Theory of the Resonant Neutron Scattering of High-T_c Superconductors

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Recent polarized neutron scattering experiments on $YBa_2Cu_3O_7$ have revealed a sharp spectral peak at the (π, π) in reciprocal lattice centered around the energy transfer of 41 meV. We offer a theoretical explanation of this remarkable experiment in terms of a new collective mode in the particle particle channel of the Hubbard model. This collective mode yields valuable information about the symmetry of the superconducting gap.

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Recently, both unpolarized and polarized neutron scattering experiments have been performed on the $YBa_2Cu_3O_7$ high-T_c superconductors [1–3]. In particular, the polarized neutron experiment [2] shows an extremely sharp spectral feature in the spin flip channel. This feature is centered around (π, π) in reciprocal space, and peaked at 41 meV with a width narrower than the instrumental resolution. This feature also has an interesting temperature dependence. In the experiment by Mook *et al.* [2], while it exists above the superconducting transition temperature of $T_c = 92.4$ K, its intensity scales like the superfluid density below the transition. More recently, Fong *et al.* [3] performed detailed spin unpolarized neutron experiments with a careful subtraction of the phonon background. They found that the 41 meV mode disappears above the superconducting transition temperature.

In this Letter, we offer a theoretical explanation of this remarkable experiment. We first show that for a general class of tight binding Hamiltonian, including the Hubbard and the t-J model, there exist well-defined collective modes in the particle particle channel centered around momentum (π, π). The spin quantum number of this excitation can be either a singlet or a triplet. The singlet excitation has been discussed by Yang [4] and one of us [5], and is in fact an exact eigenstate of the Hubbard model. Normally, collective excitations in the particle particle channel are inaccessible experimentally. However, one of us [5] argued that if the ground state of the model in consideration is superconducting, one can couple to it through a particle hole excitation, because the BCS condensate is a coherent mixture of particles and holes. Based on this consideration, one of us [5] predicted a possible new collective mode of the high-T_c superconductors. It is a spin singlet excitation peaked at (π, π), has a well-defined energy of $U - 2\mu$, and its intensity scales like the superfluid density. Possibly because it is hard to distinguish from other excitations in the system, this mode has not yet been detected experimentally.

However, the basic arguments can be easily generalized from the singlet to the triplet case. Besides the above mentioned collective mode in the singlet particle particle channel, there also exists a well-defined collective mode in the triplet channel near total momentum (π, π). This is true for a large class of tight binding models, such as the Hubbard or the t-J model. The energy spectrum of a noninteracting pair of particles or holes generally consists of a continuum labeled by their relative momentum. However, for tight binding models, this continuum collapses to a point where the total momentum of the pair is (π, π) (see Fig. 1).

A triplet pair generally has a repulsive interaction when placed on the neighboring site. Because of the collapse of the particle particle continuum, this repulsive interaction leads to an *antibound* state near total momentum $(\pi, \pi, ...)$ *in any space dimensions* (see Fig. 1). This antibound triplet state manifests itself as a collective excitation of the many-body system, or as a pole in Green's function of the particle particle channel. Most physical probes do not couple to this channel. However, if the model in consideration is superconducting (which is an assumption of our theory), then a spin flip scattering of

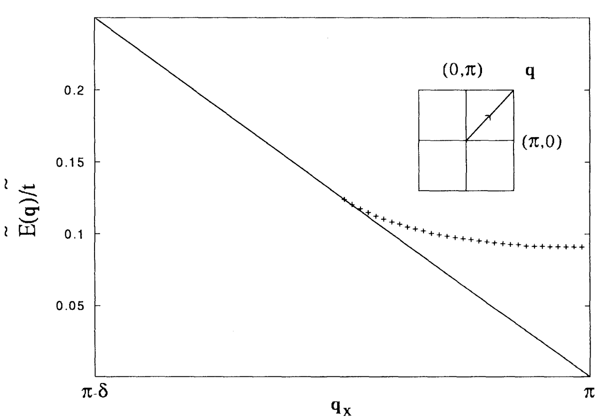

FIG. 1. The energy spectrum along the (π, π) direction. The dots correspond to the antibonding state and the solid line is the edge of the continuous spectrum. Here the numerical calculations were done for $J = \tilde{t}$ and $n = 0.85$. As δ we denoted $\pi/50$.

