Collective Excitations in High-Temperature Superconductors

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Collective, low-energy excitations in quasi-two-dimensional \textit{d}-wave superconductors are analyzed. While the long-range Coulomb interaction shifts the charge-density-wave and phase modes up to the plasma energy, the spin-density-wave excitation that arises due to a strong local electron-electron repulsion can propagate as a damped collective mode within the superconducting energy gap. It is suggested that these excitations are relevant to high-$T_c$ superconductors, close to the antiferromagnetic phase boundary, and may explain some of the exotic features of the experimentally observed spectral-density and neutron-scattering data.

An important aspect of superconductivity is concerned with collective modes as they may modify low-energy properties of superconductors \cite{1}. In principle, these excitations are either attributed to a symmetry transformation under which the system is invariant or to a spontaneously broken continuous symmetry. One might then expect that, in addition to the usual charge and spin fluctuations, collective modes associated with the phase and the amplitude of the superconducting order parameter are important. Yet, in conventional superconductors with a momentum-independent energy gap, collective modes have essentially no practical significance regarding the low-energy properties of the superconductors mainly because the long-range Coulomb interaction causes the phase mode to appear at the plasma energy \cite{2}. The only mode that is not diffusive at long wavelengths is the amplitude mode, but it has no direct coupling to charge or spin degrees of freedom making its observation demanding \cite{2}.

Moreover, excitonic states that are bound pairs of quasiparticles have been predicted to exist in superconductors \cite{3}. Such states appear in angular-momentum channels other than the one in which the Cooper pairing occurs. Theoretically, they should be present once the effective electron-electron interaction has an attractive partial-wave component with a given angular momentum. Nonetheless, there is no experimental evidence for these kind of states possibly because of their small binding energy \cite{3}.

While for most phenomena collective excitations in conventional superconductors can be ignored — for instance, the superconducting energy gap in the electronic spectrum at the Fermi energy is not affected by these modes — there are both theoretical and experimental reasons to expect that high-$T_c$ superconductors may behave differently in this respect. Theoretically, the energy gap has a strong momentum dependence and there is a large local electron-electron repulsion which may qualitatively change the nature of collective excitations and allow new ones to develop that are not related to the broken gauge symmetry. Experimentally, neutron-scattering studies \cite{4} in superconducting La$_{1.6-x}$Nd$_x$Sr$_2$CuO$_4$ have established the existence of elastic peaks in the magnetic structure factor at wave vectors $(\frac{1}{2}, \frac{1}{2}, \epsilon)$ and $(\frac{1}{2}, \frac{1}{2}, -\epsilon)$ (measured in units of $2\pi/a$), providing direct evidence for (diffusive) spin-density-wave fluctuations in high-$T_c$ superconducting materials. These excitations can be regarded as a manifestation of fluctuating charge stripes and antiphase spin domains which have shown to provide a natural explanation for the unusual features observed in angle-resolved photoemission experiments \cite{5}. We may also argue that they account for the broad “bosonic” feature in the electronic spectral density near the $(\frac{1}{2}, 0)$ and $(0, \frac{1}{2})$ points seen only in the underdoped materials \cite{6}.

In this Note, we examine under what conditions collective excitations develop in \textit{d}-wave superconductors and what are their experimental implications, when the quasi-particle picture is appropriate. Our most important observation is that \textit{d}-wave superconductors close to the antiferromagnetic phase boundary support a low-energy spin-density-wave mode. As a consequence of the superconducting energy gap, this mode can propagate coherently with reduced damping, unlike its precursor in the normal state where only a diffusive spin-density wave is realized. It can be excited by magnetic processes making it observable, for instance, by inelastic neutron scattering. The mode is “massive” because rotational symmetry in spin space is not broken. Under specific conditions, however, the mass of the mode may vanish and may even become negative signaling an instability of the superconducting ground state against a spontaneous creation of a spin-density-wave state.

