Two-Particle Excitations in Antiferromagnetic Insulators

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The energy of one and two holes in a Hubbard antiferromagnet for nonzero exchange, J, in the Ising limit is calculated within the Brinkman-Rice approximation. Only the p- and d-symmetry states bind with an energy of order J. The implications for superconductivity and antiferromagnetism of doped Hubbard insulators is discussed.

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Interest in nonphonon-induced pairing has been stimulated by the discovery of the copper-oxide–based superconductors[1] and has prompted a reexamination of strongly correlated systems. In insulating materials closely related to those with the highest T_c, a three-dimensional antiferromagnetic (AF) transition was observed, as well as finite but long-range two-dimensional AF correlations.[2] Both the superconductive and magnetic properties are sensitive to doping. One class of models, therefore, hypothesizes that pairing originates from purely Coulombic effects, and the AF correlations thereby induced in narrow-d-band materials.[3-7]

A natural starting point is the strong Hubbard model[8] near half filling which describes an antiferromagnet with

$$\mathcal{H} = -w\sum_{(ij)}(c_{i\sigma}^{\dagger}c_{j\sigma}+\text{H.c.})+J_z\sum_{(ij)}S_i^zS_j^z+J_\perp\sum_{(ij)}(S_i^xS_j^x+S_i^yS_j^y). \tag{1}$$

The summations extend over all pairs of nearest-neighbor sites on a simple two-dimensional square lattice; $c_{i\sigma}$ are the usual fermion operators (σ=spin); the spin operators $\mathbf{S}_i \equiv \frac{1}{2}\sum c_{i\sigma'}^{\dagger}\boldsymbol{\tau}_{\sigma'\sigma}c_{i\sigma}$ ($\boldsymbol{\tau}$=Pauli matrices); and in the state space associated with (1), double occupancy of a site is forbidden.

While (1) with $w \gg J_z = J_\perp = J$ is essentially the large Coulomb limit of the Hubbard model, it can also arise in more complicated settings when charge fluctuations can be integrated out and attention is focused on the spin degrees of freedom.[3] We have split the magnetic interaction into two pieces in order to have a parameter, $J_\perp/J_z$, which turns off the quantum fluctuations in the AF ground state. For real materials $J_\perp = J_z$.[2]

In the extreme $J/w \ll 1$ limit, the ground state of a hole involves a magnetic polaron.[10] The kinetic energy is minimized by making a disk of radius R ferromagnetic so that the hole can sit at the free-particle band edge with an energy $-4w$. A continuum calculation can only be done precisely for $J_\perp = 0$ (the polaron walls are sharp holes.[9] In fact, a single hole is not in a simple Bloch state since when it hops, it scrambles the AF arrangement of spins in the ground state. The calculation of its kinetic energy is a nontrivial many-body problem, which has been studied by Brinkman and Rice (BR).[10] They found that in the limit of $J/w \to 0$ the AF correlations in the ground state lead to a narrowing of the band and push the energy of the hole above the value it would have in an empty band. The essence of the BR approximation is to label the configuration space by distinct paths. They also neglect the spin dynamics, which is not unreasonable for $w/J \gg 1$, since the hole will hop many times (creating a "string" of overturned spins) before the spins can relax.

It will suffice to consider the Hamiltonian (1) for any $J_\perp/J_z \leq 1$), and yields

$$E = -4w + 8.5(J_z w)^{1/2} + O(J_z). \tag{2}$$

While (1) predicts that it is energetically favorable for many holes to share the same polaron, phase separation is prevented by the full Coulomb potential. Purely as a result of numerical factors, there is a sizable region of small J/w, $1 \gtrsim J/w \gtrsim 5\times 10^{-3}$, where the BR approximation with $J_z \neq 0$, described in the next paragraph, gives a lower energy than (2) for a single hole. A strong-coupling, lattice calculation is still appropriate and we henceforth confine our attention to this regime.[11,12]