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the neutron can couple directly to the triplet particle particle excitation. Physically, this process is nothing but the spin flip scattering of a Cooper pair in the BCS condensate. Since the Cooper pair in the high-T_c materials is a spin singlet with total momentum zero [6], therefore a spin flip scattering of the Cooper pair necessarily creates a triplet state. The orbital angular momentum or the parity selection rule forbids such a transition if the momentum loss of the neutron is zero, on the other hand, it can be shown simply that the matrix element of this coupling is maximal when the momentum loss of the neutron is at (π, π).

Based on the above reasoning, we shall interpret the sharp spectral feature observed in the polarized neutron experiment in terms of the triplet collective mode in the particle particle channel. We begin with some analytic calculations of the properties of this collective mode and its contribution to the dynamical spin-spin correlation function. Subsequently, we compare our predictions with a number of characteristic features of the resonance observed in experiments.

Our antibound state is different from the excitons of a superconductor considered by Bardasis and Schrieffer [7]. These excitons form because of an *attractive* potential in a given angular momentum channel, and they exist inside the superconducting gap and near total momentum $q = 0$. The energy of the exciton mode has a temperature dependence similar to the superconducting gap, and would approach to zero near the superconducting transition, whereas the energy of our mode does not depend significantly on temperature. Our model is also different from the magnetic susceptibility in the superconducting state computed using the random phase approximation [8], since it involves multiple scattering in the particle hole channel. More recently, Bulut and Scalapino [9] considered a model of the bilayer superconductors and argued that a dynamic nesting from the bonding to antibonding Fermi surface could give rise to a collective resonance at (π, π). Our main difference lies in the fact that their resonance is in the particle hole channel and as such it can exist well above T_c. The bilayer band structure plays a crucial role in their model, while it is irrelevant in our case.

We consider the following model defined on a two-dimensional square lattice:

$$\mathcal{H} = -t\sum_{\langle ij\rangle} c_{i\sigma}^{\dagger} c_{j\sigma} + J\sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j + U\sum_i n_{i\uparrow} n_{i\downarrow} - \mu \sum_i c_{i\sigma}^{\dagger} c_{i\sigma}. \quad (1)$$

In the limit $U \to \infty$, we recover the t-J model. On the other hand, if we keep U small, we can regard the above model as an effective Hamiltonian of the weak coupling Hubbard model, where the J term arises from the paramagnon interaction on the nearest neighbor sites. To study the spectrum in the triplet particle particle channel, we consider the operator

$$O_q^{\dagger} = \sum_p f_q(p) c_{p+q\uparrow}^{\dagger} c_{-p\uparrow}^{\dagger}, \quad (2)$$

which describes a pair of particles with center of mass momentum q and relative momentum p. The equation of motion for this operator is given by its commutator with the Hamiltonian, $[\mathcal{H}, O_q^{\dagger}]$. If we factorize the resulting commutator in terms of $O_q^{\dagger}$ and the expectation values of the density $n_{k\sigma}$, we obtain

$$[\mathcal{H}, O_q^{\dagger}] = \sum_p (\epsilon_{p+q} + \epsilon_{-p}) f_q(p) c_{p+q\uparrow}^{\dagger} c_{-p\uparrow}^{\dagger} + 2(Un_{\downarrow} - \mu) O_q^{\dagger} + \frac{J}{8} \sum_p c_{p+q\uparrow}^{\dagger} c_{-p\uparrow}^{\dagger} \frac{1}{N} \sum_{p'} f_q(p') \times [\eta(p - p') - \eta(p + p' + q)](1 - n_{p'+q\uparrow} - n_{-p'\uparrow}) - \frac{3J}{8} \sum_p f_q(p) c_{p+q\uparrow}^{\dagger} c_{-p\uparrow}^{\dagger} \times \frac{1}{N} \sum_{p'} n_{p'} [\eta(p + p') + \eta(p' - p - q)], \quad (3)$$

