CHARGE TRANSFER POLARISATION WAVE IN HIGH T_c OXIDES AND SUPERCONDUCTIVE PAIRING

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ABSTRACT

A general formalism of quantised charge transfer polarisation waves has been developed. The nature of possible superconductive pairing between oxygen holes is discussed. Unlike optical phonons these polarisation fields will give rise to dielectric bipolarons or bipolaron bubbles. In the weak coupling limit a new class of superconductivity is to be expected.

INTRODUCTION

The relevance of charge transfer excitation to superconductivity was first pointed out by Varma [1] et al. The idea of such a local short range interaction has since been extended to a variety of charge transfer excitations [2] in the high T_c oxides. This paper has its inspiration in the idea of polarisation waves that goes back to Hopfield [3] and revived recently by Aschcroft [4] with respect to the high T_c oxides. The basic notion of long range dipole-dipole interaction giving rise to a collective and quantised charge transfer wave is the key ingredient. Some of the detailed calculations are presented elsewhere [5].

A - POLARISATION WAVE FORMALISM

We want to make essential points clear. In the Hartree-Fock approximation, the state of an **Unit Cell of CuO_2** square is a Slater determinant of the occupied lowest energy orbital states. Consider an excitation α corresponding to moving one electron from a ground state to an excited state orbital costing an energy α. Figure 1 gives the relevant energy levels [6] ; figure 1a shows the charge transfer insulator gap energy E_g for zero-doping, while the arrows in figure 1b indicate the charge transfer excitation energy ω_g to the Fermi level on p-type doping (1 signifying a ligand hole or $2p^5$ configuration).

