Neutron Scattering and Superconducting Order Parameter in YBa$_2$Cu$_3$O$_7$

I. I. Mazin

Geophysical Laboratory, Carnegie Institution, 5251 Broad Branch Road, NW, Washington, DC 20015

Victor M. Yakovenko

Department of Physics and Center for Superconductivity, University of Maryland, College Park, Maryland 20742-4111

We discuss the origin of the neutron scattering peak at 41 meV observed in YBa$_2$Cu$_3$O$_7$ below T_c. The peak may occur due to spin-flip electron excitations across the superconducting gap which are enhanced by the antiferromagnetic interaction between Cu spins. In this picture, the experiment is most naturally explained if the superconducting order parameter has s-wave symmetry and opposite signs in the bonding and antibonding electron bands formed within a Cu$_2$O$_4$ bilayer.

Neutron scattering is an important tool in the study of high-T_c superconductors. A sharp magnetic neutron scattering peak was observed in the superconducting state of YBa$_2$Cu$_3$O$_x$ at a 2D wave vector $\mathbf{Q} = (\pi/a, \pi/b)$ (a and b are the lattice spacings of the CuO$_2$ plane) and energy ω equal to 41 meV at $x \approx 7$ [1]. This effect was confirmed by several groups for $x = 7$ [2,3] and $6.5 < x < 7$ [4,5]. The peak has the following remarkable features: (a) It appears only below the superconducting transition temperature T_c [6]; (b) it is localized in both energy and wave vector; and (c) it has sinusoidal dependence on q_z, the wave vector perpendicular to the CuO$_2$ planes, which implies perfect antiferromagnetic correlation of the two planes in a Cu$_2$O$_4$ bilayer.

A number of theories, suggested to explain this effect, presume that the peak occurs due to spin-flip electron excitations across the superconducting gap [1,3,7–15]. It was emphasized in Ref. [3] that, since magnetic scattering is odd with respect to the time reversal, the BCS coherence factor in the neutron scattering amplitude vanishes unless Δ_k has opposite signs for the electron wave vectors $\mathbf{k}$ and $\mathbf{k} + \mathbf{q}$ connected by the 2D neutron wave vector transfer $\mathbf{q}$ (see the inset to Fig. 1):

$$\Delta_\mathbf{k} \Delta_\mathbf{k+q} < 0. \tag{1}$$

Through this condition, neutron scattering can probe the symmetry of the superconducting state. For $\mathbf{q} = \mathbf{Q}$, condition (1) is not satisfied for a simple s-wave state, but is satisfied for the $d_{x^2-y^2}$ state. So, it was suggested in Refs. [3,8–15] that the peak in question is a manifestation of the $d_{x^2-y^2}$ pairing in YBa$_2$Cu$_3$O$_7$.

However, since most of these theories, as well as Ref. [16], dealt with only one CuO$_2$ layer, they were unable to consider important feature (c). Because YBa$_2$Cu$_3$O$_7$ consists of Cu$_2$O$_4$ bilayers, there should be two electron bands formed by bonding and antibonding states. An important issue is the relative sign of the superconducting order parameter in these two bands. Along with a regular d-wave state which has the same sign in the two bands, another state, which we will call

$s^\pm$, was discussed in the framework of weakly [7,17,18] or strongly correlated electrons [19–21]. In this state, the order parameter has s-wave symmetry and opposite signs in the bonding and antibonding bands. We show below that this state (unlike the d-wave state) provides the best explanation of the neutron scattering experiments, particularly, of feature (c).

