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# Effect of Interlayer Interaction in Outer and Inner Layers of Cuprate Superconductor's Energy Gap and Excitonic Correlations\*

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Abstract: The role of interlayer interaction in outer and inner layers of Cuprate Superconductors has been investigated with in the BCS theory irrespective of the pairing mechanism. The expression for interlayer, intralayer order parameter ( $\Delta$ ) and excitonic correlation parameter ( $\gamma$ ) for inner and outer layers has been obtained and numerically solved. We have explained relevance of our results in context of observation of two gaps and presence of pseudogap in underdoped cuprates. It is also found that interlayer order parameter for inner layers is considerably lower then for outer layers and mere addition of layers cannot increase  $T_c$  indefinitely.

Keywords: Multilayer; Superconductors; Oxides; Superconductivity.

### 1. Introduction

After the discovery of Superconductivity in 1911 by Kamerling Onnes, for about 70 years scientists were not able to push the transition temperature beyond about twenty degrees of absolute zero (Tc~23K for Nb<sub>3</sub>Ge). Only after the discovery<sup>1</sup> in 1986 of materials that become superconducting at much higher Tc, the pursuit of room temperature superconductivity is revived. The highest Tc at the moment is 170K for Hg based superconductors.

The high Tc Superconductors have many peculiar properties. One of them is the link between their layered structure and transition temperature

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and other is development of pseudogap well above Tc in underdoped cuprates.

It is now well established that increase in CuO<sub>2</sub> layers per unit cell increases Tc, being maximum for n=3 and then decreases<sup>2,3</sup>. Role of interlayer interaction is investigated by many workers<sup>4-11</sup> and are found to play significant role in enhancement of Tc and stabilising superconducting order with respect to fluctuations. Khandka and Singh<sup>12</sup> recently showed that increasing number of layers alone cannot increase Tc but density of state also should increase to ensure enhancement of Tc. But increasing no. of layers beyond n=3 results in redistribution of charge, giving rise to underdoped inner layers and overdoped outerlayers<sup>13</sup>, and density of state no more remains a monotonous function of number of layers. This limits the increasing Tc. Chakraverty et al.<sup>14</sup> suggested a competing order along with interlayer interaction in the inner layers which could effectively bring down Tc for n > 3 and explained the Tc Vs n behaviour satisfactorily. Therefore a critical attention to the inner layers of these materials is needed. When inner layers are underdoped, bulk Tc is low<sup>13</sup>. A similar situation is present in underdoped cuprates, which have low Tc and are accompanied by pseudogap<sup>15-17</sup>. A question arises whether a common cause is manifested in the two situations?

It is still unclear what relationship pseudogap is having with superconductivity, whether they are different manifestation of same correlations or they are different phenomenon altogether. Giving this debate a fresh look is a recent experiment of Angle resolved photoemission spectroscopy (ARPES) and Scanning Tunnelling microscopy<sup>18-20</sup> which suggest the presence of two gaps in underdoped cuprates, and one of the two (gaps) vanishes at Tc. This along with a distinct temperature and doping dependence of gap seems to suggest a competing nature between Superconducting gap and pseudogap<sup>18, 19</sup>. A similar competing order is also being suggested<sup>14</sup> in inner layers of cuprate superconductors by Chakrevarty et al. as stated earlier.

In view of above here we present a microscopic approach to investigate the pairing gap with in the superconducting region by considering interlayer interaction for inner as well as outer layers with in BCS theory but irrespective of the pairing mechanism to have some insight into the pseudogap region.

## 2. Mathematical formulation

The Hamiltonian for our system can be described as:

(1) 
$$H = \sum_{ik\sigma} E_k C^{\dagger}_{ik\sigma} C_{ik\sigma} - \sum_{ikk'} V_{ii}(kk') C^{\dagger}_{ik'\uparrow} C^{\dagger}_{i-k'\downarrow} C_{i-k\downarrow} C_{ik\uparrow} + C_{i-k\downarrow} C_{ik\uparrow} + C_{i-k\downarrow} C_{ik\uparrow} + C_{i-k\downarrow} C_{ik\uparrow} + C_{i-k\downarrow} C_{j-k\downarrow} C_{jk\uparrow} + C_{i\neq j} + C_{i\neq j} + C_{i\neq j} + C_{i+k\downarrow} + C_$$

where,  $C^+_{ik\sigma}$ ,  $C_{ik\sigma}$  denoted the fermion creation & annihilation operator respectively, K is the wave vector and  $\sigma$  is spin index for fermions.

