mu/2)} \frac{1}{3^{(l+m)/2}} \\ & \times \sum_{\nu_l \neq -\nu_{l-1}} \cdots \sum_{\nu_1 \neq -\mu} \sum_{\rho_m \neq -\rho_{m-1}} \cdots \sum_{\rho_1 \neq -\mu} c_{\sigma_l}(\mathbf{r}_l) c_{\lambda_m}^\dagger(\mathbf{r}_m) \\ & \times \{[c_{\sigma_l}^\dagger(\mathbf{r}_{l-1}) c_{\sigma_{l-1}}(\mathbf{r}_{l-1})] \\ & \times \cdots [c_{\sigma_1}^\dagger(\mathbf{r}) c_\sigma(\mathbf{r})][c_{\lambda_m}(\mathbf{r}'_{m-1}) c_{\lambda_{m-1}}^\dagger(\mathbf{r}'_{m-1})] \\ & \times \cdots [c_{\lambda_1}(\mathbf{r}+\mu) c_\sigma^\dagger(\mathbf{r}+\mu)]\}|0\rangle \ . \end{aligned} \tag{5}$$

where $\mathbf{r}_j = \mathbf{r} - \sum_{i=1}^{j} \nu_i$ and $\mathbf{r}'_j = \mathbf{r}+\mu+\sum_{i=1}^{j}\rho_i$.

Up to normalization $|l,m;\mu,\sigma\rangle$ is the state generated from $|\mu,\sigma\rangle$ by hopping the hole l lattice spacings and the double m lattice spacings without retracing at any stage. It consists of (a superposition of) strings of $l+m$ flipped spins, with the hole at one end of the string, and the double at the other.

The operators in square brackets above have the effect of adding a unit of length to the string, either at the hole end or the double end. We therefore introduce ladder operators $h^\dagger$, $d^\dagger$, h, d connecting the $|l,m;\mu,\sigma\rangle$ states, specifically,

$$(h^\dagger)^l (d^\dagger)^m |\mu,\sigma\rangle \equiv |l,m;\mu,\sigma\rangle \ , \tag{6}$$

which satisfy $hh^\dagger=1$, $dd^\dagger=1$, $[h^\dagger,d^\dagger]=0$, $[h,d^\dagger]=0$, and $h|\mu,\sigma\rangle = d|\mu,\sigma\rangle = 0$. That is, $h^\dagger$ extends the string (and h contracts it) at the *hole* end, while $d^\dagger$ extends the string (d contracts it) at the *double* end. Then

$$H_t^h + H_t^d = -\sqrt{3}t(h^\dagger + d^\dagger + h + d) \ . \tag{7}$$

The action of $\tilde{H}_{\rm eff}$ on the string states is straightforward; the only complications arise with $\tilde{H}_{\rm eff}|0,m;\mu,\sigma\rangle$ and $\tilde{H}_{\rm eff}|l,0;\mu,\sigma\rangle$, for in these cases the string "origin" moves. This is where the important physics lies; without these terms the hole and double would be essentially noninteracting.

Using operator algebra and the properties of the Ising ground state one can derive

$$hd^\dagger|\mu,\sigma\rangle = \frac{1}{3\sqrt{N}} \sum_{\mathbf{r}} \sum_{\nu \neq -\mu} e^{i\mathbf{k}\cdot(\mathbf{r}-\mu/2)} c_{-\sigma}(\mathbf{r}) \times c_{-\sigma}^\dagger(\mathbf{r}+\nu)|0\rangle$$

and

$$dh^\dagger|\mu,\sigma\rangle = \frac{1}{3\sqrt{N}} \sum_{\mathbf{r}} \sum_{\nu \neq -\mu} e^{i\mathbf{k}\cdot(\mathbf{r}+\nu+\mu/2)} c_{-\sigma}(\mathbf{r}) \times c_{-\sigma}^\dagger(\mathbf{r}+\nu)|0\rangle \ .$$

Observing a spin (sublattice) symmetry under $\mathcal{S}: |\mu,\sigma\rangle \to |\mu,-\sigma\rangle$ we define

$$|\mu\rangle_\pm \equiv (1/\sqrt{2})[|\mu,\uparrow\rangle \pm |\mu,\downarrow\rangle] \tag{8}$$

and finally arrive at the Hamiltonian (measuring energies relative to the Ising AF ground state, and shifting by a trivial $-U$ constant),

TABLE I. $\mathcal{S}=+1$ and $\mathcal{I}=+1$ binding energies for $\tilde{H}_{\text{eff}}$ in the Ising limit.