Consider, for example, the Hamiltonian

\begin{equation}
H = -\sum_{\mathbf{r}\mathbf{r}'\sigma} t_{\mathbf{r}\mathbf{r}'} \psi_{\mathbf{r}\sigma}^\dagger \psi_{\mathbf{r}'\sigma} + \frac{1}{2} \sum_{\mathbf{r}\mathbf{r}'} v(\mathbf{r} - \mathbf{r}') n_\mathbf{r} n_{\mathbf{r}'} - \mu \sum_\mathbf{r} n_\mathbf{r},
\end{equation}

where $n_\mathbf{r} = \sum_\sigma \psi_{\mathbf{r}\sigma}^\dagger \psi_{\mathbf{r}\sigma}$ is the electron number operator at site $\mathbf{r}$, $t_{\mathbf{r}\mathbf{r}'}$ is the tunneling-matrix element between sites $\mathbf{r}$ and $\mathbf{r}'$, $\mu$ is the chemical potential, and $v(\mathbf{r})$ is
an instantaneous electron-electron interaction. In describing superconducting order, it is useful to express the Hamiltonian in the form \( H = H_{\text{BCS}} + H_{\text{int}} \), where the BCS and interaction Hamiltonians are

\[
H_{\text{BCS}} = \sum_k \Psi_k^\dagger \left( \epsilon_k \hat{\tau}_3 - \Delta_k \hat{\tau}_1 \right) \Psi_k
\]

\[
H_{\text{int}} = \frac{1}{2N} \sum_{kk'qq} v(q)(\Psi_{k+q}^\dagger \hat{\tau}_3 \Psi_k)\left(\Psi_{k'-q}^\dagger \hat{\tau}_3 \Psi_{k'}\right) + \sum_k \Delta_k \Psi_k^\dagger \hat{\tau}_1 \Psi_k.
\]

Here, \( \Psi_k = (\psi_k^\uparrow \psi_k^{\downarrow})^T \) is the Gor’kov-Nambu spinor, \( \epsilon_k = \sum_r \epsilon(r)e^{-qr} - \mu \) is the single-particle energy relative to the chemical potential, and \( v(q) = \sum_r v(r)e^{-qr} \).

The fermion operators in real and momentum space are related by the unitary transformation \( \psi_{k\sigma} = N^{-1/2} \sum_k \psi_{k\sigma} \epsilon_k^{1/2}e^{ikr}, \) where \( N \) is the number of sites in the system. Also, suppose that there exists a wave vector \( Q \) such that \( \epsilon_{k+Q} \approx -\epsilon_k \); i.e., the Fermi surface is approximately nested. This kind of situation may qualitatively arise in lightly doped high-\( T_c \) superconductors.

The energy gap \( \Delta_k \) is determined by requiring that the interaction Hamiltonian does not give any self-energy corrections to the energy gap \( \hat{G} \). This condition leads to the gap equation

\[
\Delta_k = -\frac{i}{2} \sum_p v(k - p) \text{Tr} \hat{\tau}_1 \hat{G}(p, \omega),
\]

where \( \hat{G}(p, \omega) = 1/(\omega \hat{\tau}_3 - \epsilon_p \hat{\tau}_3 + \Delta_p \hat{\tau}_1) \) denotes the Green’s function of the BCS Hamiltonian. We use the notation in which \( \sum_p = N^{-1} \sum_p \int (d\omega/2\pi) \) and \( p = (p, \omega) \).