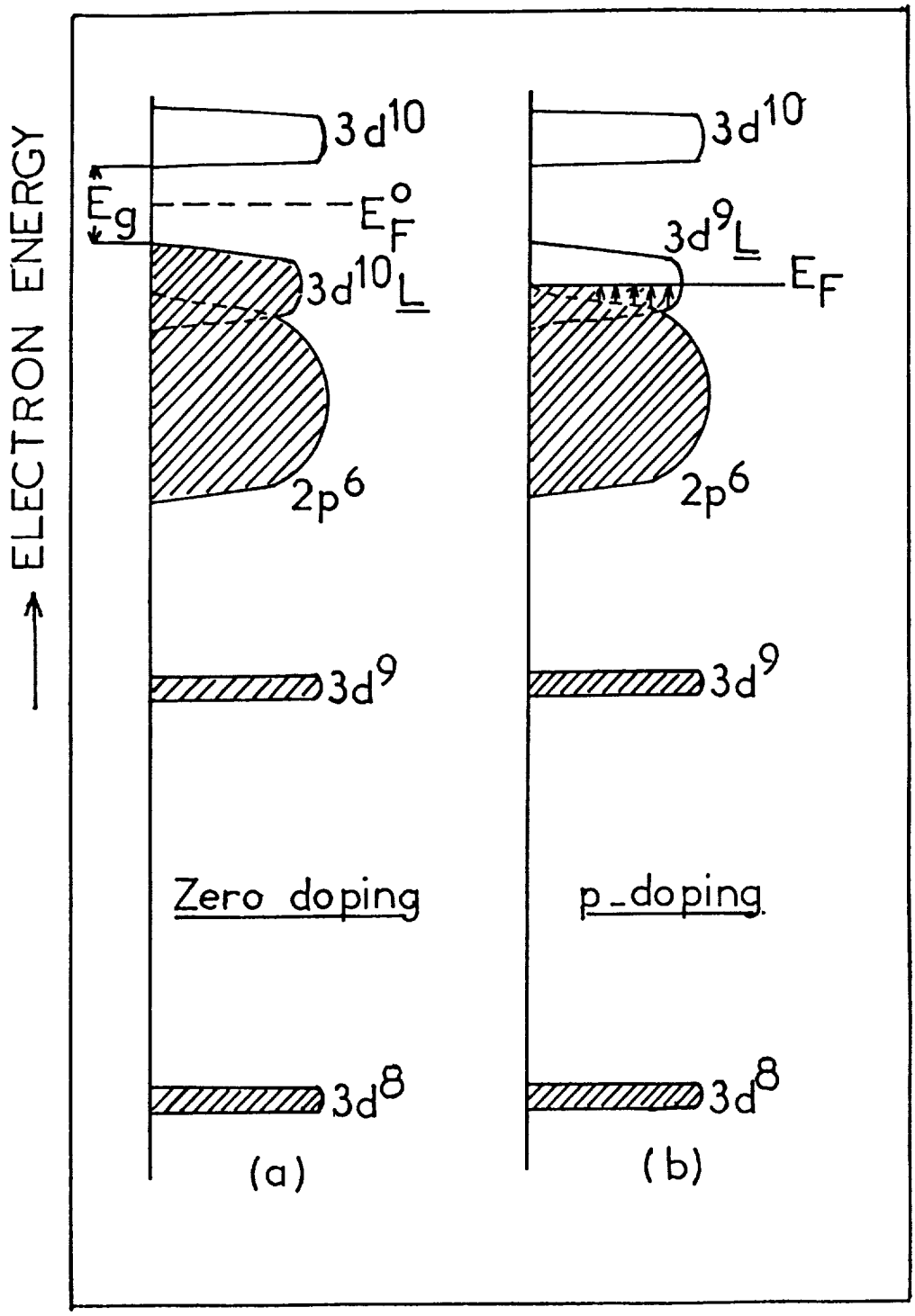

Figure 1a. Energy band--updoped material
Figure 1b. Energy band--doped material

We define by b_α^+ the operator to describe this excitation, which can be written in terms of the operator a_n of the electronic state

$$b_\alpha^+ = a_m^+ a_n \tag{1}$$

We approximate Hamiltonian of a **single cell** by

$$H = E_g + \sum_\alpha \omega_\alpha b_\alpha^+ b_\alpha \tag{2}$$

E_g : ground state energy

Define a **dipole matrix element**

$$\mu(\alpha) = \int d^3 r \ \phi_m^*(r) \ \vec{x} \ \phi_n(r) \tag{3}$$

Where the two orbitals ϕ_n and ϕ_m differ by one unit of angular momentum. Introduce the **polarisation operator** in the vector direction $\vec{x}$ for the unit cell by

$$P_x = e \sum_\alpha \mu_x(\alpha) \ (b_\alpha + b_\alpha^+) \tag{4}$$

This operator has the units of polarisation, which is charge times length - analogous to the displacement operator for a harmonic oscillator, which has the unit of length $\sim \left(\frac{\hbar}{2 m \omega}\right)^{\frac{1}{2}}$. We use the symbol $G_{xy}(i\omega)$ to denote the denote the retarded correlation function of the P operator with itself by

$$G_{xy}^0(i\omega) = - \int_0^\beta e^{i\omega\tau} \left\langle T_\tau \ P_x(\tau) \ P_y(0) \right\rangle d\tau \tag{5}$$

It is easily evaluated for the non interacting Hamiltonian (2).

$$G_{XY}^0(i\omega) = \sum_\alpha \frac{2 e^2 \ \mu_x \mu_y \omega_\alpha}{(i\omega)^2 - \omega_\alpha^2} \tag{6}$$

The quantity $- G^0(i\omega)$ is **actually the polarisability $\alpha^0(i\omega)$ of the unit cell.**

Assuming isotropy

$$\alpha(i\omega) = 2e^2 \sum_{\alpha} \frac{\mu^2(\alpha)\,\omega_\alpha}{\omega_\alpha^2 - (i\omega)^2} \tag{7}$$

So far our discussion has been confined to the properties of a single unit cell CuO_2. However when one excites dipoles in a cell i, it has **long-range dipole-dipole interaction** with dipoles of the cell j, given by

$$H' = \vec{X_i} \,.\, \phi(R_i - R_j) \,.\, \vec{X_j} \tag{8}$$

Where :

$$\phi_{xy}(R) = \frac{\delta_{xy}}{R^3} - \frac{3R_x R_y}{R^3} \tag{8a}$$

Defining a Fourier transform

$$T_{xy}(k) = \frac{\Omega_0}{4\pi} \sum_{l} \phi_{xy}(R_l) \exp i k \,.\, R_l \tag{8b}$$