J_z/t	$\Delta_2(0,0)$	$\Delta_2(0,\pi/3)$	$\Delta_2(0,2\pi/3)$	$\Delta_2(0,\pi)$	$\Delta_2(0,0)$	$\Delta_2(\pi/3,\pi/3)$	$\Delta_2(2\pi/3,2\pi/3)$	$\Delta_2(\pi,\pi)$
0.2	−0.473	−0.309	−0.073	0.000	−0.473	−0.174	+0.088	+0.133
0.4	−0.683	−0.498	−0.183	−0.062	−0.683	−0.329	+0.105	+0.200
0.6	−0.836	−0.641	−0.285	−0.133	−0.836	−0.456	+0.104	+0.256
0.8	−0.959	−0.761	−0.383	−0.210	−0.959	−0.568	+0.082	+0.291
1.0	−1.065	−0.865	−0.475	−0.290	−1.065	−0.668	+0.043	+0.304

$$\tilde{H}_{\text{eff}}|l,m;\boldsymbol{\mu}\rangle_{\pm}=-\sqrt{3}t[|l+1,m;\boldsymbol{\mu}\rangle_{\pm}+|l,m+1;\boldsymbol{\mu}\rangle_{\pm}+|l-1,m;\boldsymbol{\mu},\rangle_{\pm}+|l,m-1;\boldsymbol{\mu}\rangle_{\pm}]$$
$$+(2+l+m)J_z|l,m;\boldsymbol{\mu}\rangle_{\pm}\quad(l,m\geq 1)\,,$$

$$\tilde{H}_{\text{eff}}|0,m;\boldsymbol{\mu}\rangle_{\pm}=-\sqrt{3}t[|0,m+1;\boldsymbol{\mu}\rangle_{\pm}+|0,m-1;\boldsymbol{\mu}\rangle_{\pm}+|1,m;\boldsymbol{\mu}\rangle_{\pm}]$$
$$\mp\frac{t}{\sqrt{3}}\sum_{\boldsymbol{\nu}\neq-\boldsymbol{\mu}}e^{-i(\mathbf{k}/2)\cdot(\boldsymbol{\nu}+\boldsymbol{\mu})}|0,m-1;\boldsymbol{\nu}\rangle_{\pm}+(2+m)J_z|0,m;\boldsymbol{\mu}\rangle_{\pm}\,,\tag{9}$$

$$\tilde{H}_{\text{eff}}|l,0;\boldsymbol{\mu}\rangle_{\pm}=-\sqrt{3}t[|l+1,0;\boldsymbol{\mu}\rangle_{\pm}+|l-1,0;\boldsymbol{\mu}\rangle_{\pm}+|l,1;\boldsymbol{\mu}\rangle_{\pm}]$$
$$\mp\frac{t}{\sqrt{3}}\sum_{\boldsymbol{\nu}\neq-\boldsymbol{\mu}}e^{i(\mathbf{k}/2)\cdot(\boldsymbol{\nu}+\boldsymbol{\mu})}|l-1,0;\boldsymbol{\nu}\rangle_{\pm}+(2+l)J_z|l,0;\boldsymbol{\mu}\rangle_{\pm}\,,$$

$$\tilde{H}_{\text{eff}}|\boldsymbol{\mu}\rangle_{\pm}=-\sqrt{3}t[|1,0,\boldsymbol{\mu}\rangle_{\pm}+|0,1,\boldsymbol{\mu}\rangle_{\pm}]+\tfrac{7}{4}J_z|\boldsymbol{\mu}\rangle_{\pm}\,.$$