where $n_{\sigma} = \frac{1}{N}\sum_k n_{k\sigma}$ is the average density of electrons with spin σ, N is the total number of lattice sites, and $\eta(p) = \sum_{\vec{a}} \exp[i\vec{p} \cdot \vec{a}]$ is a geometrical factor coming from the summation over the nearest neighbors. In this equation, the first term describes the kinetic energy of the pair of particles in consideration. For tight binding models with nearest neighbor hopping, $\epsilon_k = -2t(\cos k_x + \cos k_y)$. In this case, the kinetic energy of the pair vanishes when the total momentum $q = (\pi, \pi)$. The second term describes the Hartree interaction of the spin up pair with the average density of the down spins in the background and the chemical potential energy of adding a pair of particles. In the large U limit, the leading contribution to the chemical potential is given by $Un/2$. Therefore, the second term cancels in the leading order in U and reaches a finite limit as $U \to \infty$. The third term gives the multiple scattering of the particles with each other on a restricted phase space due to the filled Fermi sea. The last term describes the Fock self-energy of the quasiparticles. The method of factorizing the operator equation of motion is equivalent to the T matrix approximation in the diagrammatic calculations; it is exact in the low density limit. The collective mode for the triplet particle particle excitation is obtained by equating the right hand side of

(3) to $E_q O_q^\dagger$, the resulting eigenvalue equation is given by

$$\det \left\| \frac{J}{2} \frac{1}{N} \sum_p \frac{\sin(p_\alpha)\sin(p_\beta) z_{pq}}{\tilde{E} - \Omega_{pq}} - \delta_{\alpha\beta} \right\| = 0, \quad (4)$$

with α and β being x or y, $z_{pq} = 1 - n_{q/2-p} - n_{q/2+p}$ and $\Omega_{pq} = \epsilon_{q/2-p} + \epsilon_{q/2+p}$.

It is straightforward to see that the last equation has two discrete eigenvalues, they correspond to the bonding and antibonding wave functions $f_q^{(\pm)}(p - \frac{q}{2}) \propto \sin(p_x) \pm \sin(p_y)$. It turns out that the bonding combination is very close to continuum and this mode exists only in a very small region of the q space. So, in the future, we will consider only the antibonding wave function whose energy is given by

$$1 = \frac{J}{N} \sum_p \frac{\sin^2(p_x)(1 - n_{q/2-p\uparrow} - n_{q/2+p\uparrow})}{\tilde{E}_q^- + 4\tilde{t}[\cos(p_x)\cos(\frac{q_x}{2}) + \cos(p_y)\cos(\frac{q_y}{2})]}, \quad (5)$$

where $\tilde{t} = t + (3J/4N)\sum_p n_{p\downarrow}\cos(p_x)$ and $E_q = \tilde{E}_q + 2(Un_\downarrow - \mu)$. The above eigenvalue equation can easily be solved numerically, and the dispersion of the collective mode is shown in Fig. 1. $\tilde{t}$ is basically the renormalized hopping matrix element of the quasiparticle. The range in momentum space over which the collective mode exists depends on the ratio of $J/\tilde{t}$. A number of numerical calculations indicate that the quasiparticle bandwidth is of the order of J [10]. Here we take a semiphenomenological approach and choose $\tilde{t}$ rather than t as a free parameter. For a ratio of $J/\tilde{t} = 1$, and $n = 0.85$, we see that the collective mode exists over $\pi/50$ of the momentum space. At $q = (\pi, \pi)$ the energy of the mode is

$$\tilde{E}^- = \frac{J}{2}\left(1 - \frac{2}{N}\sum_p n_p \cos^2 p_x\right). \quad (6)$$