We get the resultant Hamiltonian in the k-space as

$$H = E_g + \sum_{\alpha,k} \omega_\alpha b^+_{\alpha k} b_{\alpha k} + \frac{1}{2} \sum_{k,\alpha\beta} V_{\alpha\beta}(k) \left[b_{\alpha,k} + b^+_{\alpha,-k}\right]\left[b_{\beta,-k} + b^+_{\beta,k}\right] \tag{9}$$

where :

$$V_{\alpha\beta}(k) = \frac{4\pi e^2}{\Omega_0} \sum_{xy} \mu_x(\alpha)\, T_{xy}(k)\, \mu_y(\beta) \tag{9b}$$

Thus the Dyson equation for the correlation function is the matrix equation

$$G_{xy}(k, i\omega) = G^0_{xy}(k, i\omega) + \frac{4\pi}{\Omega_0} \sum_{x'y'} G^0_{x\,x'}(k, i\omega)\, T_{x'y'}(k)\, G_{y\,y'}(k, i\omega) \tag{10}$$

Solution of this equation will give us the collective excitation wave, that we have called charge transfer polarisation waves. In a cubic symmetry, it is solved to give

$$G_{xy} = -\frac{\alpha}{1 - \frac{4\pi\alpha}{3\Omega_0}} \left[\delta_{\mu\nu} - \frac{\frac{4\pi\alpha}{\Omega_0}}{\left(1 + \frac{8\pi\alpha}{3\Omega_0}\right)} \frac{k_x k_y}{k^2}\right] \tag{11}$$

The quantity-G_{xy} is the **static polarisability of the solid**, just as - G^0_{xy} is the static polarisability of the unit cell. This gives us the dielectric function at long wave-length which has the Lorentz-Lorentz form

$$\varepsilon(\omega) = \frac{1 + \frac{8\pi}{3\Omega_0}\,\alpha(k,\omega)}{1 - \frac{4\pi}{3\Omega_0}\,\alpha(k,\omega)} \qquad (12)$$

The pole and zero of the dielectric function gives us the transverse and longitudinal charge transfer polarisation waves respectively.

$$\omega_t^2 = \omega_g^2 - \frac{1}{3}\,\Omega_p^2$$

$$\omega_l^2 = \omega_g^2 + \frac{2}{3}\,\Omega_p^2 \qquad (13)$$

B - THE GAP EQUATION

Interaction between two carriers due to the longitudinal charge transfer waves is given by

$$V(q,\omega) = \frac{4\pi e^2}{q^2\,\varepsilon(q,\omega)} \qquad (14)$$

Where $\varepsilon(q,\omega)$ is the dielectric constant as shown in fig (2), for a finite value of ω_t.

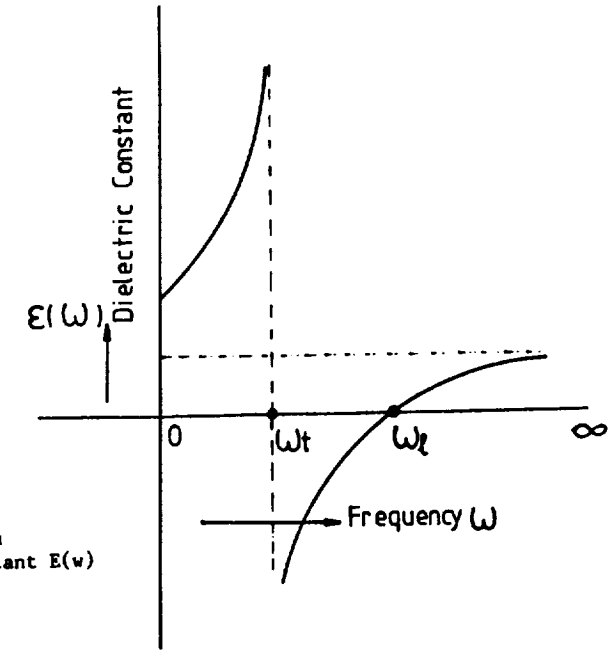

Fig. (2) - Long wave length dielectric constant E(w)