The neutron scattering cross section is proportional to the imaginary part of the electron spin susceptibility $\chi(\mathbf{q}, q_z, \omega)$. In a bilayer system, it is given by the following expression [5,20]:

$$\chi(\mathbf{q}, q_z, \omega) = \chi^{(+)} \cos^2(q_z d/2) + \chi^{(-)} \sin^2(q_z d/2), \tag{2}$$

where d is the distance between the CuO$_2$ planes in the
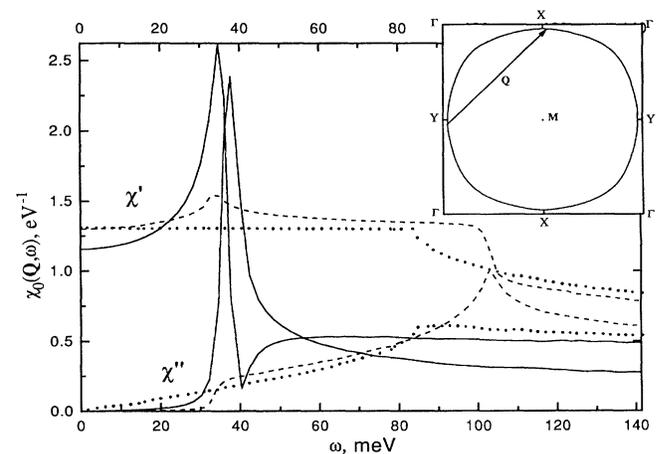
FIG. 1. The imaginary $\chi_0''^{(-)}(\mathbf{Q}, \omega)$ and real $\chi_0'^{(-)}(\mathbf{Q}, \omega)$ parts of the electron spin susceptibility in the superconducting $s^\pm$ (solid and dashed lines) and normal (dots) states at $T = 0$. Dashed lines and dots correspond to the Fermi energy $\mu = -370$ meV, solid lines to $\mu = -440$ meV. Inset: The Fermi surface for $\mu = -440$ meV.

bilayer, and

$$\chi^{(+)}(\mathbf{q}, \omega) = \chi^{(aa)}(\mathbf{q}, \omega) + \chi^{(bb)}(\mathbf{q}, \omega), \quad (3)$$

$$\chi^{(-)}(\mathbf{q}, \omega) = \chi^{(ab)}(\mathbf{q}, \omega) + \chi^{(ba)}(\mathbf{q}, \omega). \quad (4)$$

Here b and a refer to the bonding and antibonding electron bands of the bilayer. The susceptibilities $\chi^{(ij)}$ account for the transitions between the respective bands.

In experiments [1–5], only the second term in Eq. (2) was observed [feature (c)]. According to Eq. (3), the coefficient $\chi^{(+)}$, which appears in the first term of Eq. (2), involves transitions within the same band. Following the coherence factor arguments, we conclude that condition (1) is not satisfied within the same band, that is, the gap has s symmetry. On the other hand, the coefficient $\chi^{(-)}$ of the second term of Eq. (2), which involves transitions between the different bands (4), is not suppressed. Thus the order parameters of the different bands, $\Delta^{(a)}$ and $\Delta^{(b)}$, have opposite signs. This means that the state is the $s^\pm$ state described above. In this picture, the neutron scattering peak is due to the excitation of electrons, say, from the bonding band below the superconducting gap to the antibonding band above the superconducting gap, and the gaps have opposite signs.

In order to illustrate the above discussion, we performed explicit calculations for a well-known realistic tight binding model of $YBa_2Cu_3O_7$, which has the following electron dispersion law: $\xi_{\mathbf{k}} = -2t[\cos(k_x a) + \cos(k_y a)] - 4t' \cos(k_x a)\cos(k_y a) - \mu$, where $t = 250$ meV and $t'/t = -0.45$ [22]. We have chosen the Fermi energy $\mu = -440$ meV, so that the Van Hove singularity lies at an energy $\xi_{VH} = 10$ meV below the Fermi level [22,23]. The corresponding Fermi surface is shown in the inset to Fig. 1. To simplify the calculations, we set the hopping amplitude between the layers, $t_\perp$, equal to zero. This assumption does not qualitatively change our conclusions.