Since the BR approximation will readily generalize to two holes, we describe the one-hole calculation first for a Néel state and $J_z > 0$. Starting from the ground state, successive applications of the kinetic term in (1) create a series of many-body states that can be labeled as a walk or string of bond directions $|\boldsymbol{\tau}_1, \boldsymbol{\tau}_2, \ldots, \boldsymbol{\tau}_l\rangle$, $\boldsymbol{\tau}_i = \pm\hat{\mathbf{x}}$, $\pm\hat{\mathbf{y}}$, with no retracings, $\boldsymbol{\tau}_{i+1} \neq -\boldsymbol{\tau}_i$. The tree or BR ap-

proximation assumes that all allowed strings are distinct states. The first real lattice state to be double counted occurs when the hole runs around a plaquette three times in succession.[10] Most spin states with a common value of the hole position, $\mathbf{r}=\sum_i^l \boldsymbol{\tau}_i$, are truly distinct, which is why the approximation works so well. (In three dimensions it gave a band edge of $-2\sqrt{5}=-4.47$ versus an "exact" series estimate of -4.45.[10])

If $|0\rangle$ denotes a hole with no string, we define mutually adjoint "raising" and "lowering" operators, $h^\dagger$, h as

$$h^\dagger|\boldsymbol{\tau}_1,\ldots,\boldsymbol{\tau}_l\rangle=\frac{1}{\sqrt{3}}\sum_{\boldsymbol{\tau}_{l+1}\neq-\boldsymbol{\tau}_l}|\boldsymbol{\tau}_1,\ldots,\boldsymbol{\tau}_{l+1}\rangle,$$
$$h|\boldsymbol{\tau}_1,\ldots,\boldsymbol{\tau}_l\rangle=\frac{1}{\sqrt{3}}|\boldsymbol{\tau}_1,\ldots,\boldsymbol{\tau}_l\rangle, \tag{3}$$

and $h|0\rangle=0$, $h^\dagger|0\rangle=\frac{1}{3}\sqrt{3}\sum_{\boldsymbol{\tau}_1}|\boldsymbol{\tau}_1\rangle$. Then the kinetic energy is just $-\sqrt{3}(h^\dagger+h)$, and we can find its minimum exactly within the orthonormal set of states $|l\rangle=\frac{1}{2}\sqrt{3}h^{\dagger l}|0\rangle$, $l>0$. If for the moment we compute the exchange as if the walk is rectilinear, (1) reduces to[13]

$$H|0\rangle=-2|1\rangle+J_z|0\rangle,$$
$$H|1\rangle=-2|0\rangle-\sqrt{3}|2\rangle+\tfrac{5}{2}J_z|1\rangle,$$
$$H|l\rangle=-\sqrt{3}(|l-1\rangle+|l+1\rangle)+(\tfrac{3}{2}+l)J_z|l\rangle, \tag{4}$$
$$l\geq 2.$$

(Henceforth $w\equiv 1$ in all formulas, and we take the Néel state to be the zero of energy.) A numerical solution yields a ground-state energy $E_1=-2\sqrt{3}+2.74J_z^{2/3}$ (the coefficient 2.74 is accurate to a few percent for $1\gtrsim J_z\gtrsim 5\times10^{-3}$), and an average $\bar{l}$ which scales as $J^{-1/3}$ and equals 5.4 for $J\simeq 0.02$. Our various approximations have not seriously biased either result greatly when $J\ll 1$ since the exchange energy will always scale as Jl.

It is very misleading to think of the one-particle density of states found in Ref. 10 as a "band." Although its width is of order $4\sqrt{3}$, states near $-2\sqrt{3}$ have very large (infinite for BR) mass for $J_\perp=0$.