We see immediately **that excitations up to $\omega = \omega_t$ has repulsive interactions** between two carriers and is attractive between ω_t and ω_l. **A B.C.S. like gap nonetheless appears at the Fermi Surface.** The gap equation is given by

$$\Delta(\xi) = - \int_0^{\infty} \frac{K(\xi, \xi')}{\sqrt{\Delta^2(\xi') + \xi'^2}} \tanh \left[\frac{\sqrt{\Delta^2(\xi') + \xi'^2}}{2kT} \right] d\xi' \qquad (15)$$

We can mimic the repulsive interaction at frequencies $0 < \omega < \omega_t$ and an attractive interaction between $\omega_t < \omega < \omega_l$, by two **piecewise constant Kernels**

$$\begin{aligned} k(\xi,\xi') &= \lambda_1 && 0 < \xi,\xi' < \omega_1 \\ &= -\lambda_2 && \omega_1 < \xi,\xi' < \omega_2 \end{aligned} \qquad (16)$$

We also assume two energy gaps

$$\begin{aligned} \Delta(\xi) &= \Delta_1 && 0 < \xi < \omega_1 \\ &= \Delta_2 && \omega_1 < \xi < \omega_2 \end{aligned} \qquad (17)$$

The gap equation is easily solved to give these two gaps Δ_1 and Δ_2.

We have $$kT_c = 1.14\ \omega_1 \exp - \frac{1}{\tilde{\lambda}} \qquad (17a)$$

where $$\frac{1}{\tilde{\lambda}} = \frac{\lambda_2 g - 1}{D}$$

With $$g = \ln \frac{\omega_2}{\omega_1}$$

and $$D = \lambda_1 - \lambda_2^2 g$$

For $\frac{\omega_2}{\omega_1} = 6$, $\lambda_1 = 0.5$, $\frac{\lambda_2}{\lambda_1} = 0.76$ and $h\omega_1 = 0.5$ eV, we obtain a $T_c \sim 300°$ k.

The same formalism will apply for optical phonons. Using $\hbar\omega_1 = 0.1$ eV, $\frac{\omega_2}{\omega_1} = 2.25$ (corresponding to $\frac{\varepsilon_0}{\varepsilon_\infty} = 5$), $\lambda_1 = 0.3$, $\lambda_2 = 0.7$ gives a $\tilde{\lambda} = 0.6$ and a zero temperature gap $\Delta_1 = 36$ m eV. We thus see that a dipole lattice or a **Lorentz lattice in general** can give rise to a B.C.S. kind of pairing and an energy gap at the Fermi surface, inspite of a net repulsive interaction in the small frequency range, and the most general solution admits at least more than one energy gap ($\Delta_2 > \Delta_1$).

C - DIELETRIC POLARONS AND BIPOLARONS

The interaction of the carriers with the longitudianl polarisation field gives rise to polarons (Feynmann [7]). Will it give rise to bipolarons ? The literature is quite scarce on the subject. While the answer is quite affirmative with the acoustic bipolarons, it seems to be less so with its optical counterpart. Early [8] calculations seemed to indicate that for a favorable range of the dielectric constant ratios ($\frac{\varepsilon_0}{\varepsilon_\infty} \sim$ 10 - 20) we may be able to have a dielectric bipolaron. More recent path [9] integral calculations put this ratio even higher. What is certain [10] is that if the carrier behaves like a localized classical charge (i.e its interaction with polarisation field is recoil less), the phonon-mediated interaction will at best reduce the coulomb repulsion between the two charges form $\frac{e^2}{\varepsilon_\infty r}$ to $\frac{e^2}{\varepsilon_0 r}$ but does not lead to any attraction. We can see from figure 1b, that as the p-type doping is increased, there is an increasing component of low energy excitation ω_g (ω_g going down with ε_F) such that we expect $\omega_t \rightarrow 0$, at some critical doping value x_c, given by equation [13]. With the longitudinal frequency $\omega_l \rightarrow \sqrt{\frac{2}{3}}\,\Omega_p$ and because of the collapse of the Lorentz lattice, we can have the whole frequency range up to ω_l where the dielectric constant is negative. This low frequency attraction will give **instantaneous local interaction** between carriers, giving rise to (fig. 4b) **dielectric bipolarons or bipolaron bubble** (if its energy is embedded in the continuum of 1-particle states). We can model the effective Hamiltonian by

$$H_{eff} = \sum \varepsilon_k C_k^+ C_k - V \sum C_{k\sigma}^+ C_{k'\sigma'}^+ C_{k'\sigma'} C_{k\sigma} \qquad (18)$$