In order to determine whether the system can support collective excitations, consider an effective two-particle interaction \( \hat{\Gamma} \) that describes mutual scattering of two quasiparticles. The poles of the effective interaction then yield the energy and lifetime of two-particle collective excitations. In the ladder approximation, the Bethe-Salpeter equation for \( \hat{\Gamma} \) may be written formally as

\[
\hat{\Gamma} = \hat{\Gamma}_0 + \sum_p \hat{\Gamma}_0(p, q)\hat{\Lambda}(p, q)\hat{\Gamma}(p, q),
\]

with \( \hat{\Lambda}(p, q) = \hat{G}(p + q/2) \otimes \hat{G}(p - q/2)^T \); here, \( q \) is the total four-momentum of a quasiparticle pair. The bare interaction vertex, denoted as

\[
\hat{\Gamma}_0(k, k'; q) = v(k - k')\hat{\tau}_3 \otimes \hat{\tau}_3 - v(q)\hat{P},
\]

\( \hat{P} = (\hat{\tau}_1) \). Moreover, the outer product, \( [A \otimes B]_{ab} = A_{a}B_{b} \), is defined so that there is a one-to-one correspondence between the ordered index lists \( a, b \) and \( (ij), (kl) \in \{(11,22), (12,12), (21,11)\} \).

On a square lattice, it is convenient to define orthogonal functions \( \eta_\alpha(k) \) in terms of which the vertex functions and the electron-electron interaction can be expanded \( [\hat{\Gamma}] \). For example, \( v(k - k') = U + V_1 [\cos(k_x - k'_x)a + \cos(k_y - k'_y)a] \) may be written as \( v(k - k') = \sum_{\alpha\beta} v_{\alpha\beta}(k)\eta_\alpha(k)\eta_\beta(k') \), where, for \( \alpha = 0 \), \( v_\alpha = U \) is the on-site electron-electron interaction and, for \( \alpha = 1, \ldots, 4 \), \( v_\alpha = V_1/2 \) is the nearest-neighbor electron-electron interaction. Below, we will assume that the on-site interaction is repulsive, \( U > 0 \), and the nearest-neighbor interaction is attractive, \( V_1 < 0 \), as should be appropriate for a phenomenological model describing high-\( T_c \) superconductors. Similarly, \( \hat{\Gamma}(k, k'; q) = \sum_{\alpha\beta} \hat{\Gamma}_{\alpha\beta}(q)\eta_\alpha(k)\eta_\beta(k') \) and \( \hat{\Gamma}_0(k, k'; p) = \sum_{\alpha\beta} \hat{\Gamma}_{\alpha\beta}(0)\eta_\alpha(k)\eta_\beta(k') \), with \( \hat{\Gamma}_{\alpha\beta}(q) = v_{\alpha\beta}\delta_{\alpha\beta}\delta_{13} \otimes \hat{\tau}_1 - v(q)\delta_{\alpha\beta}\delta_{13} \hat{P} \). Thus, the Bethe-Salpeter equation becomes

\[
\hat{\Gamma}_{\alpha\beta}(q) = \hat{\Gamma}_{\alpha\beta}^{(0)}(q) + \sum_{\gamma\gamma'} \hat{\Gamma}_{\alpha\beta}^{(0)}(q)\hat{\Lambda}_{\gamma\gamma'}(p)\hat{\Gamma}_{\gamma\gamma'}(q),
\]

where \( \hat{\Lambda}_{\alpha\beta} = i \sum_p \eta_\alpha(p)\hat{\Lambda}(p, q)\eta_\beta(p) \).

To compactify the notation, define new matrices such that \( [\hat{\Gamma}]_{\alpha\beta} = \hat{\Gamma}_{\alpha\beta}^{(0)} \), etc. Then, it is immediately clear that the effective interaction vertex is

\[
\hat{\Gamma}(q) = [1 - \hat{\Gamma}^{(0)}(q)\hat{\Lambda}]^{-1}\hat{\Gamma}^{(0)}(q),
\]