In the experiments with the polarized neutron scattering one measures the dynamic spin-spin correlation function

$$S(q,\omega) = \sum_n |\langle n | S_q^\dagger | 0\rangle|^2 \delta(\omega - \omega_{n0}), \quad (7)$$

where $|0\rangle$ and $|n\rangle$ are the ground and excited states of the system and $S_q^\dagger = \sum_p c_{p+q\uparrow}^\dagger c_{p\downarrow}$. Using the operator equation $[\mathcal{H}, O_{q,\alpha}^\dagger] = E_q^\alpha O_{q,\alpha}^\dagger$ we can construct a class of approximate excited states of the Hubbard Hamiltonian as $|n\rangle = \frac{1}{\mathcal{N}_q} O_{q,\alpha}^\dagger |0\rangle$, where $1/\mathcal{N}_q$ is a normalization factor. The same operator equation for $O_{q,\alpha}$ shows that $O_{q,\alpha}|0\rangle = 0$ when the system is less than half filled. With these relations, we can calculate the contribution of this approximate eigenstate to $S(q,\omega)$ at zero temperature:

$$\begin{aligned} S_0(q,\omega) &= \frac{1}{\mathcal{N}_q^2}\langle 0 | O_q^- S_q^\dagger | 0\rangle|^2 \delta(\omega - E_q^-) + \sum_{n'} \\ &= \frac{1}{\mathcal{N}_q^2}\langle 0 | [O_q^-, S_q^\dagger] | 0\rangle|^2 \delta(\omega - E_q^-) + \sum_{n'}, \end{aligned} \quad (8)$$

where $\sum_{n'}$ denotes the contribution from states other than $O_q^{-\dagger}|0\rangle$, and we used the fact that $O_{q,\alpha}|0\rangle = 0$ to replace the product of two operators by their commutator. Evaluating the commutator we find the contribution of our collective mode to the density density correlation function:

$$\begin{aligned} S_0(q,\omega) &= \frac{1}{\mathcal{N}_q^2}\left| \langle 0 | 2\sum_p f_q^{(-)}(p) c_{-p\uparrow} c_{p\downarrow} | 0\rangle \right|^2 \delta(\omega - E_q^-) \\ &= 2\frac{|\sum_p f_q^{(-)}(p)\Delta_p/2E_p|^2}{\sum_p |f_q^{(-)}(p)|^2(1 - n_{p\uparrow} - n_{p+q\uparrow})} \delta(\omega - E_q^-). \end{aligned} \quad (9)$$

The overlap matrix element is finite for d-wave pairing in the ground state only. For the momentum transfer of $Q = (\pi, \pi)$ the wave functions have an extremely simple form $f_Q^{(-)}(p) = \cos(p_x) - \cos(p_y)$, and one can see that the numerator of (9) reduces to the BCS self-consistency equation. Also, close to half filling the denominator of (9) is equal to $\frac{1}{2}(M - N)/M$, where M is the number of sites and N is the number of electrons. Finally we have

$$\begin{aligned} S(Q,\omega,T) = \frac{16}{9}\frac{\Delta_0^2}{J^2}\frac{M}{M-N}\left[1 + n_B\left(\frac{\omega}{T}\right)\right] \\ \times \delta(\omega - E_0), \end{aligned} \quad (10)$$

where the Boltzmann factor takes care of the finite temperature in spin-spin correlation functions. One can easily see, though, that the main temperature dependence of $S(q,\omega,T)$ comes from $\Delta_0(T)$. The spectral intensity is simply proportional to the BCS order parameter or the superfluid density. This is so because the BCS order parameter provides the coupling from the particle hole channel to the particle particle channel [5]. This extra spectral weight at energy $E_{q,\alpha}$ is transferred from the low energy sector, since the singlet BCS pairing removes the low energy spin fluctuation. Our theory is consistent with the fact that the 41 meV peak intensity seems to scale with the superfluid density below T_c, in agreement with the experimental results obtained by Fong *et al.* [3]. We can see that expression (10) gives a linear dependence of the intensity of the peak on doping. It comes from the proportionality of the order parameter to the density of holes in the t-J model and an extra factor $M/(M - N)$ due to normalization.

