Is the π Particle Responsible for the 41 meV Peak in $YBa_2Cu_3O_7$?

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It is argued that there is no low-energy resonance associated with the π operators introduced by Demler and Zhang. This implies that the Hubbard model does not possess an approximate SO(5) symmetry generated by these operators. Recent finite-size studies are reinterpreted accordingly. [S0031-9007(97)04746-7]

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Two years ago, Demler and Zhang [1] proposed a new collective mode for the positive U Hubbard model, the so-called π resonance. They calculated the energy of this mode within the T-matrix approximation, and found

$$\omega_0 \approx \frac{J}{2}(1 - n) - 2\mu\,, \tag{1}$$

where $J \approx t^2/4U$ is the antiferromagnetic coupling between nearest neighbors, $n < 1$ is the electron density, and μ is the "chemical potential measured from half filling" [2] (by which, I presume, they mean the difference between the chemical potential at electron density n and at half filling ($n = 1$), as there is no meaning in measuring a chemical potential from a particular reference point). They then interpreted the sharp magnetic resonance peak at 41 meV observed by inelastic neutron scattering [3–6] in the superconducting phase of $YBa_2Cu_3O_7$ in terms of this resonance, and identified ω_0 at the appropriate electron density with the observed resonance frequency.

In order to explain why I do not agree with the reasoning leading to this identification, let me first review the essential steps in their derivation [1]. The π operator is given by

$$\pi_d^\dagger = \sum_k (\cos k_x - \cos k_y) c^\dagger_{\mathbf{k}+\mathbf{Q}\uparrow} c^\dagger_{-\mathbf{k}\uparrow} \qquad \mathbf{Q} \equiv (\pi, \pi) \tag{2}$$

and is argued to be an approximate eigenoperator of the t-J-U-μ Hamiltonian

$$H = -t \sum_{\langle ij\rangle\sigma} c^\dagger_{i\sigma} c_{j\sigma} + \frac{J}{2} \sum_{\langle ij\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + U \sum_i n_{i\uparrow} n_{i\downarrow} - \mu \sum_{i\sigma} c^\dagger_{i\sigma} c_{i\sigma}\,, \tag{3}$$

where electron density is determined by the chemical potential. This property they deduce from the commutator

$$[H, \pi_d^\dagger] \approx 2(Un_\downarrow - \mu)\pi_d^\dagger + \text{a term linear in } J\,, \tag{4}$$

where $n_\downarrow = \frac{1}{N}\sum n_{i\downarrow}$ is the average density of down-spin electrons. Careful scrutiny [1] then reveals that the term linear in J can be approximated by $\frac{J}{2}(1 - n)\pi_d^\dagger$. Demler and Zhang obtained (1) from (4) by asserting that "the leading contribution to the chemical potential is [in the large U limit] given by $Un/2$," and that the first term in (4) "cancels in the leading order in U and reaches a finite limit as $U \to \infty$" [1]. With this I disagree.

First of all, let us go back and find the source of the mistake. In an earlier paper [7], Zhang observes that the particle-hole symmetry of the Hubbard model implies

$$\mu(n) = U - \mu(2 - n) \tag{5}$$

and infers that $\mu = U/2$ at half filling. This is not unambiguously correct at $T = 0$ as the chemical potential of the positive-U Hubbard model is (in general) discontinuous at $n = 1$ [8], with a discontinuity which becomes more and more pronounced as U tends to infinity:

$$\mu(1 + \varepsilon) - \mu(1 - \varepsilon) = U - 2\mu(1 - \varepsilon)\,, \tag{6}$$

where ε is a positive infinitesimal and $\mu(1 - \varepsilon)$ approaches a constant as $U \to \infty$.

The origin of this discontinuity is best understood in the same limit: Up to half filling, the ground state is to zeroth order composed of configurations without doubly occupied sites; doubly occupied sites enter only through virtual processes. The chemical potential in this regime will, therefore, approach a (density dependent) constant in the large U limit. Once we exceed half filling, however, the ground state must necessarily contain as many doubly occupied sites as there are electrons exceeding half filling, with a zeroth order energy cost of U associated with each. The leading contribution to the chemical potential in that regime is thus given by U.

The chemical potential for the large U Hubbard model with $n < 1$ is consequently much smaller than $Un/2$, and the energy of the π particle

$$\omega_0 \approx 2Un_\downarrow - 2\mu(n) + \frac{J}{2}(1 - n) \tag{7}$$

is of order 2 eV. This particle cannot possibly account for the magnetic resonance peak observed at 41 meV.