where \( q = (q, \Omega + i0^+) \). Because, for \( q = 0 \), \( \hat{\Lambda}(-p, q) = \hat{\Lambda}(p, q) \), there is no mixing between even and odd parity sectors of \( \hat{\Gamma}_{\alpha\beta}(q) \): the subspaces \( \{\eta_k^{(0)}, \eta_k^{(\pm)}\} \) and \( \{\zeta_k^{(\pm)}\} \) describing scattering in the singlet and triplet channels decouple. In contrast, for \( q = \Omega \), the subspaces \( \{\eta_k^{(0)}, \zeta_k^{(\pm)}\} \) and \( \{\eta_k^{(0)}, \eta_k^{(\pm)}\} \) are decoupled. When the system has particle-hole symmetry at the Fermi energy and \( \Omega \approx 0 \), a further factorization can be shown to occur; namely, particle-hole and particle-particle channels decouple. This means that, for \( q = \Omega \), the subspace \( \{\zeta_k^{(\pm)}\} \) decouples from \( \{\eta_k^{(0)}, \zeta_k^{(\pm)}\} \) and, for \( q = 0 \), the same occurs for \( \{\eta_k^{(0)}, \eta_k^{(\pm)}\} \). This is particularly convenient because \( \eta_k^{(\pm)} \) determines the \( d \)-wave gap function, \( \Delta_k = (\Delta_0/2)\eta_k^{(\pm)} \). At zero temperature, Eq. (6) becomes
1 = -(V_1/N) \sum_k |\tilde{\eta}^{(-)}_k|^2/4E_k. Here, \( E_k = \sqrt{\xi_k^2 + \Delta_k^2} \) is the quasiparticle energy in the superconductor. We find \( \Delta_0 \approx 4W e^{-1/N_F|V_1|} \), where \( W \) is the half bandwidth and \( N_F = N(\mu) \) is the density of states at the Fermi energy in the normal state.

The collective excitations are described by the poles of the effective two-particle interaction, Eq. (1), and they are conveniently classified by the symmetry properties of the system. The Hamiltonian (2) possesses a number of symmetries which lead to conserved currents. First, gauge symmetry, generated by the transformation \( \Psi_k \to \Psi_k' = e^{i\xi_kq} \Psi_k \), yields charge conservation. On general grounds, one then expects that there is a collective mode associated with the phase of the superconducting order parameter. However, due to the long-range nature of the Coulomb interaction, the Anderson-Higgs mechanism shifts it at small momenta to the plasma energy. Second, the symmetry transformation, \( \Psi_k \to \Psi_k' = e^{i\xi_kq} \Psi_k \), is associated with an amplitude mode of the order parameter \( \Psi(0) \). In contrast to \( s \)-wave superconductors, this mode is always damped in \( d \)-wave superconductors.

Third, spin-rotational symmetry leads to a new mode in the particle-hole sector, which is driven by the on-site Coulomb repulsion. As an example, consider a cylindrical Fermi surface and a wave vector \( q = Q \) nesting two given \( k \) points with vanishing quasiparticle energies. For \( U \sim |V_1| \), the energy \( \Omega_Q \) of the collective excitation is

\[
\beta \left( \frac{\Omega_Q}{2\Delta_0} \right) \approx \frac{1}{4} \left( \frac{V_0}{U} - 1 \right) - \kappa_Q,
\]

when \( \beta > 0 \). For \( \Delta_0, |\mu| \ll W, \) \( V_0^{-1} \approx \frac{1}{N} \sum_k \frac{\xi_k}{2E_k} \sim |V_1|^{-1} \). The parameter \( \beta \) measures the magnitude of the particle-hole symmetry breaking in the density of states \( N(\epsilon) \) at the Fermi energy and \( \kappa_Q \) describes the contribution due to the mixing between the particle-hole and particle-particle channels. For \( U, |V_1| \ll W \), it can be expanded as \( \kappa_Q = \kappa_Q^{(1)} + \kappa_Q^{(2)} \), where \( \kappa_Q^{(1)} \approx 2 \left( \frac{\gamma_5}{16} \right)^2 + \frac{\gamma_5}{2\gamma_3} \) and \( \kappa_Q^{(2)} \approx \frac{1}{4U} \left( \frac{\gamma_5}{16} \right)^2 \). For \( \beta < 0 \), the collective excitation remains massive \( \left( \Omega_Q > 0 \right) \) for all values of \( V_0/U \), down to the point where the superconductor becomes unstable. The excitation is damped, because it overlaps with the quasiparticle continuum. For the other values of \( q \), the continuum does not necessarily start from zero but at some finite value \( \bar{\delta}_q \equiv \min_k (E_k + E_{k+q}) \). It is then possible to have collective excitations with an infinite lifetime, when \( \Omega_q < \bar{\delta}_q \).