In equation (1) the first term is the energy of the free charge carriers within the  $CuO_2$  planes. The second term describes BCS type intralayer attractive interaction originating from any proposed mechanism<sup>7</sup> and the third term represents interlayer pairing<sup>8</sup> and V<sub>ij</sub> represents the attractive interactions containing contribution from any proposed mechanism mediated interaction<sup>9, 10-16</sup>.

Here we obtain self consistent expression for interlayer correlation parameter ( $\gamma_{\perp}$ ) and order parameter for outer layers and inner layers. We apply Green's function technique<sup>24</sup>.

In our present analysis we use a Green's function, defined as-

(2) 
$$G_{rrqq}^{\uparrow\uparrow} = \left\langle \left\langle C_{rq}\uparrow, C_{rq}^{+}\uparrow\right\rangle \right\rangle,$$

and writing equation of motion as-

(3) 
$$\omega G \stackrel{\uparrow \uparrow}{rrqq} = \frac{1}{2\pi} + \left\langle \left\langle [C_{rq}\uparrow,H],C_{rq}^{+}\uparrow\right\rangle \right\rangle.$$

Now, evaluating the commutator  $[C_{rq\uparrow}, H]$  using the Hamiltonian (1), we get

$$[C_{rq}\uparrow,H] = E_q C_{rq}\uparrow -\sum_{K} V_{rr}(kq)C_{r-q}^{\dagger}\downarrow C_{r-k}\downarrow C_{rk}\uparrow -\sum_{\substack{j \\ k \\ j\neq r}} V_{rj}(kq)C_{r-q}^{\dagger}\downarrow C_{j-k}\downarrow C_{jk}\uparrow$$

Putting the value of commutator  $[C_{rk\uparrow}, H]$  in the equation (3) we get

$$(4) \quad \omega G_{rrqq}^{\uparrow\uparrow} = \frac{1}{2\pi} + \left\langle \left\langle E_q C_{rq} \uparrow, C_{rq}^+ \right\rangle \right\rangle - \sum_K V_{rr}(kq) \left\langle \left\langle C_{r-q}^+ \downarrow C_{r-k} \downarrow C_{rk} \uparrow, C_{rq}^+ \right\rangle \right\rangle - \sum_{\substack{jk \\ j \neq r}} V_{rj}(kq) \left\langle \left\langle C_{r-q}^+ \downarrow C_{j-k} \downarrow C_{jk} \uparrow, C_{rq}^+ \right\rangle \right\rangle \right\rangle.$$

Now we introduce the order parameter  $\Delta$  such as

$$\Delta = \sum_{k} V_{rr} (kq) \left\langle C_{rk}^{+} \uparrow C_{r-k}^{+} \right\rangle$$
$$\Delta = \sum_{k} V_{rj} (kq) \left\langle C_{jk}^{+} \uparrow C_{j-k}^{+} \right\rangle.$$

Substituting these order parameters in equation (4), finally we obtained the equation

(5) 
$$(\omega - E_q) G_{rrqq}^{\uparrow\uparrow} = \frac{1}{2\pi} - (\Delta_{\coprod} + \sum_{\substack{j \\ j \neq r}} \Delta_{\perp j}) G_{rr-qq}^{\downarrow\uparrow},$$

where,  $G_{rr-qq}^{\downarrow\uparrow}$  is another Green's function, which may be written as:-

(6) 
$$G_{rr-qq}^{\downarrow\uparrow} = \left\langle \left\langle C_{r-q\downarrow}^+, C_{rq\uparrow}^+ \right\rangle \right\rangle .$$