The pair states correspond to two holes connected by a string of reversed spins.[14] Whether binding occurs is subtle as is illustrated by a one-dimensional example[15] where the hopping is nonzero only along the x axis but the exchange is two dimensional and still given by (1) with $J_\perp=0$. A simple but exact calculation shows that there is no binding for $J_z\lesssim 0.2$ and the relevant energy difference for small J_z is large, i.e., $\sim -J_z^{2/3}$, as a result of the antisymmetry. The binding for larger J occurs purely because, when hopping is suppressed, the exchange energy is minimized with the two holes contiguous.

While the single-hole ground state could be found on a set of states $|l\rangle$ corresponding to sums over nonretracing configurations of the string of length l, the fermionic nature of holes dictates a more complex structure for a pair. For the latter we shall construct a set of "string states" labeled by two integers, which satisfies the requirements of statistics and, although not exactly closed under the hopping term in H, provides a good variational basis.

A general state with a pair of holes on the nearest-neighbor sites can be written as ($\boldsymbol{\mu}=\pm\hat{\mathbf{x}},\pm\hat{\mathbf{y}}$)

$$|\chi\rangle\equiv\sum_{\boldsymbol{\mu}}\sum_{\mathbf{r}}e^{i\mathbf{k}\cdot\mathbf{r}}e^{(i/2)\mathbf{k}\cdot\boldsymbol{\mu}}\chi_{\sigma\sigma'}(\boldsymbol{\mu})c_\sigma(\mathbf{r})c_{\sigma'}(\mathbf{r}+\boldsymbol{\mu})|0\rangle, \tag{5}$$

where repeated indices are summed over, $\chi_{\sigma\sigma'}(\boldsymbol{\mu})$ is a form factor that we will determine later, and $|0\rangle$ is some singly occupied ground state. The anticommutation of fermionic operators imposes the constraint

$$\chi_{\sigma'\sigma}(-\boldsymbol{\mu})=-\chi_{\sigma\sigma'}(\boldsymbol{\mu}). \tag{6}$$

The hopping term in Hamiltonian (1) acting on $|\chi\rangle$ yields

$$H_0|\chi\rangle=-\sum_{\boldsymbol{\mu},\mathbf{r}}e^{i\mathbf{k}\cdot\mathbf{r}}e^{(i/2)\mathbf{k}\cdot\boldsymbol{\mu}}\chi_{\sigma\sigma'}(\boldsymbol{\mu})\sum_{\boldsymbol{\tau}\neq-\boldsymbol{\mu}}\{c_\sigma(\mathbf{r})[c_\nu^\dagger(\mathbf{r}+\boldsymbol{\mu})c_{\sigma'}(\mathbf{r}+\boldsymbol{\mu})]c_\nu(\mathbf{r}+\boldsymbol{\mu}+\boldsymbol{\tau})$$
$$+c_\nu(\mathbf{r}-\boldsymbol{\tau})[c_\nu^\dagger(\mathbf{r})c_\sigma(\mathbf{r})]c_{\sigma'}(\mathbf{r}+\boldsymbol{\mu})\}|0\rangle. \tag{7}$$

This state lies in the space of states defined by ($\{\ldots,\ldots\}$ denotes anticommutator)

$$|m,n\rangle\equiv\sum_{\boldsymbol{\mu},\mathbf{r}}e^{i\mathbf{k}\cdot\mathbf{r}}e^{(i/2)\mathbf{k}\cdot\boldsymbol{\mu}}\chi_{\sigma\sigma'}(\boldsymbol{\mu})3^{-(n+m)/2}\sum_{\boldsymbol{\tau}_m\neq-\boldsymbol{\tau}_{m-1}}\cdots\sum_{\boldsymbol{\tau}_1\neq-\boldsymbol{\mu}}\sum_{\boldsymbol{\tau}_n'\neq-\boldsymbol{\tau}_{n-1}'}\cdots\sum_{\boldsymbol{\tau}_1'\neq-\boldsymbol{\mu}}c_{\nu_m}(\mathbf{r}_m)c_{\nu_n'}(\mathbf{r}_n')$$
$$\times\tfrac{1}{2}\{[c_{\nu_m}^\dagger(\mathbf{r}_{m-1})c_{\nu_{m-1}}(\mathbf{r}_{m-1})]\cdots[c_{\nu_1}^\dagger(\mathbf{r})c_\sigma(\mathbf{r})],[c_{\nu_n'}^\dagger(\mathbf{r}_{n-1}')c_{\nu_{n-1}'}(\mathbf{r}_{n-1}')]\cdots[c_{\nu_1'}^\dagger(\mathbf{r}+\boldsymbol{\mu})c_{\sigma'}(\mathbf{r}+\boldsymbol{\mu})]\}|0\rangle, \tag{8}$$

