Mechanism for high-temperature superconductivity in cuprates

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A treatment of the Coulomb interaction in cuprates by a time-dependent multiple-scattering theory leads to coherently excited holes in the "antibonding" band with the appearance of a pairing interaction between the (coherently excited) holes. This pairing interaction leads to superconductivity in doped cuprates. The essential points of the mechanism are discussed by making some illustrative calculations and by examining the behavior of a few parameters.

In high-temperature superconductors, Coulomb interaction competes with the banding interaction.[1] A large number of theories have been developed for these systems,[2] but none of them is satisfactory. In this paper we develop a theory of high-temperature superconductors by keeping in mind the comparable strengths of the interelectron Coulomb interaction and the banding interaction. The method that we use to deal with such a situation is based on the idea that scattering theory can be used to investigate the evolution of the system from band states with use of the matrix element of the interelectron Coulomb interaction between the band states, which determines the transition probabilities between these states. We consider the screened Coulomb interaction in a simplified form, where the screening effects of the medium are represented by a dielectric constant ϵ. Such a screened interaction is justified for cuprates as these systems are ionic in nature[1] and have a small carrier concentration.[1] We write

$$U_{\rm Coul}(\mathbf{r}_i-\mathbf{r}_j)=e^2/\epsilon|\mathbf{r}_i-\mathbf{r}_j| \ . \tag{1}$$

Here $\mathbf{r}_i$ and $\mathbf{r}_j$ are positions of the interacting electrons, and e is the charge of a hole. (In doped cuprates the carriers are of hole nature.[1])

For simplicity we do not consider interlayer interactions and so restrict ourselves to the two-dimensional CuO_2 planes only. The Hamiltonian of the system (doped cuprates) can be written as

$$H=H_{\rm band}+H_{\rm Coul} \ . \tag{2}$$

Here $H_{\rm band}$ describes the holes in the band states. Written explicitly

$$H_{\rm band}=\sum_{b,\mathbf{k},\sigma}\varepsilon_{b\mathbf{k}}a^{\dagger}_{b\mathbf{k}\sigma}a_{b\mathbf{k}\sigma} \ , \tag{3}$$

where $a^{\dagger}_{b\mathbf{k}\sigma}$ and $a_{b\mathbf{k}\sigma}$ denote creation and annihilation operators, respectively, of a hole in the band b with momentum $\mathbf{k}$, spin σ, and energy $\varepsilon_{b\mathbf{k}}$. The $b=1$ band corresponds to the partially filled antibonding band,[3] while the $b>1$ bands lie below the $b=1$ band in terms of electron energy. In this way, the b bands are quasi-two-dimensional. In the process of interband transitions a hole will make transition from the $b=1$ to the $b>1$ bands.

The interaction $H_{\rm Coul}$ in Eq. (2) describes the (screened) Coulomb interaction between holes. Its explicit form is

$$H_{\rm Coul}=\sum_{i>j}\sum_{\text{all } b,\mathbf{k},\sigma}(b''\mathbf{k}''\sigma;b'''\mathbf{k}'''\sigma'|U_{\rm Coul}(\mathbf{r}_i-\mathbf{r}_j)|b\mathbf{k}\sigma;b'\mathbf{k}'\sigma')a^{\dagger}_{b''\mathbf{k}''\sigma}a^{\dagger}_{b'''\mathbf{k}'''\sigma'}a_{b'\mathbf{k}'\sigma'}a_{b\mathbf{k}\sigma} \ . \tag{4}$$

The interaction $H_{\rm Coul}$ forces a band hole of the system to undergo multiple scattering with other holes of the system.[4] Let $t_0,t_1,t_2,\ldots$ form a sequence of time ($t_n>t_{n-1}$ for all n) such that the nth scattering starts at t_{n-1} and is completed[4] by t_n. Let $f_{n,b\mathbf{k}}$ be the distribution function after the nth scattering, and $P_n(b\mathbf{k}\rightarrow b''\mathbf{k}'')$ be the transition probability per unit time for the $b\mathbf{k}\rightarrow b''k''$ transition. Then we may write

$$f_{n,b\mathbf{k}}=f_{n-1,b\mathbf{k}}\left[1-\sum_{b'',\mathbf{k}''}P_n(b\mathbf{k}\rightarrow b''\mathbf{k}'')\tau_n\right]+\sum_{b'',k''}f_{n-1,b''\mathbf{k}''}P_n(b''\mathbf{k}''\rightarrow b\mathbf{k})\tau_n \ , \tag{5}$$