This Green's function may also may be written in term of equation of motion as:

(7) 
$$\omega_{G_{rr}-qq} = \left\langle \left\langle [C_{r-q\downarrow}^{+},H],C_{rq\uparrow}^{+} \right\rangle \right\rangle.$$

Evaluating the Commutator  $[C_{r-q\downarrow}^+, H]$  using the Hamiltonian (1)

$$\begin{split} [C_{r-q\downarrow}^{+},H] = [C_{r-q\downarrow}^{+},\sum_{ik\sigma} E_{K}C_{ik\sigma}^{+}C_{ik\sigma}] - [C_{r-q\downarrow}^{+},\sum_{ikk'} V_{ii}(kk')C_{ik'\uparrow}^{+}C_{i-k'\downarrow}^{+}\\ C_{i-k\downarrow}C_{ik\uparrow\uparrow}] - [C_{r-q\downarrow}^{+},\sum_{\substack{ijkk'\\i\neq j}} V_{ij}(kk')C_{ik'\uparrow}^{+}C_{i-k'\downarrow}^{+}C_{j-k\downarrow}C_{jk\uparrow}]. \end{split}$$

280

Substituting the value of  $[C^+_{r-q\downarrow}, H]$  in equation (7)

$$\begin{split}
\omega G_{rr-qq}^{\downarrow\uparrow} &= -E_{-q} G_{rr-qq}^{\downarrow\uparrow} - \sum_{k'} V_{rr}(qk') \left\langle C_{rk'\uparrow}^+, C_{r-k'\downarrow}^+ \right\rangle \left\langle \left\langle C_{rq\uparrow}, C_{rq\uparrow}^+ \right\rangle \right\rangle \\
\end{split}$$
(8)
$$- \sum_{\substack{k'\\i\neq r}} V_{ir}(k'q) \left\langle C_{ik'\uparrow}^+, C_{i-k'\downarrow}^+ \right\rangle \left\langle \left\langle C_{rq\uparrow}, C_{rq\uparrow}^+ \right\rangle \right\rangle.
\end{split}$$

But from the law of conservation of energy  $E_{-q} = E_q$ . So

(9) 
$$(\omega + E_q) G_{rr-qq}^{\downarrow\uparrow} = -(\Delta_{\coprod} + \sum_j \Delta_{\perp j}) G_{rrqq}^{\uparrow\uparrow} .$$

Now, by the help of equation (5) & equation (9), we obtain both Green's functions  $G_{rrqq}^{\uparrow\uparrow} \& G_{rr-qq}^{\downarrow\uparrow}$  as

(10) 
$$G_{rrqq}^{\uparrow\uparrow} = \frac{(\omega + E_q)}{2\pi \left[\omega^2 - E_q^2 - (\Delta \coprod + \sum_j \Delta_\perp j)^2\right]},$$

(11) 
$$G_{rr-qq}^{\downarrow\uparrow} = \frac{-(\Delta \coprod + \Sigma \Delta \perp j)}{2\pi \left[\omega^2 - E^2 q - (\Delta \coprod + \Sigma \Delta \perp j)^2\right]},$$

Using these Green's functions, we can obtain the expression for order parameter  $\Delta$  & correlation parameter  $\gamma$ . The interlayer order parameter  $\Delta_{\perp}$  may be written as

(12) 
$$\Delta_{\perp} = \sum_{k'} V_{jr} (k'q) \left\langle C^{+}_{jk'} \uparrow C^{+}_{j-k'} \right\rangle.$$

Correlation function  $\langle C_{jk'\uparrow}^+ C_{j-k'\downarrow}^+ \rangle$  is related to Green's function  $_{G_{jj-qq}}^{\downarrow\uparrow}$  as

(13) 
$$\left\langle C_{\omega}C_{\omega}\right\rangle = -\frac{1}{i}\int_{-\infty}^{\infty}\frac{G_{jj-qq}^{\downarrow\uparrow}(\omega+i\varepsilon)-G_{jj-qq}^{\downarrow\uparrow}(\omega-i\varepsilon)}{e\frac{\omega}{kT}-\eta}d\omega,$$

where  $\eta = -1$ , for fermion, K = Boltzmann constant & T = Temperature.