The physical reason why the energy of the π excitation is of order U is trivial. The π operator places two up-spin electrons with a center of mass momentum of (π, π) on two neighboring lattice sites, with an amplitude insensitive to whether these sites are occupied by holes (that is, unoccupied) or by down-spin electrons. (If one of the sites is occupied by an up-spin electron, the amplitude vanishes due to the Pauli principle.) Therefore, as the π operator acts on the ground state for the Hubbard model

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with electron density $n < 1$, there is a large amplitude to find doubly occupied sites. Those give rise to the first term in (7).

Unlike the η excitation proposed by Yang [9] and further elaborated by Zhang [10], the π excitation is hardly a resonance. The η operator places a pair of electrons with opposite spin on the same lattice site, and as the Pauli principle excludes the remaining electrons from this site, there is no scattering between this pair and any of the other electrons by the Hubbard interaction U. The situation is different for the π operator, which places a pair of up-spin electrons on neighboring sites, and thereby allows U to scatter any down-spin electron in the liquid off this pair. This scattering induces an almost instantaneous decay of the π excitation, with a decay rate of order U.

The fact that the π operators are not approximate eigenoperators of the Hubbard Hamiltonian implies that they do not generate an approximate SO(5) symmetry for the Hubbard model [11].

In light of these considerations, it comes as a surprise when Meixner *et al.* [2] present finite-size studies which "verify that the π operators are approximate eigenoperators of the Hubbard model" and "lend support to an interpretation of the recent neutron scattering peaks" in terms of the π excitation. In Figs. 1(b), 1(f), 2(b), and 2(f) of [2], they compare the π-π correlation function

$$\pi_d^\dagger(\omega) = -\frac{1}{\pi} \times \mathrm{Im}\langle\Psi_0^{N-2}|\pi_d \frac{1}{\omega - H + E_0^N + i\varepsilon} \pi_d^\dagger|\Psi_0^{N-2}\rangle \tag{8}$$

with the dynamical spin-spin correlation function

$$\chi_{\mathbf{Q}}^+(\omega) = -\frac{1}{\pi} \times \mathrm{Im}\langle\Psi_0^N|S_{\mathbf{Q}}^- \frac{1}{\omega - H + E_0^N + i\varepsilon} S_{\mathbf{Q}}^+|\Psi_0^N\rangle, \tag{9}$$

where Im denotes the imaginary part and H the t-U Hubbard Hamiltonian (which does not contain a chemical potential term). The π_d operator is as defined in (2), and

$$S_{\mathbf{Q}}^+ = \sum_{\mathbf{k}} c_{\mathbf{k}+\mathbf{Q}\uparrow}^\dagger c_{\mathbf{k}\downarrow}, \qquad S_{\mathbf{Q}}^- = S_{\mathbf{Q}}^{+\dagger}, \qquad \mathbf{Q} \equiv (\pi, \pi) \tag{10}$$

is the spin density wave operator. For a ten site Hubbard cluster with $N = 10$, they find that both of these correlation functions (as well as a mixed correlation function involving both π_d and $S_{\mathbf{Q}}^+$) have sharp resonance peaks at the same energy. They conclude that this "clearly demonstrates that the peak in the spin correlation function is due to a particle-particle intermediate state $\pi_d^\dagger|\Psi_0^{N-2}\rangle$" [2].

In order to explain why I disagree, let me first review the reasoning leading to this interpretation [10]. For this purpose, let us assume that the π operator was an exact eigenoperator of H,

$$[H, \pi_d^\dagger] = \omega_\pi \pi_d^\dagger. \tag{11}$$

Then the only intermediate state which would contribute to the π-π correlation function defined in (8),

$$\pi_d^+(\omega) = -\frac{1}{\pi} \mathrm{Im} \sum_n \frac{|\langle\Psi_n^N|\pi_d^\dagger|\Psi_0^{N-2}\rangle|^2}{\omega - E_n^N + E_0^N + i\varepsilon}, \tag{12}$$

where the sum extends over all of the excited states of H, would be

$$|\Psi_\pi^N\rangle = \mathcal{N}_\pi \pi_d^\dagger|\Psi_0^{N-2}\rangle, \tag{13}$$

where $\mathcal{N}_\pi$ is a normalization constant; we choose $\mathcal{N}_\pi$ real. Therefore, (12) would reduce to

$$\pi_d^+(\omega) = \frac{1}{\mathcal{N}_\pi^2} \delta(\omega - \omega_\pi). \tag{14}$$