The exact nature of the collective excitation is deduced by examining its coupling to the spin operator \( S_z(q) = \sum_k \tilde{\psi}_{1/2}^{(0)} \tilde{\psi}_{1/2+q} \) where \( \tilde{\psi}_{1/2}^{(0)} = \frac{1}{2} \tilde{\psi}_0 \). That the mode is a spin-density wave becomes evident by computing the spin correlation function, \( \chi(q, \tau) = \langle T_x S_z(q, \tau) S_z(-q, 0) \rangle \).

![Image]( attachment://figure1.png)

**FIG. 1.** The energy \( \Omega_q \) of the spin-density-wave excitation as a function of wave vector \( q \) at zero temperature for \( U/W = 0.485 \) and \( V_1/W = -0.608 \). Here, the superconducting energy gap is \( \Delta_0/W = 0.1 \) and the chemical potential is chosen so that the density of holes equals 10% (relative to half filling). The excitation energy \( \Omega_q \) is computed numerically from the exact result, Eq. (8).

Specifically, its temporal Fourier transform is given by the formula

\[
\chi(q) = \langle \gamma_z |\hat{\Lambda}(q)\{1 + \hat{\Gamma}(q)\hat{\Lambda}(q)\}|\gamma_z \rangle, \tag{8}
\]

where the column vector \( |\gamma_z \rangle \) is defined as \( \langle \alpha\alpha |\gamma_z \rangle = \frac{1}{2} \delta_{\alpha^0} |\alpha \rangle \). In the limit \( \Omega \to 0 \), our result reduces to the form obtained by non-conserving approximation \([12]\), when the energy spectrum has particle-hole symmetry at the Fermi energy. However, for energies \( \Omega \gtrsim \Delta_0 \) or in the absence of particle-hole symmetry, one must use the general result, Eq. (8). Note that, by incorporating the mixing of the particle-hole and particle-particle degrees of freedom, \( \kappa_Q \) accounts for, for example, the effect of any two-particle, spin-triplet excitations \([13]\) at the momentum \( Q \). Similar calculation shows that no resonance develops for the charge response near \( q = Q \).

The most favorable conditions for observing these excitations are most likely found in underdoped high-\( T_c \) superconductors close to the antiferromagnetic phase boundary. In the antiferromagnetic phase, the above collective mode is replaced by the Goldstone mode of the antiferromagnet. Interestingly, for \( U \gtrsim V_0 \), the system is unstable against a spontaneous creation of quasiparticle-quasihole virtual bound states at \( q = Q \). This implies a phase transition to an antiferromagnetic state. In contrast to \( d \)-wave superconductors, the Ward identity \([14]\) excludes the spin-density-wave collective mode in conventional superconductors with a momentum independent gap function.