Green's function  $G_{jj-qq}^{\downarrow\uparrow}(\omega + i\varepsilon) \& G_{jj-qq}^{\downarrow\uparrow}(\omega - i\varepsilon)$  may be expressed as

(14) 
$$G_{jj-qq}^{\downarrow\uparrow}(\omega+i\varepsilon) = \frac{-(\Delta \coprod + \Sigma \Delta \bot i)}{2\pi [(\omega+i\varepsilon)^2 - Eq^2 - (\Delta \coprod + \Sigma \Delta \bot i)^2]},$$

(15) 
$$G_{jj-qq}^{\downarrow\uparrow}(\omega-i\varepsilon) = \frac{-(\Delta \coprod + \Sigma \Delta_{\perp i})}{2\pi[(\omega-i\varepsilon)^2 - Eq^2 - (\Delta \coprod + \Sigma \Delta_{\perp i})^2]}.$$

Substitute both Green's functions  $G_{jj-qq}^{\downarrow\uparrow}(\omega + i\epsilon) \& G_{jj-qq}^{\downarrow\uparrow}(\omega - i\epsilon)$  from equations (14) and (15) in equation (13) and then after solving, we get Correlation function

(16) 
$$\left\langle C_{jk'\uparrow}^{+}C_{j-k'\downarrow}^{+}\right\rangle = \frac{\left(\Delta\coprod+\sum\Delta_{\perp i}\right)}{\sqrt{E_{q}^{2}+(\Delta\coprod+\sum\Delta_{\perp i})^{2}}} \operatorname{Tanh}\frac{\sqrt{E_{q}^{2}+(\Delta\coprod+\sum\Delta_{\perp i})^{2}}}{2KT}$$

Then we can obtained the expression for interlayer order parameter  $\Delta_{\perp}$  by substituting correlation function in equation (12)-

(17) 
$$\Delta_{\perp} = \sum_{k'} V_{jr}(k'q) \frac{(\Delta \coprod + \sum \Delta_{\perp i})}{\sqrt{E_q^2 + (\Delta \coprod + \sum \Delta_{\perp i})^2}} Tanh \frac{\sqrt{E_q^2 + (\Delta \coprod + \sum \Delta_{\perp i})^2}}{2KT}.$$

Converting summation over k' into an integration with cut-off energy  $+\hbar\omega_D$  from the fermi level, we get

(18) 
$$\Delta_{\perp} = N_{0} \int_{0}^{\pm \hbar \omega D} \frac{\int_{i}^{(\Delta_{\coprod} + \Sigma \Delta_{\perp i})}}{\sqrt{E_{q}^{2} + (\Delta_{\coprod} + \Sigma \Delta_{\perp i})^{2}}} T_{anh} \frac{\sqrt{E_{q}^{2} + (\Delta_{\coprod} + \Sigma \Delta_{\perp i})^{2}}}{\sum_{2KT} dE_{q}}.$$

Interlayer order parameter is also calculated using Green's function from eq. (10) and (11), neglecting Interlayer interactions, we get

(19) 
$$\Delta_{\coprod} = N_0 \int_0^{\hbar \omega_D} V_{rr} \frac{(\Delta_{rr})}{\sqrt{E_q^2 + (\Delta_{rr})}} Tanh \frac{\sqrt{E_q^2 + (\Delta_{rr})^2}}{2KT} dE_q.$$

Equation (18) and (19) are numerically solved to find interlayer ( for inner and outer layers) and intralayer order parameter for different values of temperatures for multilayer cuprates (n=4) .Curve between  $\Delta$  Vs T deferent is plotted.

The interlayer and intralayer excitonic correlation is expressed as

(20) (a) 
$$\gamma_{\perp} = \sum_{K} \left\langle C_{sk\uparrow}^{+}, C_{rk\uparrow} \right\rangle$$
, (b)  $\gamma_{\coprod} = \sum_{K} \left\langle C_{rk\uparrow}^{+}, C_{rk\uparrow} \right\rangle$ .