with $f_{0,b\mathbf{k}}$ as the usual Fermi factor for holes, and

$$P_n(b\mathbf{k}\rightarrow b''\mathbf{k}'')=\frac{2\pi}{\hbar}\rho_{b''\mathbf{k}''}\Bigg|\sum_{b',b''',\mathbf{k}',\mathbf{k}'''}A_{b\mathbf{k},b''\mathbf{k}''}A_{b'\mathbf{k}',b'''\mathbf{k}'''}(b''\mathbf{k}''\sigma;b'''\mathbf{k}'''\sigma'|T(t_n)|b\mathbf{k}\sigma;b'\mathbf{k}'\sigma')$$
$$\times f_{n-1,b\mathbf{k}}f_{n-1,b'\mathbf{k}'}(1-f_{n-1,b''\mathbf{k}''})(1-f_{n-1,b'''\mathbf{k}'''})\Bigg| \ . \tag{6}$$

In Eq. (5) $\tau_n = t_n - t_{n-1}$. We have written Eq. (5) by using the fact that $P_n(b\mathbf{k} \rightarrow b''\mathbf{k}'')\tau_n \ll 1$. In Eq. (6) $\hbar$ is Planck's constant divided by 2π. $\rho_{b''\mathbf{k}''}$ is the density of hole states (per unit energy) at the energy $\varepsilon_{b''\mathbf{k}''}$, and $T(t_n)$ is the transition matrix corresponding to the interaction H_{Coul} during the nth scattering.

The quantities $A_{b\mathbf{k},b''\mathbf{k}''}$ in Eq. (6) are introduced to account for the nonconservation[5] of energy during the collision of two holes. When two holes, which are initially in the states $|b\mathbf{k}\sigma\rangle$ and $|b'\mathbf{k}'\sigma'\rangle$, collide with each other, then, due to the presence of other holes, they will be scattered to the non-energy-conserving (NEC) states

$$|b''\mathbf{k}''\sigma\rangle_{\text{NEC}} = \sum_{g,\mathbf{p}} A_{b''\mathbf{k}'',g\mathbf{p}} |g\mathbf{p}\sigma\rangle \tag{7}$$

and

$$|b'''\mathbf{k}'''\sigma'\rangle_{\text{NEC}} .$$

In order to calculate the coefficients $A_{b''\mathbf{k}'',g\mathbf{p}}$ we consider a very simple description of the non-energy-conserving processes. In fact, due to low hole concentration we may assume that non-energy-conservation arises when a third hole forces the scattered hole to change its state from $b''\mathbf{k}''$ to $g\mathbf{p}$. When it is so, the share of the $g\mathbf{p}$ states, may be related with the transition probability (per unit time) for the $b''\mathbf{k}'' \rightarrow g\mathbf{p}$ transition. In this sense we may write

$$A_{b''\mathbf{k}'',g\mathbf{p}} = A_0[P_n(b''\mathbf{k}'' \rightarrow g\mathbf{p})]^{1/2} . \tag{8}$$

Here A_0 is to be determined by requiring that

$$\sum_{g\mathbf{p}} |A_{b''\mathbf{k}'',g\mathbf{p}}|^2 = 1 . \tag{9}$$

We now present calculations for the distribution function at zero temperature. For this calculation we assume that there are only two (two-dimensional) bands, $b=1$, and $b=2$. Both the bands are assumed parabolic and truncated so that the $b=1$ band has a width of 4 eV, while the $b=2$ band has a width of 2 eV. The $b=1$ band is of positive effective mass, while the $b=2$ is of negative effective mass so that there is a 0.5-eV gap between the bottom of the $b=1$ band and the top of the $b=2$ band. The Fermi energy E_F is taken in the $b=1$ band at 3 eV up from the bottom of the band. In Eq. (1) ϵ is taken to

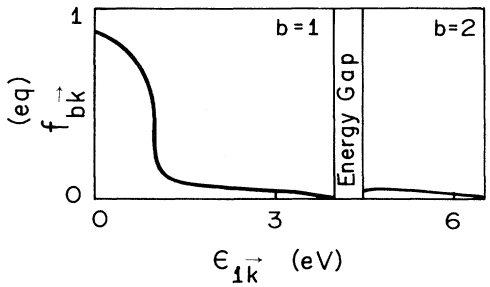

FIG. 1. Values of the equilibrium distribution function $f^{(eq)}_{b\mathbf{k}}$ for various hole energies.