where $\mathbf{r}_j=\mathbf{r}-\sum_{i=1}^j\boldsymbol{\tau}_i$ and $\mathbf{r}_j'=\mathbf{r}+\boldsymbol{\mu}+\sum_{i=1}^j\boldsymbol{\tau}_i'$. It is straightforward to verify from (6) that $|m,n\rangle=|n,m\rangle$. The operator product on the right-hand side of (8) can be associated with a pair of holes connected to the link $(\mathbf{r},\mathbf{r}+\boldsymbol{\mu})$ by nonretracing paths of length m and n.

Generalizing the single-hole case we introduce operators $h_1^\dagger$ and $h_2^\dagger$, $[h_1^\dagger,h_2^\dagger]=0$, which add all allowed links at the ends of the string. Operators h_1,h_2, such that $h_\alpha h_\alpha^\dagger=1$ (for $\alpha=1,2$), then annihilate the end links. Also, $h_\alpha|0,0\rangle=0$

($|0,0\rangle \equiv |\chi\rangle$). The $|m,n\rangle$ states can then be represented as $|m,n\rangle = \frac{1}{2}(h_1^{\dagger m}h_2^{\dagger n} + h_1^{\dagger n}h_2^{\dagger m})|\chi\rangle$. We have $(h_1^\dagger + h_2^\dagger)|m,n\rangle = |m,n+1\rangle + |m+1,n\rangle$ and $(h_1+h_2)|m,n\rangle = |m-1,n\rangle + |m,n-1\rangle$, the latter as long as $m, n \neq 0$. The hopping part of the Hamiltonian is represented symbolically by

$$H_0 = -\sqrt{3}\sum_a (h_a^\dagger + h_a). \tag{9}$$

The representation (9) for H_0 is only as faithful as the labeling of configuration space by walks. Problems arise when the two walks associated with m,n intersect, since as they are retraced by acting with (1), nontrivial commutator terms are generated.[16] (For the Néel ground state, these anomalous components are orthogonal to the $|m,n\rangle$ space.) The $h_a^\dagger$ are nevertheless a convenient bookkeeping device for displaying the lattice walk representation of our states. They also facilitate the construction of a rather nontrivial *first quantitized* version of (8) with proper Fermi statistics. Finally, while $|m,n\rangle$ states for $n+m=$const are not orthogonal, they can be orthogonalized by diagonalization of the operator $h_1h_2^\dagger + h_2h_1^\dagger$.

Proceeding with the calculation, we have

$$\begin{aligned} H|0,0\rangle &= -\sqrt{3}(|0,1\rangle + |1,0\rangle) + \tfrac{7}{4}J_z|0,0\rangle, \\ H|0,n\rangle &= -\sqrt{3}(|0,n+1\rangle + |1,n\rangle + \beta|0,n-1\rangle) + (2+n)J_z|0,n\rangle, \\ H|m,n\rangle &= -\sqrt{3}(|m+1,n\rangle + |m,n+1\rangle + |m-1,n\rangle + |m,n-1\rangle) + (2+n+m)J_z|m,n\rangle. \end{aligned} \tag{10}$$