The correlation function  $\gamma_{\perp}$  may be expressed in terms of Green's function as

(21) 
$$\left\langle C_{sq\uparrow}^{+}C_{rq\uparrow}\right\rangle = -\frac{1}{i}\int_{-\infty}^{\infty}\frac{G_{rsqq}^{\uparrow\uparrow}(\omega+i\varepsilon)-G_{rsqq}^{\uparrow\uparrow}(\omega-i\varepsilon)}{\frac{\omega}{e\,kT}-\eta}d\omega$$
,

where  $\eta = -1$ , for fermion, K = Boltzmann constant & T = Temperature. The Green's functions  $G_{rsqq}^{\uparrow\uparrow}$  ( $\omega + i\epsilon$ ) &  $G_{rsqq}^{\uparrow\uparrow}$  ( $\omega - i\epsilon$ ) may be written as

$$G_{rsqq}^{\uparrow\uparrow}(\omega+i\varepsilon) = \frac{(\omega+i\varepsilon)+E_q}{2\pi[(\omega+i\varepsilon)^2 - E^2_q - (\Delta_{\coprod} + \sum_{j}\Delta_{\perp j})^2]}$$

$$G_{rsqq}^{\uparrow\uparrow}(\omega-i\varepsilon) = \frac{(\omega-i\varepsilon)+E_q}{2\pi[(\omega-i\varepsilon)^2 - E^2_q - (\Delta \coprod + \sum_j \Delta_{\perp j})^2]}.$$

Substitute both Green's functions  $G_{rsqq}^{\uparrow\uparrow}$  ( $\omega + i\varepsilon$ ) &  $G_{rsqq}^{\uparrow\uparrow}$  ( $\omega - i\varepsilon$ ) in equation (21) & than after solving, we get Correlation function for  $\gamma$ 

(22) 
$$\left\langle C_{sk}^{\dagger}\uparrow^{*}C_{rq}\uparrow\right\rangle = \sum_{K} \frac{E_{q}}{2\sqrt{E_{q}^{2}+(\Delta \coprod^{+\sum\Delta_{\perp}j})^{2}}} Tanh \frac{\sqrt{E_{q}^{2}+(\Delta \coprod^{+\sum\Delta_{\perp}j})^{2}}}{2KT} - \frac{1}{2}.$$

Substituting eq. (22) in eq. (20), the expression for excitonic correlation is obtained as

$$\gamma_{\perp} = \sum_{k} E_{q} \frac{1}{2\sqrt{E_{q}^{2} + (\Delta_{\perp} + \sum_{j} \Delta_{\perp} j)^{2}}} Tanh \frac{\sqrt{E_{q}^{2} + (\Delta_{\perp} + \sum_{j} \Delta_{\perp} j)^{2}}}{2KT} - \frac{1}{2}.$$

Converting summation over K' into an integration with cut-off energy  $+\hbar\omega_D$  from the fermi level, and also multiply by N<sub>o</sub> then we get the correlation parameter  $\gamma$  as

(23) 
$$\gamma_{\perp} = N_0 \int_{0}^{\hbar\omega_{\rm D}} E_q \left[ \frac{1}{\sqrt{E_q^2 + (\Delta_{\perp} + \sum \Delta_{\perp j})^2}} Tanh \frac{\sqrt{E_q^2 + (\Delta_{\perp} + \sum \Delta_{\perp j})^2}}{2KT} - \frac{1}{2} \right] dE_q$$

and

(24) 
$$\gamma_{I]} = N_0 \int_{0}^{\hbar\omega_D} E_q \left[ \frac{1}{\sqrt{E_q^2 + (\Delta \Pi)^2}} Tanh \frac{\sqrt{E_q^2 + (\Delta \Pi)^2}}{2KT - 2} \right] dE_q.$$

By using equation (23) and (24), we numerically calculated interlayer (inner and outer layers) and intralayer excitonic correlations for different values of temperature respectively and curve of  $\gamma$  Vs T is drawn.