To obtain a quantitative understanding of the dispersion relation of the collective mode, we resort to numerical methods. Figure 1 shows the energy of the spin-density wave as a function of wave vector near half filling with hole density equal to 10%. The minimum value of the excitation energy, \( \Omega_q/2\Delta_0 \approx 0.08 \), is located at the wave vector \( q \approx (0.95\pi/a, \pi/a) \), implying that the
superconductor becomes unstable against a spontaneous creation of an antiferromagnetically ordered state with vertical (horizontal) antiphase domain walls. In contrast to neutron scattering data, in superconducting La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ which show that the mean separation $\ell$ between antiphase domain walls should scale with the hole density $x$ as $\ell \approx a/2x$, we find that $\ell$ is more than by a factor of two longer than the experimental one. (In the present approximation, $\ell$ also depends on $\Delta_0$.) The failure to predict correctly the domain-wall periodicity is similar to the problem of describing the static, incommensurate stripe order in the Hartree-Fock approximation. Although at zero temperature exciting vertical spin-density-wave fluctuations requires the least amount of energy, the rotation of their orientation relative to the underlying lattice constitutes a relatively soft mode; see Fig. 1. For example, the excitation energy at the saddle point $q = 0.98Q_0$ is about 60% larger than the minimum energy required to excite a vertical mode. The form of the dispersion relation $\Omega_q$ is affected by the lifetime effects: the troughs clearly visible in Fig. 1 mark the boundaries between damped ($\Omega_q > \vartheta_q$) and undamped ($\Omega_q < \vartheta_q$) excitations. For example, the excitations with $q = Q_0$ and $0.94Q_0$ are undamped whereas the excitation with $q = 0.97Q_0$ is damped.

Figure 2 illustrates the behavior of the imaginary part of the spin correlation function, Eq. (3). The collective excitation produces a distinctive resonance structure in $\chi''(q, \Omega)$ at energies specified by the dispersion relation, $\Omega = \Omega_q$. At low energies, the resonance becomes narrower as the excitation energy $\Omega_q$ decreases. However, in the vicinity of the $(\pi/a, \pi/a)$ point, for example, the collective excitation appears below the onset energy of the two-particle continuum $\vartheta_q$. Accordingly, the collective excitation would acquire an infinite lifetime yielding a resolution-limited peak in $\chi''(q, \Omega)$ — also, below this onset energy, the usual quasiparticle contribution to $\chi''(q, \Omega)$ would vanish — if the lifetime of quasiparticles were infinite. Clearly the collective mode is an important new feature describing the low-energy spin correlation function.

In a $d$-wave superconductor, strongly-scattering impurities induce virtual bound states by modifying the local potential energy of electrons at the impurity site. In addition, they may also change local Coulomb interaction between electrons at the impurity site. Such an effect can be accounted for by including the term $U_{\text{imp}}n_{\text{r}_0\uparrow}n_{\text{r}_0\downarrow}$, where $\text{r}_0$ is the location of the impurity. A straightforward calculation shows that this effect leads to a virtual bound state with the energy $\Omega_0/2\Delta_0 \sim 2\sqrt{U_c/U_{\text{imp}}} - 1$, where $U_c \sim 1/\pi N_F$. In contrast to an impurity potential, the impurity interaction has a critical value $U_c$, below which the ground state is nonmagnetic and the spin quantum number of the resonance state equals to $\frac{1}{2}$, and above which the impurity becomes magnetic in the sense that one electron spin is trapped to the impurity site.

Finally, one may ask whether excitonic states of bound pairs of quasiparticles are feasible in $d$-wave superconductors. It is immediately clear that while the effective interaction has an attractive partial wave in the (extended) $s$-wave channel, it is nevertheless too weak near half filling to support any excitons with the same angular quantum number. It is only far away from half filling that these states might appear as virtual bound states because of the proximity to a superconducting state with extended $s$-wave symmetry.

In conclusion, we have shown that $d$-wave superconductors can support propagating collective modes that are best described as spin-density waves. It is then natural to anticipate that the most favorable conditions for detecting them are found close to the antiferromagnetic phase boundary. Furthermore, superconducting fluctuations couple particle-hole and particle-particle excitations allowing the latter ones to be probed by the usual means. This coupling is particularly important if the two excitations are (nearly) degenerate.

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Historically, collective excitations had an important role in establishing the gauge-invariant theory of superconductivity; for example, see Ref. [1] below.

[1] Historically, collective excitations had an important role in establishing the gauge-invariant theory of superconductivity; for example, see Ref. [9] below.

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