In BCS theory, at zero temperature, the susceptibilities are given by the following formula [24]:

$$\chi_0^{(i,j)}(\mathbf{q}, \omega) = \frac{1}{2}\sum_{\mathbf{k}}\left(1 - \frac{\xi_{\mathbf{k}+\mathbf{q}}\xi_{\mathbf{k}} + \Delta_{\mathbf{k}+\mathbf{q}}^{(i)}\Delta_{\mathbf{k}}^{(j)}}{E_{\mathbf{k}+\mathbf{q}}E_{\mathbf{k}}}\right)$$
$$\times \left(\frac{1}{\omega + E_{\mathbf{k}+\mathbf{q}} + E_{\mathbf{k}} + i\Gamma} - \frac{1}{\omega - E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} + i\Gamma}\right),$$

$$(5)$$

where $E_{\mathbf{k}} = \sqrt{\xi_{\mathbf{k}}^2 + \Delta_{\mathbf{k}}^2}$ is the quasiparticle dispersion law in the superconducting state, Γ is the damping constant, and the indices i and j label the bonding and antibonding electron bands. Because of the simplifying assumption $t_\perp = 0$, the dispersion laws of the bonding and antibonding bands are identical and do not need to be distinguished in Eq. (5). Only the signs of the order parameters $\Delta^{(i)}$ may depend on the index i. Note that only when (1) is satisfied does the coherence factor [the first line of Eq. (5)] not vanish at small energies $\xi \ll \Delta$.

We have calculated the spin susceptibility $\chi_0^{(-)}$ directly from Eqs. (4) and (5) using $\Gamma = 1$ meV and the $s^\pm$ gap computed in Ref. [17] for $T_c = 90$ K. The gap attains its maximal value $\Delta_0 = 17.5$ meV near the points X and Y in the Brillouin zone (see the inset to Fig. 1). Imaginary $\chi_0''^{(-)}(\mathbf{Q}, \omega)$ and real $\chi_0'^{(-)}(\mathbf{Q}, \omega)$ parts of the susceptibility are shown in Fig. 1 for the superconducting and normal states for two values of the Fermi energy μ. We observe that in the superconducting state, for the energies ω lower than the absorption threshold $E_{th} \approx 35$ meV ("spin gap"), $\chi_0''(\omega)$ is zero. Furthermore, for the realistic $\mu = -440$ meV both real and imaginary parts (solid curves) have sharp peaks at 35 and 38 meV, respectively. In order to clarify the origin of these features, we repeated the calculations (dashed curves) with an unrealistic choice of the Fermi energy $\mu = -370$ meV, which moves the Van Hove singularity much deeper below the Fermi level: $\xi_{VH} = 80$ meV. In this case, the peak in $\chi_0'(\omega)$ stays at the same energy E_{th}, and $\chi_0''(\omega)$ develops a step at the same energy. On the other hand, the peak in $\chi_0''(\omega)$ shifts to much higher energy of about 100 meV, which is close to $\Delta_0 + \xi_{VH}$. $\chi_0'(\omega)$ develops a negative step at the same energy. Similar results were obtained in Ref. [14] for the $d_{x^2-y^2}$ state.

To gain a qualitative understanding of this behavior, we set $\Gamma \to 0$ and set the coherence factor in Eq. (5) to 1. In this approximation, $\chi_0''(\mathbf{q}, \omega)$ is proportional to the joint density of states $A(\mathbf{q}, \omega) = \sum_{\mathbf{k}}\delta(\omega - E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}})$. The two-particle energy $E_2(\mathbf{k}, \mathbf{q}) = E_{\mathbf{k}+\mathbf{q}} + E_{\mathbf{k}}$, considered as a function of the 2D wave vector $\mathbf{k}$ for a fixed $\mathbf{q}$, has a minimum and several saddle points. The minimum defines the threshold energy E_{th}. In a 2D case, the joint density of states has a step at the threshold and logarithmic divergencies at the saddle point energies. Correspondingly, the real part $\chi_0'(\omega)$ which is related to $\chi_0''(\omega)$ by the Kramers-Kronig relations, has a logarithmic singularity at the threshold. Exactly that behavior is observed in Fig. 1. The minimum of $E_2(\mathbf{k}, \mathbf{Q})$ is achieved at a vector $\mathbf{k}$ such that both $\mathbf{k}$ and $\mathbf{k} + \mathbf{Q}$ belong to the Fermi surface (see the inset to Fig 1), where both $E_{\mathbf{k}}$ and $E_{\mathbf{k}+\mathbf{Q}}$ attain their minimal values approximately equal to Δ_0. Thus $E_{th} \approx 2\Delta_0 = 38$ meV.