Only the second line of (10) is nontrivial (β is defined below), since in that case h_a can act directly on the "base" link $(\mathbf{r},\mathbf{r}+\boldsymbol{\mu})$. That is,

$$(h_1+h_2)|0,n\rangle = |0,n-1\rangle + \tfrac{1}{2}(h_1^{\dagger(n-1)} + h_2^{\dagger(n-1)})M_+|\chi\rangle + \tfrac{1}{2}(h_1^{\dagger(n-1)} - h_2^{\dagger(n-1)})M_-|\chi\rangle, \tag{11}$$

where $M_\pm \equiv h_1h_2^\dagger \pm h_2h_1^\dagger$. More explicitly,

$$M_\pm|\chi\rangle = -\tfrac{1}{3}\sum_{\mathbf{r}}\sum_{\boldsymbol{\mu}} e^{i\mathbf{k}\cdot\mathbf{r}}e^{(i/2)\mathbf{k}\cdot\boldsymbol{\mu}} \sum_{\boldsymbol{\tau}\neq-\boldsymbol{\mu}} [\chi_{\sigma'\sigma}(-\boldsymbol{\mu}) \mp \chi_{\sigma\sigma'}(\boldsymbol{\mu})] c_\nu^\dagger(\mathbf{r}+\boldsymbol{\mu}) c_{\sigma'}(\mathbf{r}+\boldsymbol{\mu}) c_\nu(\mathbf{r}-\boldsymbol{\tau}) c_\sigma(\mathbf{r})|0\rangle. \tag{12}$$

Because of the constraint (6), $M_-|\chi\rangle = 0$. Furthermore, when $|0\rangle$ is the Néel ground state, we can evaluate M_+ exactly by noticing that the string of annihilation operators is nonvanishing only for $\sigma' \neq \sigma$ and $\nu \neq \sigma$ because of the staggered spin order in $|0\rangle$. Hence $\nu = \sigma'$.

We now fix β in (10) by choosing χ to satisfy $M_+|\chi\rangle = (\beta-1)|\chi\rangle$. The eigenstates may be labeled by parity which then fixes the spin dependence by (6). There are two states with a k-independent eigenvalue $\beta = \frac{4}{3}$ and hence infinite mass, namely,

$$\chi^{p_1}_{\sigma\sigma'}(\boldsymbol{\mu}) = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}_{\sigma\sigma'} [\sin(k_y/2), -\sin(k_y/2), -\sin(k_x/2), \sin(k_x/2)] \tag{13a}$$

and

$$\chi^{d}_{\sigma\sigma'}(\boldsymbol{\mu}) = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}_{\sigma\sigma'} [\cos(k_y/2), \cos(k_y/2), -\cos(k_x/2), -\cos(k_x/2)], \tag{13b}$$

where the label $\boldsymbol{\mu}$ is arrayed as $(\hat{x}, -\hat{x}, \hat{y}, -\hat{y})$. There are two additional states with a finite mass, viz. (suppressing spin dependence),

$$\chi^s(\boldsymbol{\mu}) = [\cos(k_x/2), \cos(k_x/2), \cos(k_y/2), \cos(k_y/2)], \quad \beta^s = \tfrac{2}{3}[2 - \cos^2(k_x/2) - \cos^2(k_y/2)] \tag{13c}$$

and

$$\chi^{p_2}(\boldsymbol{\mu}) = [\sin(k_x/2), -\sin(k_x/2), \sin(k_y/2), -\sin(k_y/2)], \quad \beta^{p_2} = \tfrac{2}{3}[2 - \sin^2(k_x/2) - \sin^2(k_y/2)].$$

By combining the various definitions one verifies that the transformation $\mathbf{k} \to \mathbf{k} + (\pi,\pi)$ interchanges χ^{p_1} with χ^d, and χ^{p_2} with χ^s, and thus "singlet" with "triplet." It is important to note that the "spin" is a property of the entire state and that it does not reside on the ends but is rather associated with the sublattices.