We also note that our calculation is not fully self-consistent, since the factorization of the equations of motion is taken with respect to the normal state $|0\rangle$.

We have performed the fully self-consistent calculation using the equations of motion method first developed by Anderson [11], where all expectation values are taken with respect to the superconducting state. We find the collective mode still exists and is not changed significantly from the simple calculation presented here. The details of the fully self-consistent calculation will be presented in the longer version of this paper [12].

All the above discussions were restricted to the two-dimensional CuO plane, which we model by the two-dimensional Hubbard model. However, it can be simply generalized to three dimensions. If one takes a three-dimensional Hubbard model, one finds that the collective mode exists near (π, π, π) rather than $(\pi, \pi, 0)$. This is consistent with the experiment where the third component of the momentum transfer is also π.

The fact that the particle particle collective mode always exists at $(\pi, \pi, \ldots)$ is a special property of the tight binding model on a bipartite lattice. How would a next-nearest neighbor hopping term change the results? In this case, Eq. (4) still holds but with the dispersion relation now given by

$$\epsilon_k = -2\tilde{t}(\cos k_x + \cos k_y) - 2t'[\cos(k_x + k_y) + \cos(k_x - k_y)],$$

where t' denotes the amplitude of the next-nearest neighbor hopping. It is easy to see that the antibonding state $E^-_{\pi,\pi}$ always exists, while there is a critical coupling $t'_{\mathrm{cr}} = 1.2 \times 10^{-4} J$ for the bonding state, so that for $t' > t'_{\mathrm{cr}}$, it ceases to exist at (π, π). In real experiments t' and J are of the same order of magnitude, therefore, one can safely conclude that the bonding state disappears into the continuum. Below the superconducting transition temperature, the intensity of the antibonding state as measured in the neutron scattering experiment is proportional to

$$\left| \sum_p f_q^{(\alpha)}(p - q/2)\Delta_{p-q/2} \right|^2, \tag{11}$$

where $f_q^{(\alpha)}(p - q/2) \propto \sin(p_x) - \sin(p_y)$.

From Eqs. (9) and (11), we see immediately that the intensity is nonvanishing if and only if the gap symmetry is of the d-wave type. We therefore argue that the existence of the neutron resonance determines the *symmetry* of the pairing gap of the high-T_c superconductors to be of the d-wave type, consistent with the theories where pairing interaction arises from the spin fluctuations [13–15]. It could also be consistent with more exotic possibilities of $d_{x^2-y^2} + id_{xy}$ pairing symmetry [16].

We conclude that the basic features of the observed polarized neutron scattering experiment can be explained in terms of a new particle particle collective mode in the Hubbard model. The energy of the mode at (π, π) is given by Eq. (6) in the case of $t' = 0$, and is basically a fraction of J, which can be easily 41 meV. This formula also predicts that the mode energy should scale like $1 - n$, and should be lower for underdoped systems. The mode is centered around (π, π, π) for two different reasons, both because the particle particle continuum at this momentum is minimal so that the antibound state could exist and because of the conservation of angular momentum for exciting a singlet Cooper pair to a triplet state. This is exactly the momentum transfer of the excitation observed in experiment. We predict that similar modes should exist in other high-T_c materials as well, at the commensurate momentum (π, π, π). The intensity of the mode scales with the superfluid density because the BCS pairing amplitude is involved in converting a particle hole pair into a particle particle pair. The antibonding collective mode only has an overlap with the d-wave order parameter, and we conclude that the experimental observation of the collective resonance in the neutron scattering experiment can only be consistent with the d-wave symmetry of the pairing gap.

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