The operator,

$$\mathcal{I}:|l,m;\boldsymbol{\mu},\sigma\rangle\rightarrow|m,l;-\boldsymbol{\mu},-\sigma\rangle\,,$$

which interchanges hole and double commutes with $\tilde{H}_{\text{eff}}$. Furthermore, $\mathcal{I}$ commutes with the translation operator, and since $\mathcal{I}^2=1$ we may label our eigenstates by $\mathbf{k}$ and "s" or "p", "s" states being even under $\mathcal{I}$, "p" states being odd. Under $\mathbf{k}\rightarrow\mathbf{k}+(\pi,\pi)$, $\mathcal{S}\rightarrow-\mathcal{S}$ while $\mathcal{I}$ is invariant. So we may restrict $\mathbf{k}$ to the magnetic Brillouin zone or, alternatively, consider just one spin channel for the full Brillouin zone, $[-\pi,\pi]\times[-\pi,\pi]$. We choose the latter.

The eigenvalues $E_2(\mathbf{k})$ of $\tilde{H}_{\text{eff}}$ were determined for $\mathbf{k}\in[-\pi,\pi]\times[-\pi,\pi]$ in the $\mathcal{S}=+1$, $\mathcal{I}=\pm1$ channels. The binding energies $\Delta_2(\mathbf{k})$ (in units of t) of the lowest-lying states are defined by

$$\Delta_2(\mathbf{k})\equiv E_2(\mathbf{k})-2E_1^{\text{Ising}}\,,\tag{10}$$

where the (dispersionless) single-hole energy in the Ising limit is given by[5]

$$E_1^{\text{Ising}}=-2\sqrt{3}t+2.74J_z^{2/3}t^{1/3}\,.$$

We find that the s-wave states bind most strongly at $\mathbf{k}=(0,0)$ with binding energies $\sim O(J_z^{2/3}t^{1/3})$ as $J_z/t\rightarrow0$, weakening as one moves away from the zone center. For $\mathbf{k}=(0,0)$ a fit of

$$\Delta_2(0,0)=-1.88J_z^{2/3}t^{1/3}+0.83J_z$$

is good to better than 1%, at least for $0.05\leq J_z/t\leq1.0$. Further results are presented in Table I. In contrast, the p-wave states form a flat band and bind weakly, with an approximate fit $\Delta_2(\mathbf{k})\sim J_z/2$.

H_{ph} acts like a short-range $O(J)$ repulsion and thus raises the s-wave binding energies (the p-wave energies are unaffected, because they do not couple to H_{ph}) as shown in Table II. However, there is still a large $O(J_z^{2/3}t^{1/3})$ binding.

The spectrum for the Ising limit approaches that of H_{eff} *asymptotically* as $J/t\rightarrow0$. However, for the systems of physical interest $0.1<J/t<1$. We expect the excitonic binding to be weaker in this regime simply because the action of $J_\perp$ is to flip spins and thus weaken the "tension" of the Ising strings. In order to study the effect of $J_\perp$ we have undertaken an 18-site cluster exact diagonalization of the UHB of H_{eff}. We limit ourselves here to presenting the main results; further discussion of the technical

TABLE II. $\mathcal{S}=+1$ and $\mathcal{I}=+1$ binding energies for $\tilde{H}_{\text{eff}}+H_{\text{ph}}$ in the Ising limit.

J_z/t	$\Delta_2(0,0)$	$\Delta_2(0,\pi/3)$	$\Delta_2(0,2\pi/3)$	$\Delta_2(0,\pi)$	$\Delta_2(0,0)$	$\Delta_2(\pi/3,\pi/3)$	$\Delta_2(2\pi/3,2\pi/3)$	$\Delta_2(\pi,\pi)$
0.2	−0.437	−0.282	−0.061	+0.006	−0.437	−0.156	+0.089	+0.133
0.4	−0.561	−0.396	−0.126	−0.026	−0.561	−0.250	+0.115	+0.198
0.6	−0.592	−0.430	−0.150	−0.036	−0.592	−0.281	+0.136	+0.248
0.8	−0.568	−0.413	−0.139	−0.026	−0.568	−0.269	+0.152	+0.271
1.0	−0.506	−0.358	−0.100	+0.006	−0.506	−0.223	+0.169	+0.263

TABLE III. s-wave particle-hole binding energies.