be 6, which is a typical value for cuprates.[6] From a practical viewpoint, in Eq. (5) we have taken the time interval as $\tau = 0.01$ sec instead of τ_n. Using these simplifications, and applying the wave packet approach[7] for the calculation of T-matrix elements in the first-order approximation, we found that after the 237th step the distribution function attains an equilibrium value $f^{(eg)}_{b\mathbf{k}}$ that is shown in Fig. 1. It is clear that the distribution function has a discontinuity of 0.23 at E_F.[8] In a realistic calculation we would expect to get a different magnitude of discontinuity at E_F, but discontinuity will be there because the number 0.23 is not so small that one may doubt the discontinuity of the distribution function. Thus we can conclude that in (doped) cuprates the distribution function will be discontinuous at the Fermi energy. In this sense superconductivity in (doped) cuprates will be based on Cooper pairs.

We emphasize that the plane-wave functions used in the above calculations are considered for simplicity only. For a realistic calculation one should use realistic wave functions as obtained from a band-structure calculation.[1] Within the plane-wave approximation the values of the intrasite and intersite Coulomb interactions are 6.3 and 1.2 eV, respectively.

As a result of the above multiple-scattering process, a hole will move, at all times in a coherent mixture[9] of the excited $|1\mathbf{k}\sigma\rangle$ states. We can describe such a coherent state by the creation operator

$$\gamma^{\dagger}_{1\mathbf{k}\sigma}(t_n) = \sum_{\mathbf{k}',\mathbf{k}'',\mathbf{k}''',\sigma'} (1\mathbf{k}''\sigma; 1\mathbf{k}'''\sigma' | T(t_n) | 1\mathbf{k}\sigma; 1\mathbf{k}'\sigma') A_{1\mathbf{k},1\mathbf{k}''} A_{1\mathbf{k}',1\mathbf{k}'''} f_{n1\mathbf{k}} f_{n1\mathbf{k}'} (1-f_{n1\mathbf{k}''})(1-f_{n1\mathbf{k}'''}) a^{\dagger}_{1\mathbf{k}''\sigma} \tag{10}$$

along with a corresponding annihilation operator $\gamma_{1\mathbf{k}\sigma}(t_n)$.

The wave vector $\mathbf{k}''$ of the component states of the coherent mixture takes all such values (including $\mathbf{k}$ itself) for which the band states $|1\mathbf{k}''\sigma\rangle$ are in coherence. As the Coulomb interaction, which leads to coherence, is repulsive, we assume that the energies for $\mathbf{k}'' \neq \mathbf{k}$ are higher than those for $\mathbf{k}'' = \mathbf{k}$, where $\mathbf{k}$ is restricted to $k \leq k_F$ with k_F as the Fermi wave number. The energy range for the coherent state resulting from the $|1\mathbf{k}\sigma\rangle$ state will be $\Delta\varepsilon_{1\mathbf{k}} \sim \hbar/\tau_n$, where $\tau_n \sim r_s/v_g$ with r_s as the average intercarrier spacing, and v_g is the group velocity of wave packet in the band states. For cuprates $r_s \geq 3$ Å,[10] and v_g will be less than or equal to the Fermi velocity, which is of the order of 10^7 cm/sec (Ref. 11) in cuprates. This leads to $\Delta\varepsilon_{1\mathbf{k}} \leq 0.2$eV. This is at least one order of magnitude less than the typical band width[1] of cuprates.

It may further be noted that the strength of the coherence within the energy range $\Delta\varepsilon_{1\mathbf{k}}$ will depend on the strength of the interaction $U_{\text{Coul}}(\mathbf{r}_i - \mathbf{r}_j)$. In the present case, in the above model calculations, we have found that the transition probabilities $P_n(1\mathbf{k} \rightarrow 1\mathbf{k}'')$ are comparable for many $\mathbf{k}''$ values. This means that the usual Fermi-

liquid theory, which is based on single $\mathbf{k}''$ ($\mathbf{k}''=\mathbf{k}$) quasiparticle states is not applicable to (doped) cuprates. Rather, a consideration of the coherent nature of the hole states is necessary for a reasonable description of cuprates. We emphasize that such a deviation of the present "hole liquid" from the Fermi liquid is due to the nonenergy conservation approximation.