The logarithmic peak in $\chi_0''(\omega)$ occurs because of transitions between the occupied states located near X and Y points and empty quasiparticle states above the superconducting gap. The points X and Y are the saddle points of the normal-state dispersion law $\xi_{\mathbf{k}}$. They produce the van Hove singularity in the single-particle density of states in the normal state at the energy ξ_{VH}. The logarithmic divergence in the joint density of states in the superconducting state is located, then, at the energy $E^* \approx \Delta_0 + \sqrt{\Delta_0^2 + \xi_{VH}^2}$. The value of ξ_{VH} depends on the Fermi energy μ. For the two choices of μ in Fig 1, the values of ξ_{VH} are equal, respectively, to 10 and 80 meV, which gives values of $E^* \approx 38$ and ≈ 100 meV, respectively, in agreement with the positions of the peaks

in $\chi_0''(\omega)$ in Fig 1. Note that both local density approximation calculations [22] and photoemission experiments [23] place the Van Hove singularity very close to the Fermi level, which corresponds to the first choice.

Comparison of $\chi_0''(\mathbf{Q}, \omega)$ in the normal state at zero temperature (dotted curve in Fig 1) and in the superconducting state (dashed line) shows that the logarithmic peak due to the Van Hove singularity is absent in the normal state. Instead, we see a cusp at an energy close to $\xi_{VH} = 80$ meV, which mark the threshold where transitions from the saddle points to the Fermi level become allowed. The reason that, instead of a cusp in the normal state, transforms a divergence develops in the superconducting state is that in the latter case, roughly speaking, the transitions take place between the Van Hove singularity and the coherence peak in the quasiparticle density of states. Since there is no threshold of absorption in the normal state, $\chi_0'(\omega)$ has no divergence at a finite ω in this state (dotted line in Fig. 1).

Summarizing, we conclude that the peaks in Fig. 1 appear to manifest two different physical mechanisms. The peak in $\chi_0'(\mathbf{Q}, \omega)$ is due to the transitions between the states just below the gap to the states right above it at the energy $E_{th} \approx 2\Delta_0$, which depends only on the superconducting gap. On the other hand, the peak in $\chi_0''(\mathbf{Q}, \omega)$ is due to the transitions from an occupied saddle point to the empty states just above the superconducting gap. The energy of this peak depends on the Van Hove energy of the normal state ξ_{VH}. For a realistic Fermi energy $\mu = 440$ meV, $\xi_{VH} = 10$ meV is small compared to $\Delta_0 = 17.5$ meV, therefore the peaks in χ_0' and χ_0'' approach each other, which is shown below to have a major effect on neutron scattering. None of these peaks appear in the normal state.

Curves in Fig. 1 are reminiscent of the experimental curves [1,4], where $\chi''(\mathbf{Q}, \omega)$ was found to exhibit a spin gap and a peak at different energies in underdoped samples with $x < 7$. It is tempting [14] to identify the logarithmic peak in $\chi_0''(\mathbf{Q}, \omega)$ with the peak observed in the experiment. However, the plot of $\chi_0''(\mathbf{q}, \omega)$ reveals a structure quite different from that found experimentally. While there is a local maximum at $\mathbf{q} = \mathbf{Q}$ and $\omega \approx 2\Delta_0$ in Fig 2, the corresponding peaks, although weaker, exist also at other values of $\mathbf{q}$, in contradiction with feature (b). Even worse, when $\mathbf{q} \rightarrow 0$, $\chi_0''(\mathbf{q}, \omega)$ increases to much higher values than at $\mathbf{q} = \mathbf{Q}$. We conclude that $\chi_0''(\mathbf{q}, \omega)$ does not provide a satisfactory explanation of the experimentally observed neutron scattering.