J/t	$E_1(\pi/2,\pi/2)$	$E_2^s(0,0)$	$E_2^s(\pi,\pi)$	$\mathcal{E}_2^s(0,0)$	$BE^s(0,0)$
0.4	-1.974	-4.401	-4.139	-4.270	-0.322
0.6	-1.59 ± 0.02[a]	-3.650	-3.298	-3.474	-0.29 ± 0.03
0.8	-1.21 ± 0.02[a]	-2.955	-2.517	-2.736	-0.32 ± 0.03
1.0	-0.883	-2.300	-1.777	-2.038	-0.272
1.2	-0.55 ± 0.02[a]	-1.678	-1.069	-1.374	-0.27 ± 0.03

[a]Interpolated from Ref. 10.

details will be presented elsewhere.[4]

The main results are presented in Tables III and IV, and Fig. 1. We focus attention on the zero-momentum state, the best candidate magnetic exciton state. All energies are in units of t and measured relative to the ground state at half-filling of the Heisenberg Hamiltonian, which, on an 18-site cluster, has energy $-12.113J$. The overall energy shift U is dropped. $E_2^{s,p}(\mathbf{k})$ is the lowest cluster energy for $\tilde{H}_{\rm eff}$ (again, the effect of $H_{\rm ph}$ is postponed) for a hole-double state with momentum $\mathbf{k}$ and even ("s") or odd ("p") under hole-double interchange. $\mathcal{E}_2^{s,p}(0,0)$ is the average of $E_2^{s,p}(0,0)$ and $E_2^{s,p}(\pi,\pi)$ and is our approximation to the infinite volume result (where there will be degeneracy due to the presence of long-range order).[4,11] $E_1(\pi/2,\pi/2)$ is the energy of an isolated hole (or double) with momentum $(\pm\pi/2,\pm\pi/2)$.[10] A combination of two such states with total momentum zero represents the minimum energy state for a "noninteracting" hole and double in the $\mathbf{k}=0$ channel. The difference between this energy and the cluster energies give estimates of the binding energy. We expect that the binding energies determined by $E_2^{s,p}(0,0)$ will give approximate upper and lower bounds for the binding energies in the thermodynamic limit. Those determined by $\mathcal{E}_2^{s,p}(0,0)$ are our estimate of the binding energies in the thermodynamic limit (see Fig. 1). The results clearly support the existence of an s-wave exciton band: the binding energy is roughly independent of J, which supports the notion of binding being due to correlated hopping. In contrast, the binding in the p-channel is at best $O(J)$.

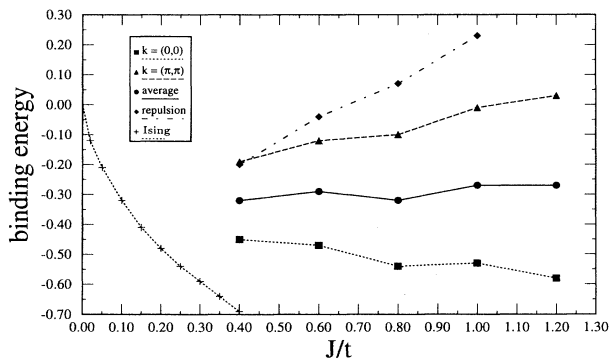

FIG. 1. A summary of the numerical results obtained on an 18-site cluster. For $\tilde{H}_{\rm eff}$ the squares and triangles denote the binding energies calculated from $E_2^s(0,0)$ and $E_s^s(\pi,\pi)$, respectively. The circles denote the average of these two estimates, and represent the estimate of the binding energies in the thermodynamic limit. The diamonds denote the estimated upper bound on the binding energies once the pair hopping repulsion $H_{\rm ph}$ has been accounted for. The crosses in the lower left are the $k=(0,0)$, $\mathcal{S}=+1$, $\mathcal{J}=+1$ binding energies for $\tilde{H}_{\rm eff}$ in the Ising limit, which are exact in the asymptotic limit of small J/t.