The holes in the coherent mixture of $|1\mathbf{k}''\sigma\rangle$ states, will polarize, through the electrons generated in the $b=1$ band, the medium of the holes of the $b>1$ bands which, in turn, will mediate an indirect interaction between the $b=1$ band holes. Such an indirect interaction has already been used in different excitonic models[10,12] of cuprate superconductivity. However, in the present case the situation is quite complicated as the indirect interaction acts between coherently excited holes. Keeping in mind that superconductivity in cuprate superconductors involves Cooper pairs of zero center-of-mass momenta,[13] we assume that $\xi^{(h\text{-}h)}(2\mathbf{k})$ is, to within a spin factor, the indirect interaction due to polarization of band electrons between holes characterized by the coherent mixture operators $\gamma^{\dagger}_{1\mathbf{k}\sigma},\gamma_{1\mathbf{k}\sigma}$ and $\gamma_{1,-\mathbf{k},-\sigma},\gamma_{1,-\mathbf{k},-\sigma}$. We can clarify the role of $\xi^{(h\text{-}h)}(2\mathbf{k})$ in terms of the indirect interactions $J^{(h\text{-}h)}(2k'')$ which, to within a spin factor, act between holes moving in states $|1\mathbf{k}'',\sigma\rangle$ and $|1,-\mathbf{k}'',-\sigma\rangle$ in the following manner:

$$\xi^{(h\text{-}h)}(2\mathbf{k})\gamma^{\dagger}_{1\mathbf{k}\sigma}(t_n)\gamma^{\dagger}_{1-\mathbf{k}-\sigma}(t_n) = \sum_{\mathbf{k}''} D_{\mathbf{k}\mathbf{k}''}(t_n)J^{(h\text{-}h)}(2\mathbf{k}'')a^{\dagger}_{1\mathbf{k}''\sigma}a^{\dagger}_{1,-\mathbf{k}'',-\sigma} \ . \quad (11)$$

Here the expression for $D_{\mathbf{k},\mathbf{k}''}(t_n)$ follows from the Eqs. (10) and (11).

To within a spin factor, we can write an expression for the interaction $J^{(h\text{-}h)}(2\mathbf{k}'')$ in a manner similar to that of Eq. (16) of Ref. 14.

In order to get an idea regarding the binding energy of Cooper pairs we proceed as follows. First of all we notice that the band states $\mathbf{k}''\neq\mathbf{k}$, as assumed above, correspond to higher energies than the $\mathbf{k}''=\mathbf{k}$ states. As the higher-energy states $\mathbf{k}''\neq\mathbf{k}$ are brought in because of the Coulomb repulsion, opposite spin holes of momenta $\mathbf{k}'\neq\mathbf{k},-\mathbf{k}''$ will form a bound state only if the pairing interaction $J^{(h\text{-}h)}(2\mathbf{k}'')$ can overcome the Coulomb repulsion associated with the $\mathbf{k}'',-\mathbf{k}''$ states. It is clear that the binding energy of the $\mathbf{k}''\neq\mathbf{k},-\mathbf{k}''$ Cooper pairs, say $\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$ will be reduced from its bare value

$$\Delta_0(\mathbf{k},\mathbf{k}'')=-\langle J_0 a^{\dagger}_{1\mathbf{k}''\sigma}a^{\dagger}_{1,-\mathbf{k}'',-\sigma}\rangle \ . \quad (12)$$

Here $\langle\cdots\rangle$ denotes an average in terms of the state of the system, and J_0 is the $l=0$ partial-wave component of the interaction $J^{(h\text{-}h)}(2\mathbf{k}'')$. (We have assumed s wave pairing.)

We calculate $\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$ through the contribution of the $\mathbf{k}''$ state to the ground-state energy of the system of superconducting quasiparticles.[15] An expression for such a contribution can be written as[16]

$$E(\mathbf{k}'',\Delta_0(\mathbf{k},\mathbf{k}''))=\varepsilon_{1\mathbf{k}''}-\frac{\{2(\varepsilon_{1\mathbf{k}''})^2+[\Delta_0(\mathbf{k},\mathbf{k}'')]^2\}}{2\{(\varepsilon_{1\mathbf{k}''})^2+[\Delta_0(\mathbf{k},\mathbf{k}'')]^2\}^{1/2}} \ . \quad (13)$$