This Hamiltonian is strikingly different form the B.C.S Hamiltonian in not having the ω-cut off and its general behavior pattern has been indicated [11]. In the usual weak-coupling approximation and when the dielectric bipolaron interaction energy $V << \varepsilon_F$, the superconductivity gap equation has been solved [5] to give

$$\Delta = 8\,\varepsilon_F \exp -\frac{\varepsilon_F}{\varepsilon_c} \qquad (19)$$

Where ε_c is an energy cut-off.

It is necessary to recall the essential ingredients leading to the equation (18) signifying an instantaneous attractive interaction V between two carriers.

We can define two key dimensionless parameters in the pairing scenarios.

- The dimensionless coupling constant

$$\lambda = \frac{V}{E_F} \qquad (20a)$$

- Antiadiabaticity parameter

$$\gamma = \frac{\hbar\omega}{E_F} \qquad (20b)$$

Signifying ratio of boson frequency to the Fermi energy.

There are several interesting differences of the electronic properties in the normal state between the adiabatic ($\gamma << 1$) and the antiadiabatic ($\gamma >> 1$) regime. These differences[12] are shown schematically in figure (3a) and (3 b), as well as in fig. (4a) and (4b).

(a) The effective mass ratio $\frac{m^*}{m}$ is exponentially large (Holstein factor) around $\gamma = 1$ ($\lambda < 1$) but is completely unrenormalised in the antiadiabatic limit [5] (fig. 3a).

(b) The electron life time τ at the Fermi surface is quite different in the two regimes. In the adiabatic regime $\frac{1}{\tau} << kT \sim \frac{T^2}{E_F}$, which gives the usual metallic conductivity. In the antiadiabatic regime $\frac{1}{\tau} >> kT$ (fig. 3b) and is conjectured to be $\sim T$, if it behaves as a marginal Fermi liquid [13].

(c) In the adiabatic regime, the usual electron-phonon interaction (fig. 4a) gives the **retarded non local** attraction between electron-pairs. In the antiadiabatic regime the attraction is expected to be instantaneous and local (fig. 4b), forming a real-space electron-pair or a **dielectric bipolaron bubble.** The resulting normal and superconducting properties are bound to be different from the classical B.C.S. behavior.

In the B.C.S. behavior

$$\frac{kT_c}{E_F} = \gamma \exp - \frac{1}{\lambda} \qquad (21a)$$

In the other limit, $\gamma >> 1$

$$\frac{kT_c}{E_F} \doteq \exp - \frac{1}{\lambda} \qquad (21b)$$

In summary, we can say that presence of high frequency bosons ($\hbar\omega > E_F$) will lead to quasiparticules which are dielectric polarons with properties in the normal and superconducting states quite different from usual metals.