In the presence of d-wave superconductivity, this resonance would also manifest itself in the spin-spin correlation function defined in (9),

$$\chi_{\mathbf{Q}}^+(\omega) = -\frac{1}{\pi} \mathrm{Im} \sum_n \frac{|\langle\Psi_n^N|S_{\mathbf{Q}}^+|\Psi_0^N\rangle|^2}{\omega - E_n^N + E_0^N + i\varepsilon}, \tag{15}$$

as the exact excited state $|\Psi_\pi^N\rangle$ would yield a singularity at ω_π:

$$\begin{aligned}\chi_{\mathbf{Q}}^+(\omega) &= -\frac{1}{\pi} \mathcal{N}_\pi^2 \mathrm{Im} \frac{|\langle\Psi_0^{N-2}|\pi_d S_{\mathbf{Q}}^+|\Psi_0^N\rangle|^2}{\omega - \omega_\pi + i\varepsilon} \\ &\quad + \text{regular at } \omega_\pi \\ &= 4\mathcal{N}_\pi^2 |\langle\Psi_0^{N-2}|\Delta_d|\Psi_0^N\rangle|^2 \delta(\omega - \omega_\pi) \\ &\quad + \text{regular at } \omega_\pi,\end{aligned} \tag{16}$$

where we have used

$$\frac{1}{2}[\pi_d, S_{\mathbf{Q}}^+] = \sum_{\mathbf{k}} (\cos k_x - \cos k_y) c_{\mathbf{k}\uparrow} c_{-\mathbf{k}\downarrow} \equiv \Delta_d. \tag{17}$$

The expectation value $\langle\Psi_0^{N-2}|\Delta_d|\Psi_0^N\rangle$ is often used as a d-wave superconducting order parameter.

This effect—the manifestation of a particle-particle resonance in the particle-hole channel in the presence of superconductivity—persists even if $|\Psi_\pi^N\rangle$ is only an approximate eigenstate. Hence, their interpretation.

These authors, however, seem to have overlooked that this argument can be run backwards. In the presence of superconductivity, a resonance in the particle-hole channel will manifest itself in the particle-particle channel. In particular, if the one-magnon state

$$|\Psi_S^N\rangle = \mathcal{N}_S S_{\mathbf{Q}}^+|\Psi_0^N\rangle \tag{18}$$

was an exact eigenstate of H,

$$[H, S_{\mathbf{Q}}^+] = \omega_S S_{\mathbf{Q}}^+, \tag{19}$$

this resonance would manifest itself in the π-π correlation function:

$$\pi_d^+(\omega) = 4\mathcal{N}_S^2 |\langle \Psi_0^{N-2} | \Delta_d | \Psi_0^N \rangle|^2 \delta(\omega - \omega_S) + \text{regular at } \omega_S . \quad (20)$$

Again, the effect persists even if $|\Psi_S^N\rangle$ is only an approximate eigenstate. This is how I would interpret the numerical studies by Meixner *et al.* [2]; a magnon manifests itself in the particle-particle channel.

In fact, a simple *gedanken experiment* can decide between the two interpretations. Suppose we destroy the d-wave superconductivity in the ten site Hubbard cluster by adding a nearest-neighbor repulsion of order $U' = 4t^2/U$. Then the resonances in the two different channels no longer have to coincide in frequency; one of them may be shifted, or might even disappear. Since the resonances in the spin-spin correlation function $\chi_Q^+(\omega)$ shown in Figs. 1(f) and 2(f) of Ref. [2] occur at an electron density $n = 1$ (that is, one electron per site), a nearest-neighbor repulsion $U' \ll U$ would induce only a higher order correction; it would not affect the one-magnon resonance significantly. The nearest-neighbor repulsion, however, would be likely to affect the resonance in the π-π correlation function $\pi_d^+(\omega)$ at $n = 0.8$ shown in Figs. 1(b) and 2(b) of [2]. Hence, my interpretation.

Finally, I wish to offer an interpretation for the low-energy resonances observed by Meixner *et al.* [2] in $\pi_d^+(\omega)$ at $n = 0.6$, a density at which the d-wave superconducting correlations are no longer present [Figs. 1(a) and 2(a) of [2]].