In this expression, the effect of the Coulomb repulsion on the ground-state energy is incorporated into the energies, $\varepsilon_{1\mathbf{k}''}$ which appear in the coherent state formed out of the initial state $\mathbf{k}(k<k_F)$ due to the presence of the Coulomb interaction. An alternate form of this very contribution to the ground-state energy from the pair $\mathbf{k}''\neq\mathbf{k},-\mathbf{k}''$ can also be written as $E(\mathbf{k},\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}''))$ where now the effect of the Coulomb repulsion appears through the effective binding energy $\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$. Since both $E(\mathbf{k}'',\Delta_0(\mathbf{k},\mathbf{k}''))$ and $E(\mathbf{k},\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}''))$ represent the same quantity, we have

$$E(\mathbf{k},\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}''))=E(\mathbf{k}'',\Delta_0(\mathbf{k},\mathbf{k}'')) \ . \quad (14)$$

This equation provides us a way of determining $\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$.

From Eq. (14) we can see that a bound hole pair will be formed only for those $\varepsilon_{1\mathbf{k}''}$ that lie within a few meV above $\varepsilon_{1\mathbf{k}}$. In fact, for cuprate superconductors $\Delta_0(\mathbf{k},\mathbf{k}'')\sim 50$ meV[2] so that we estimate the "pairing" energy range above $\varepsilon_{1\mathbf{k}}$ for bound pairs to be of order 10 meV, which is more than $\frac{1}{20}$ of the complete coherence range (of ≤ 0.2 eV). We note that the above treatment of the Coulomb interaction is different from that of Morel and Anderson.[17] In the present case, superconductivity will occur only when the probability of finding the pairing holes in the pairing energy range (of order 10 meV) is such that on average the number of pairs in this energy range is macroscopic. An estimate of this probability of finding a pair, in the model system used in our calculation at $T=0$ K, leads to a value of about 0.67, which is sufficient for the occurrence of the superconducting order.

It is clear from Eq. (14) that the pairs will assume different binding energies $\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$ in accordance with the probabilities $D_{\mathbf{k}\mathbf{k}''}(t_n)$. When it is so, then the overall value of a property of the system, say $F(T)$, may be written as

$$F(T)=\int_0^{\Delta_0(\mathbf{k},\mathbf{k})} F_0(\Delta,T)\exp(-\Delta/k_BT)d\Delta \ . \quad (15)$$

Here the integrand is the product of the value of $F(T)$ for a given Δ and the corresponding weight factor $D_{\mathbf{k}\mathbf{k}''}(t_n)$. $F_0(\Delta,T)$ is a nonexponential function of temperature T.

Evaluation of the integral of Eq. (15) will lead to a dominating power law term. In fact, there will be an exponential term also but it will be too small to have any practical significance.

In the above discussion we have considered only the contribution of bound pairs to the temperature dependence of a physical property. But as has been clarified above, there will be a finite probability for finding holes in unpaired states also, even though the interaction $J^{(h\text{-}h)}(2\mathbf{k}'')$ between these holes will be there. According to the above estimate holes will remain in the unpaired states with probability ≈ 0.33. This means that the fraction of holes of the complete system existing in the unpaired state is macroscopic. So the unpaired holes will also contribute to the properties of the system. Such a contribution is important in at least two ways. Firstly, these holes will contribute a power-law temperature dependence for some of the physical properties of the sys-

tem. This fact, along with the above-noted power-law contribution of bound pairs, may be used to understand, for example, the T dependence of the low-temperature specific heat,[18] and of NMR relaxation rate.[19] Secondly, due to the presence of holes, properties like Knight shift will not vanish at $T=0$ K. This is indeed what has been observed experimentally.[20]

Another specific feature of the above mechanism of superconductivity is related to the superconducting energy gap. In the above mechanism, the superconducting energy gap will be obtained by averaging the individual gaps $2\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$ [in accordance with the probabilities $D_{\mathbf{k}\mathbf{k}''}(t_n)$]. Because of this, those experiments that measure the superconducting energy gap (Andreev reflection, tunneling, infrared absorption) may show effects of the individual gaps $2\Delta_{\text{eff}}(\mathbf{k},\mathbf{k}'')$ also in the sense that the gap will not be sharp. For example, in infrared absorption experiments there is a possibility that infrared frequencies below the average superconducting energy gap are absorbed. Such a residual absorption has indeed been observed even for single crystals.[21]

In conclusion, we have presented a mechanism for high-temperature superconductivity by treating the Coulomb interaction in a realistic manner. Pairing between coherently excited states is the key point of the present mechanism.

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