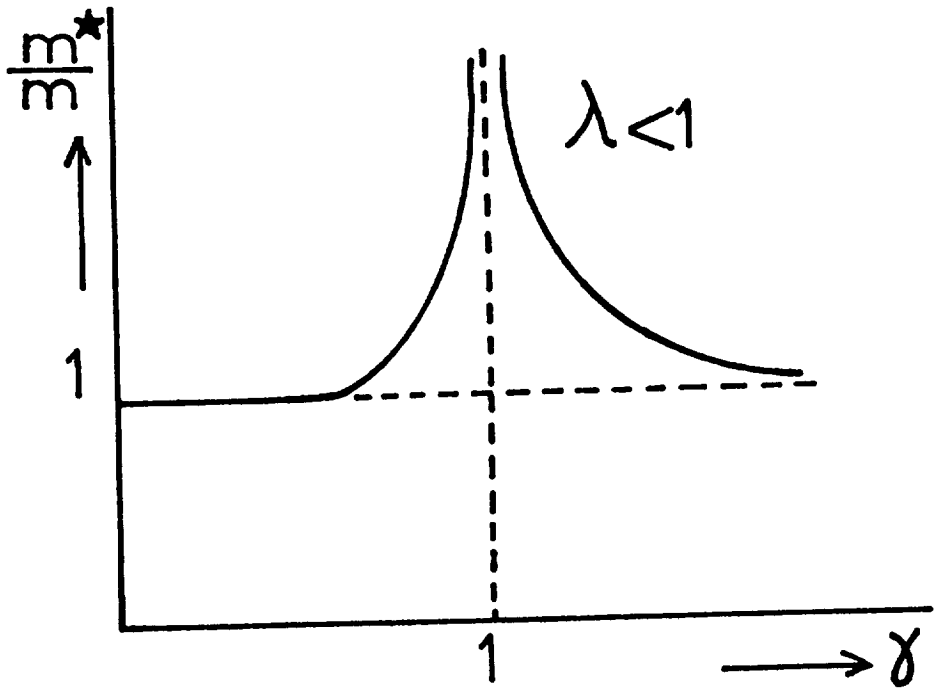

Fig. 3a

Fig. (3a) - Effective mass of carriers as function of $\gamma = \frac{\hbar\omega}{E_F}$ or antiadiabaticity.