The fact that the $\pi_d^\dagger$ operator is not an approximate eigenoperator of the Hubbard model does not imply that its projection onto a specific supspace cannot be an approximate eigenoperator. In particular, the problem with the doubly occupied sites exposed above can be circumvented if one sandwiches the $\pi_d^\dagger$ operator in between two Gutzwiller projectors

$$\mathcal{P}_G \equiv \prod_i (1 - n_{i\uparrow} n_{i\downarrow}) \quad (21)$$

and considers the resulting operator

$$\varpi_d^\dagger = \mathcal{P}_G \pi_d^\dagger \mathcal{P}_G \quad (22)$$

as a candidate for an approximate eigenoperator of the t-J model, which is obtained from the Hubbard model (3) by taking the limit $U \to \infty$. Since the ϖ excitation

$$|\Psi_\varpi^N\rangle = \mathcal{N}_\varpi \varpi_d^\dagger |\Psi_0^{N-2}\rangle \quad (23)$$

has a finite overlap with the π excitation (13) in the thermodynamic limit, I conjecture that the intermediate state (23) is responsible for the sharp resonance peak observed by Meixner *et al.* [2] in the π-π correlation function $\pi_d^\dagger(\omega)$ for $n = 0.6$.

The ϖ excitation introduced above, however, has a number of problems:

(a) Since the Gutzwiller projector $\mathcal{P}_G$ does not commute with the hopping term in (3), $|\Psi_\varpi^N\rangle$ is not an eigenstate of the kinetic part of the t-J (or Hubbard) Hamiltonian.

(b) The ϖ operators do not satisfy all of the commutation relations of the SO(5) algebra [11]. They can, however, be used to rotate an antiferromagnetic order operator into a d-wave superconducting order operator; since $\mathcal{P}_G$ commutes with the spin density wave operator $S_\mathbf{Q}^+$, the commutator (17) reduces to

$$\frac{1}{2}[\varpi_d, S_\mathbf{Q}^+] = \mathcal{P}_G \Delta_d \mathcal{P}_G . \quad (24)$$

Since the t-J Hamiltonian does not allow for doubly occupied sites, the Gutzwiller projectors in (24) have no effect on the superconducting order parameter $\langle \Psi_0^{N-2} | \Delta_d | \Psi_0^N \rangle$.

(c) Unpublished finite-size studies by Meixner and Hanke [12] show a sharp peak in the dynamical spin-spin correlation functions $\chi_\mathbf{Q}^+(\omega)$ at $n = 0.8$, which occurs at a slightly lower energy than the resonance in $\pi_d^+(\omega)$ at $n = 0.6$ shown in Figs. 1(a) and 2(a) of [2]. (The intermediate states in both correlation functions contain the same number of particles). This result constitutes significant evidence against an interpretation of the magnetic resonance peak observed in superconducting $YBa_2Cu_3O_7$ in terms of the ϖ excitation, or a resonance in the particle-particle channel in general.

(d) One of the side effects of the on-site Hubbard or t-J model descriptions of the CuO planes in high-T_c superconductors is that the antiferromagnetic exchange interaction yields an effective attraction between holes on nearest-neighbor sites, which is of order $J\langle \mathbf{S}_i \cdot \mathbf{S}_{i+1} \rangle$. In the CuO planes, this attraction is overcompensated by the Coulomb repulsion between the holes; depending on the details of the screening by the highly polarizable O atoms in between the Cu atoms, this repulsion is by a factor of at least 10 larger than the magnetic attraction.

I conjecture that most of the spectral weight in the low-energy resonance peaks observed by Meixner *et al.* [2] in the π-π correlation function $\pi_d^+(\omega)$ will disappear at all densities once a nearest-neighbor repulsion of order $J = 4t^2/U$ is introduced to compensate for this "unphysical" magnetic attraction.

Apart from the detailed considerations presented above, the π particle proposal does not explain why the neutron resonance peak is observed only in a bilayer system, and there only in the acoustical channel ($Q_\perp = \pi$); according to this proposal, a similar resonance should occur in the optical channel ($Q_\perp = 0$) as well.

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Note added.—The criticism initiated by this Letter has meanwhile been strengthened and deepened by Baskaran and Anderson [13].

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[8] The reverse of this statement is, of course, true: If we choose $\mu = U/2$, the Hubbard cluster will be half filled, as it will be for any choice of μ between $\mu(1 - \varepsilon)$ and $\mu(1 + \varepsilon)$. The chemical potential plotted as a function of the electron density is discontinuous at $n = 1$, while the electron density plotted as a function of the chemical potential has a plateau between $\mu(1 - \varepsilon)$ and $\mu(1 + \varepsilon)$. The fact that this discontinuity gets smoothed out at finite T, which indeed yields the "midgap" value $\mu = U/2$ at exactly $n = 1$, does not affect the validity of the argument presented in the text.

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