## 3. Results and Discussion

In the present work we tried to investigate the role of inner and outer layers with interlayer interaction on superconducting order parameter and excitonic type of correlations in high Tc cuprate superconductors and hence to have some insight into the pseudogap region.

For this, expression of interlayer order parameter for inner  $(\Delta_{\perp i})$  and outer layer  $(\Delta_{\perp 0})$  is obtained along with intralayer order parameter  $(\Delta_{11})$  from equation (18 & 19), for n=4 and n=1 CuO<sub>2</sub> layers per unit cell. Constant density of states is assumed at fermi level with cut off energy  $\pm\hbar\omega_D$ , taking  $\hbar\omega_D=0.0862$ ev.

These equations are numerically solved as a function of temperature. Fig (1) shows the curve between superconducting order parameter( $\Delta$ ) and temperature(T). It is clear from the curve that interlayer order parameter ( $\Delta_{\perp \perp}$ &  $\Delta_{10}$ ) exceeds intralayer ( $\Delta_{11}$ ) order parameter at temperatures close to Tc. We attribute this robustness of interlayer order parameter to their ability to either suppress or screen fluctuations close to Tc. This is supported<sup>14, 21, 22</sup> by the fact that fluctuations depresses Tc more for n=1 then for n=3. This in turn helps the superconducting state to stabilize and increase Tc. Fig.(1) also shows the interlayer order parameter for inner layers is less than for outer layers. Our results show that increasing n beyond 3 would amount for more inner layers then outer layers, and since inner layers are not contributing much towards increasing Tc, increasing no. of layers alone cannot bring about an indefinite increase in Tc. Also increasing no. of CuO<sub>2</sub> layers beyond n = 3, the inner layers become under doped and a competing order<sup>14</sup> may develop to bring the Tc down. This situation is similar to the under doped cuprates where competing order is suggested<sup>18, 19</sup> in pseudogap region and which can decrease the Tc in them. This draws support from the experiments predicting two gaps, one of which vanishes<sup>18, 20, 23</sup> at Tc.

It seems underdoped materials give rise to low Tc and if pseudogap is universal in them, presence of pseudogap in inner layers of multilayer cuprates is a distinct possibility.

In order to have an insight into the pseudogap region, expression for interlayer excitonic correlations is also obtained (Eq. (23)). It is numerically solved for inner layers ( $\gamma_{\perp i}$ ), outer layer ( $\gamma_{\perp 0}$ ) and intralayer ( $\gamma_{II}$ ) excitonic correlations (eq. (24)) as a function of temperature. Fig (2) shows the curve of  $\gamma$  Vs T. The curve shows that  $\gamma_{\perp 0}$  and  $\gamma_{\perp i}$  falls rapidly close to Tc as superconducting state is reached from above. This can be explained in terms of reduced fluctuation close to Tc due to interlayer interaction. This in turn strengthens  $\Delta_{\perp i} \& \Delta_{\perp 0}$ , which is supported by our results. Also fig (2) shows intralayer excitonic correlation ( $\gamma_{II}$ ) to be independent of temperature and thus decrease intralayer order parameter more close to Tc .

In conclusion we can say interlayer interactions are indeed helping to maintain high Tc as long as inner layers don't become underdoped. As no of layers are increased the effective contribution of inner layers goes on decreasing by virtue of redistribution of charges<sup>13</sup>. So for optimum number of  $CuO_2$  layers per unit cell, maximum Tc is obtained. Doping the cuprates also bears a close resemblance. Increasing doping first produce pseudogap with competing pairing, low Tc & later overdoped region again with low Tc .At critical doping the competing order is absent and the hole concentration is right enough to produce superconductivity at high Tc. Careful experiments to search for pseudogap in inner layers of multilayer cuprates is very much desirable and can throw light on universality of Tc variation with no. of layers n & doping x.



Fig. 1. Delta ( $\Delta$ ) vs Temperature (T)



Fig. 2. Gamma ( $\gamma$ ) vs Temperature (T)

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