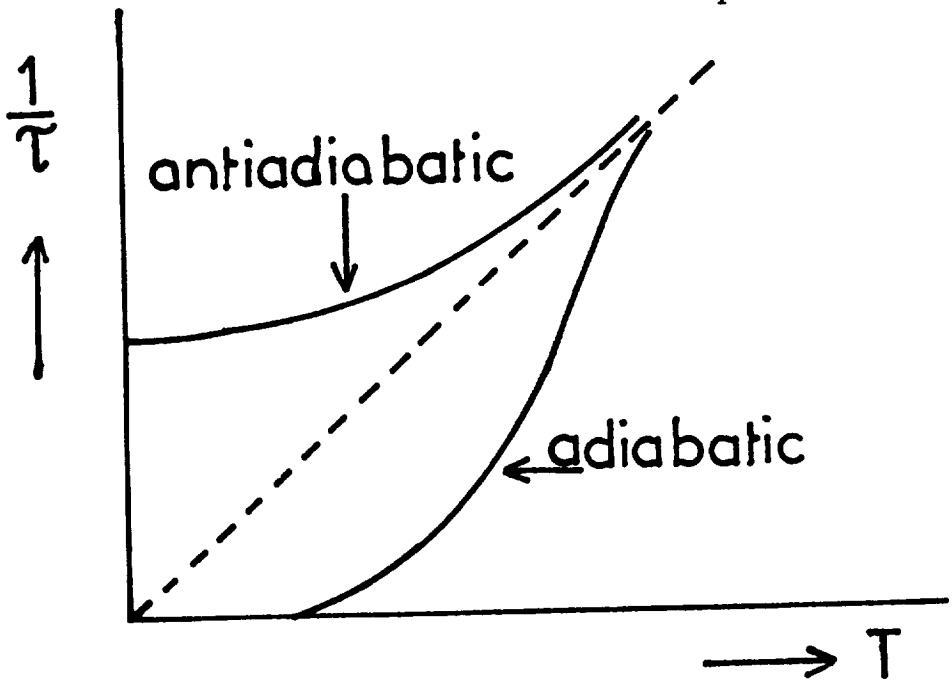

Fig. 3b

Fig. (3b) - A carrier life time τ as function of temperature

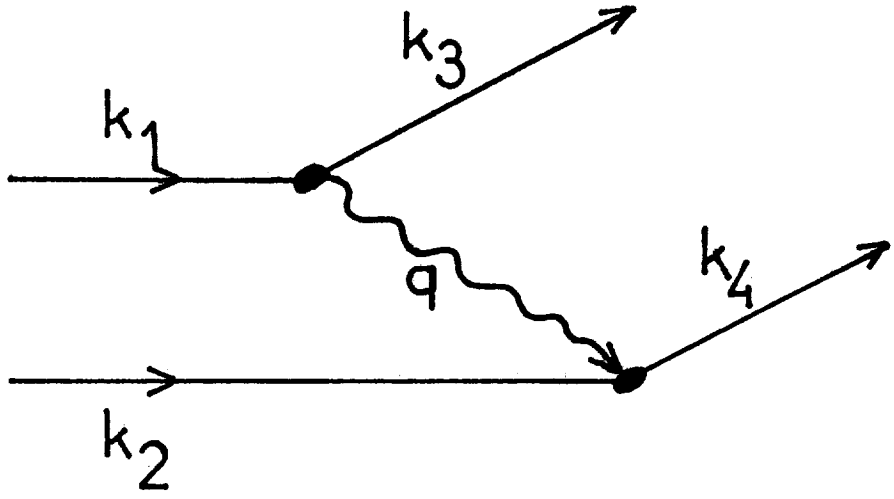

Fig. 4a

Fig. (4a) - Electron-electron interaction in the adiabatic regime

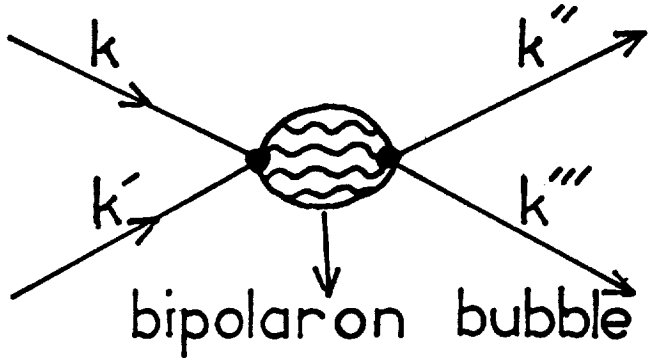

Fig. 4b

Fig. (4b) - Electron-electron interaction in the antiadiabatic regime

REFERENCES

1 - C.M.Varma, S. Schmitt-Rink and E. Abrahams
Sol. St Comm. 62, 681 (1987).

2 - B.K. Chakraverty, D. Feinberg, H. Zheng and M. Avignon
Sol. St Comm. 64, 1147 (1987.

W. Weber, Zeit Physik B 70, 323 (1988)/

J.E. Hirisch and S. Tang, Sol. St. Comm. 69, 987 (1989).

3 - J.J. Hopfield, Phys. Rev. 112, 1555 (1955).

4 - N.W. Aschcroft, "Recent Progress in Many body theories "vol. I Edited by E. Pajanne and R.F. Bishop (Plenum Publishing, N.Y. 1988) p 39.

5 - B.K. Chakraverty, Phys. Rev. B, April 1990.

6 - A. Bianconi et al
"Procceding Adriatico Research Conf. on High Temperature Superconductors"
Trieste Italy 6-8 July 1989
Edited Y. Lu, M. Tosi and E. Tosatti, World Scientific 1988.

7 - R.P. Feynman, Phys. Rev. 97, 660 (1955).

8 - V.L. Vinetskii and M. Sh. Giterman
J. Expt. Theoret. Phys. (USSR) 33, 560 (1958).

V.L. Vinetskii
Soviet Physics JETP, 13, 1023 (1961).

9 - H. Hiramoto and Y. Toyozawa
Jl. Phys. Soc. Jap 54, 245 (1985).

10 - W. Kohn
Phys Rev. 110, 857 (1958).

11 - Ph. Nozières and S. Schmitt-Rink
Jl Low Temp. Phys. 59, 195 (1985).

12 - D. Feinberg, S. Ciuchi and F. de Pasquale
Int. Jl. Modern Phys. B (1990 to be published.
Also
S. Aubry et al
"Workshop "Non-Linear Coherent Structures" Montpellier, France 1989 (Verlag-Springer)

13 - C.M. Varma et al
Phys. Rev. Letter 63, 1996 (1989).