We now take into account the effect of the pair hopping repulsive term $H_{\rm ph}$. Taking the results for $H_{\rm eff}$ and added the shift in the *Ising* s-wave exciton due to $H_{\rm ph}$ we obtain an *estimated upper bound* on the binding energy. From the various estimates presented in Fig. 1 it is plausible that the results obtained in the Ising limit, and the physical picture therein, remain at least qualitatively valid in the intermediate coupling regime, and that correlated hopping-induced binding could persist even up to $J/t\approx1$.

For the remainder of the Brillouin zone, we also expect qualitative similarity with the Ising results, in particular an s-wave dispersion $\sim O(J^{2/3}t^{1/3})$. While it is possible that the p-wave band may have a minimum at finite momentum, again the $O(J)$ terms introduced by $J_\perp$ can only lower the energy by $O(J)$, so that any p-wave binding will still be at most weak.

We now briefly discuss the effect of long-range Coulomb terms added to (1). Such terms will give rise to an *attractive* hole-double Coulomb interaction. Considering the Ising limit once again, we may write $\mathcal{U}_{\rm mag}\sim Jl$ for the magnetic energy cost of a hole-double string of length l, and $\mathcal{U}_{\rm elec}\sim -V/(\epsilon r)$ for the Coulomb energy cost to separate a hole and double by distance r. Since $r\sim l^{1/2}$ we can define a naive electrostatic length scale $l_{\rm elec}\sim(\epsilon t/V)^{2/3}$ determined by the hopping and Coulomb terms, and a magnetic length scale $l_{\rm mag}\sim(t/J)^{1/3}$ determined by the hopping term and $\mathcal{U}_{\rm mag}$. Now if $l_{\rm mag}/l_{\rm elec}\ll1$ then the long-range Coulomb terms are unimportant. It is of interest to note that if we suppose a single-band description of the high-T_c copper oxides with $t\sim0.4$ eV, $J\sim0.15$ eV, $V\sim3.6$ eV, and $\epsilon\sim6-30$ then we have $l_{\rm mag}/l_{\rm elec}\sim O(1)$. Thus, Coulomb effects should not be totally disregarded in these materials and they will certainly alter the excitonic binding energies. However, it is equally clear that the many-body nature of "excitons" in these systems cannot be ignored either.

In conclusion, we have studied the spectrum of neutral, two-particle UHB excitations in the single-band large-U Hubbard model at half-filling via a canonical transformation to a Hamiltonian of t-J type. We have given a general argument that, in the asymptotic limit $J/t\ll1$, there will always be a pocket of s-wave "magnetic" excitons, centered on $\mathbf{k}=(0,0)$ in the magnetic Brillouin zone, the result of strong magnetic correlations, with binding

TABLE IV. p-wave particle-hole binding energies.

J/t	$E_1(\pi/2,\pi/2)$	$E_2^p(0,0)$	$E_2^p(\pi,\pi)$	$\mathcal{E}_2^p(0,0)$	$BE^p(0,0)$
0.4	-1.974	-3.813	-4.196	-4.004	-0.06
0.6	-1.59 ± 0.02[a]	-2.92	-3.415	-3.17	$+0.01\pm0.03$
0.8	-1.21 ± 0.02[a]	-2.14	-2.71	-2.42	0.00 ± 0.03
1.0	-0.883	-1.43	-2.06	-1.74	$+0.03$
1.2	-0.55 ± 0.02[a]	-0.765	-1.449	-1.107	-0.01 ± 0.03

[a]Interpolated from Ref. 10.

energies $\sim O(J^{2/3}t^{1/3})$. Numerical studies of excitations on an 18-site cluster provide evidence of binding persisting up to $J/t \approx 1$, with a maximum binding of $\approx 0.3t$ at $J/t \approx 0.3$. We emphasize that these bound particle-hole states are fundamentally different from the conventional electronic excitons of nearly free-electron insulators and semiconductors.

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