Thus far we have neglected the interaction between quasiparticles in the superconducting state. If we take into account a weak antiferromagnetic interaction between Cu spins in the plane, $J(\mathbf{q})$, and between the planes, $J_\perp$, the bare spin susceptibilities $\chi_0^{(\pm)}$, discussed above, should be renormalized in a random phase approximation manner [11–13,20,25]:

$$\chi^{(\pm)}(\mathbf{q}, \omega) = \chi_0^{(\pm)}(\mathbf{q}, \omega)/[1 + J^{(\pm)}(\mathbf{q})\chi_0^{(\pm)}(\mathbf{q}, \omega)/2], \quad (6)$$

$$J^{(\pm)}(\mathbf{q}) = J(\mathbf{q}) \pm J_\perp. \quad (7)$$
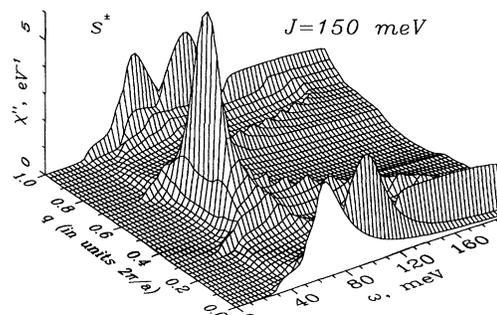
FIG. 2. Bare susceptibility $\chi_0''{}^{(-)}(q_x, q_y, \omega)$, plotted as a function of $q = q_x = q_y$ and ω for the $s^\pm$ order parameter with $\mu = -440$ meV.

Since the *real* part, $\chi_0'{}^{(-)}(\mathbf{q}, \omega)$, diverges as $\omega \rightarrow E_{th}$, as discussed above, the renormalized susceptibility $\chi^{(-)}(\mathbf{q}, \omega)$ (6) has a pole at ω close to E_{th} with singularities in both real and imaginary parts of $\chi^{(-)}$. Physically, the pole in $\chi^{(-)}(\mathbf{q}, \omega)$ describes a triplet electron-hole collective mode (an "antiparamagnon" or a "triplet exciton") with the energy slightly below E_{th}. In this picture, the peak in the neutron scattering rate occurs due to inelastic excitation of this collective mode by neutrons.

Since the singularity in $\chi_0'{}^{(-)}(\mathbf{q}, \omega)$ exists, generally speaking, for any $\mathbf{q}$, the exciton state (and, thus, a peak in neutron scattering) should exist for any $\mathbf{q}$, in contradiction with feature (b). However, if we take into account a finite lifetime of quasiparticles Γ in Eq. (5), the excitons also acquire a finite lifetime. For the antiferromagnetic interaction between the nearest neighbors, $\chi^{(-)}(\mathbf{q}, \omega)$ acquires a peak at $\mathbf{q} = \mathbf{Q}$, where $J(\mathbf{q}) = J_\parallel[\cos(q_x a) + \cos(q_y b)]$ reaches its maximal negative value. Thus the position of the neutron peak in $\mathbf{q}$ space is set by $J(\mathbf{q})$ and in ω by $\chi_0'{}^{(-)}(\mathbf{Q}, \omega)$. This statement is illustrated in Fig 3, where we show $\chi''(\mathbf{q}, \omega)$ calculated according to Eq. (6) with $J^{(-)} = 150$ meV and $\Gamma = 1$ meV for the $s^\pm$ state. In agreement with experiment, a single peak
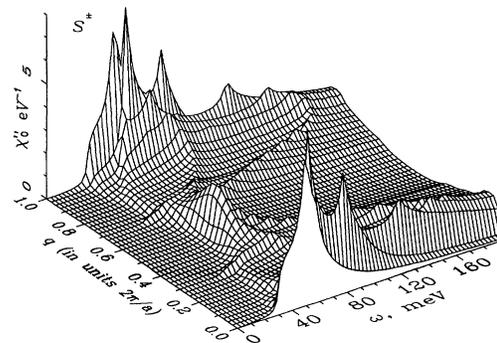
FIG. 3. Renormalized susceptibility $\chi''{}^{(-)}(q_x, q_y, \omega)$, plotted as a function of $q = q_x = q_y$ and ω for the same $s^\pm$ state as in Fig. 2.