By diagonalizing (10) numerically, we find that within the BR approximation only the p and d states bind with an energy $\sim \frac{1}{2}J_z$ for $J_z \simeq 1$ which decreases to zero by $J_z \simeq 2\times10^{-3}$. Hence, the $\mathcal{O}(J^{2/3})$ terms from (4) nearly cancel and these are the only ones whose sign will not change when $J_\perp = J_z$. (Within a Born-Oppenheimer approach, the time for a hole to run over a walk of length $l \sim J^{-1/3}$ is a factor $J^{1/3}$ shorter than the time, $J^{-2/3}$, to flip at least one spin on the walk.) We are thus uncertain whether the problem of physical interest exhibits binding. The inverse mass of χ^{p_2} from (10) is positive

and to within 20% varies as $J^{2/3}$ for $1 \gtrsim J \gtrsim 0.04$ and as J for $0.04 \gtrsim J \gtrsim 2\times 10^{-3}$.

It is instructive to redo the preceding calculation [from (5) onward] for bosons which reproduces (10) with $\beta=2$. Two bosons do bind with an energy $\sim(1.2$-$1.7)J^{2/3}$. The BR approximation, though exact for large coordination number, includes walks for which the two holes occupy the same site. We corrected this problem for bosons by adding a hard core and constructing a variational wave function which assigned a common amplitude $\psi_l(\mathbf{r})$ to all walks with l links and end-to-end distance $\mathbf{r}$.[17] The binding persisted but with a much smaller coefficient of $J^{2/3}$. The effects of the hard core for fermions should be less pronounced since the antisymmetry is already present.

The subtle structure of the spin state (8) is nicely illustrated by our repeating the variational calculation for fermions in a p state with $\psi_l(x,y)=-\psi_l(x,-y)$.[17] The binding energy, surprisingly, is strongly negative and scales roughly as $-J^{1/3}$. It is apparently a very poor approximation to treat the many-particle wave function by retaining only the end-to-end vector which forces a common nodal line in $\mathbf{r}$.

We now summarize our conclusions. First, the Nagaoka mechanism and ferromagnetic polarons are only important for J/w very small, i.e., $\lesssim 5\times 10^{-3}$. The observation of antiferromagnetism argues in favor of $Jw \lesssim 1$ and thus the BR picture of "band"-narrowing effects may be appropriate. In this range, the kinetic energy of holes can be evaluated by use of the string representation of the configuration space. First-quantized spin and space wave functions do not factorize. Also, surprisingly, the mass of a pair state may be infinite.

Finally, as yet we do not have a conclusion concerning superconductivity in the Hubbard model since only two holes were considered and the binding that we find is of the same order as $J_\perp$ exchange which was not included. The latter will introduce coupling between pairs and unpaired fermions, but can be considered perturbatively. On the other hand, the string states as defined in Eq. (8) can be generalized to many particles.

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[12]See Nagoaka, Ref. 11.

[13]Although (4) can be solved analytically in the small $-J$ limit as a one-dimensional "potential" problem on the half-line $l \geq 0$, it is not possible in more than one physical dimension to write any simple Schrödinger equation in terms of the hole position, $\mathbf{r}$, alone. In particular $H=-2\sqrt{3}w-w\nabla^2+Jr$ is incorrect since for a random walk $r\sim l^{1/2}$ and the potential energy is linear in l, not r.

[14]J. E. Hirsch, Phys. Rev. Lett. **59**, 228 (1987); Y. Takahashi, Z. Phys. B **67**, 503 (1987). Hirsch only calculates for large J while Takahashi's approximation for one and two holes is explicitly inconsistent for small J.

[15]We thank W. F. Brinkman for this example.

[16]D. Frenkel, private communication.

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[18]Similar conclusions have been reached by S. Trugman.