in $\chi'''^{(-)}(\mathbf{q}, \omega)$, localized in both $\mathbf{q}$ and ω, is observed, which is now more than twice as high as that at $q \to 0$. The magnitude of the peak depends on the chosen value of J and Γ, but qualitatively the picture remains the same for a reasonable range of these parameters. Similar results were obtained in Ref. [13] for the $d_{x^2-y^2}$ state. It is worth noting that the peak in $\chi'''^{(-)}$ is due to the peak in the real part of $\chi_0^{(-)}$. When the Van Hove singularity is close to the Fermi level, the peak in $\chi_0'^{(-)}(\mathbf{Q}, \omega)$ gets stronger and enhances the peak in the renormalized susceptibility $\chi^{(-)}(\mathbf{q}, \omega)$.

The crucial difference between the $s^{\pm}$ and $d_{x^2-y^2}$ states is due to the following: In the $s^{\pm}$ state, the divergence occurs only in $\chi_0'^{(-)}$, but not in $\chi_0'^{(+)}$, where it is suppressed by the coherence factor (5). This suppression, unlike the case of logarithmic divergence in $\chi_0'''^{(+)}$, is exact, because the divergence in the real part of $\chi_0^{(+)}$ would occur precisely at the threshold energy where the coherence factor vanishes. As a consequence, among the renormalized susceptibilities, only $\chi^{(-)}$ diverges, but not $\chi^{(+)}$. Taking into account Eq. (2), this naturally explains feature (c) of the experiment.

In the case of the d wave, the coherence factor allows divergencies in both $\chi_0'^{(-)}$ and $\chi_0'^{(+)}$, which, in turn, produce divergencies in both $\chi^{(-)}$ and $\chi^{(+)}$. The only way to reconcile this with the experiment is to assume that $J^{(+)}$ is small: $|J^{(+)}| \ll |J^{(-)}|$, or even has a wrong (positive) sign. According to Eq. (7), this would require that the antiferromagnetic interaction between the layers be stronger than within a layer: $J_{\perp} \gtrsim 2J_{\parallel}$, which is unrealistic [26].

A number of theoretical papers [7–11] consider the special case when $t' = 0$ in the dispersion law. This assumption results in nesting at the Fermi energy or at another energy ("dynamic nesting"), which produces a peak in $\chi_0''(\mathbf{Q}, \omega)$. However, this case is not relevant for YBa$_2$Cu$_3$O$_7$, where $\mathbf{Q}$ is not a nesting vector.

In conclusion, we considered a scenario, where the peak at 41 meV observed in neutron scattering experiments [1–4] occurs due to spin-flip interband electron excitations across the superconducting gap, which are enhanced by the antiferromagnetic interaction between Cu spins. We found that the experiment can be explained most naturally if the superconducting order parameter is of the $s^{\pm}$ type, that is, the order parameter which has the s-wave symmetry and the opposite signs in the bonding and antibonding bands. This state easily explains the observed dependence of the scattering intensity on the momentum perpendicular to the CuO$_2$ planes. On the other hand, the $d_{x^2-y^2}$ case can be reconciled with the observed dependence only if the antiferromagnetic interaction between the CuO$_2$ planes is stronger than the interaction within the plane, which seems to be unrealistic.

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[26] After submission of the manuscript we became aware of two preprints considering neutron scattering in a bilayer with d pairing. B. Normand, H. Kohno, and H. Fukuyama (Report No. cond-mat/9505001), in agreement with our conclusion about d symmetry, were not able to produce the required q_z modulation. At the same time, D. Z. Liu, Y. Zha, and K. Levin [preceding Letter, Phys. Rev. Lett. 75, 4130 (1995)] argued that a small $J_{\perp} = 10$ meV can account for the observed dependence on q_z. Since Liu et al. did not report the value of $J_{\parallel}$ that they used, we were not able to trace the origin of the discrepancy in the conclusions.