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Abstract. – We review the present theoretical understanding and predictions for systems exhibiting real-space pairing of electrons. As the attractive interaction between electrons increases, the superconducting properties can change the BCS like behaviour gradually into superfluid like characteristics. The relevance of real-space pairing for high temperature superconductors is discussed.

1. Introduction

Many years ago already it was observed [1] that in systems which at that time were considered high temperature superconductors (A-15, Chevrel phases, YRh_4B_4...) a clustering of metal atoms occurs. This class of materials was also considered to exhibit strong electron-phonon interaction but the high critical temperature T_c was thought not to be related in any very simple way to this strong coupling. It was believed that electron pairing arises from an electronic mechanism with metal atoms imbedded in a highly polarizable dielectric [2]. The experimental verification of diatomic clusters [3] (Ti_4O_7, $Na_xV_2O_5$) gave renewed impetus to examine real space electron pairing and their resulting superconductivity [4]. These original studies were based on the bi-polaronic mechanism whereby an electron locally deforms the lattice which surrounds it. Two such electrons sense an attractive potential via their respective lattice deformations. In the limit of very strong electron-lattice coupling and the anti-adiabatic case, the problem of such bound electron pairs (bi-polarons) reduces to the problem of the extended negative U Hubbard problem in the strong attraction limit. Any attempts to study real space pairing for intermediate electron-lattice coupling in a situation intermediate between the antiadiabatic and the adiabatic cases have so far reached only little success. The general believe nonetheless is that real space pairing exists in a much wider regime than that studied so far, i.e. the non-adiabatic strong electron-lattice coupling limit.

A different and more phenomenological approach was taken by studying real space pairing directly on the basis of an effective extended negative U Hubbard model which permitted to interpolate between weak and strong attraction [5]. Such a non-retarded static short range interaction can result from the coupling between electrons and bosonic excitations like phonons, excitons or plasmons or can be of purely electronic origine due to strong polarizibility of anions.

These first attempts of treating real space pairing were restricted to onsite electron pairs where the site refered to well defined and separable electron clusters. The resulting superconductivity was found to correspond to the condensation of a charged Bose gas on a lattice. More recently the problem of intersite electron pairing has been treated [6]. This is a particularly interesting problem since it permits to take into account a repulsive Hubbard onsite correlation (giving rise to magnetic correlations) and intersite attraction (leading to superconductivity). On the basis of such a model the mutual influence of magnetism and superconductivity can be studied.

We believe that the recently discovered high T_c superconductors can (to within a first approximation) be described by such models of real space pairing. The coherence length in those materials is known to be of the order of a few lattice constants, compatible with the idea of real space pairing. The systems are certainly highly correlated ones and those correlations are taken into account in the model. Whether the origin of the pairing is linked to the resonating valence bond or is due to oxygen hole pairing in a two band Hubbard problem [7] or -as we think- due to chemical bonding, is at present an open question. The increasing critical temperatures [8] in the prototype real space pair system $BaBi_{1-x}Pb_xO_3$ and its derivatives, provides new confirmation for real space pairing arising from chemical bonding or electronic and polaronic mechanisms, entirely unrelated to magnetic correlations.

In this lecture we shall review the main features of real space superconductivity. We shall base our analyses onto i) the negative U Hubbard model for studying onsite pairing and ii) the extended Hubbard model (positive U and negative intersite interaction) for studying intersite pairing. The latter will be done for a square lattice in view of its relevance to high T_c materials.

The idea that bound electron pairs condense into a superfluid state and hence can lead to superconductivity had been proposed well before BCS [9]. At that time however no convincing arguments for a possible real space pairing mechanism could be given and finally the experimentally observed superconductivity did not in any sense resemble the superfluidity of a

charged Bose gas. This situation may have changed with the possibility of synthezising now highly complex compounds in which real space pairing has been verified either in form of bound states or virtual bound pairs (double valence fluctuations).

2. Superconductivity of onsite real space pairs

Systems with onsite pairs of electrons have now been verified experimentally. They contain cations which exist in two valence states differing by two electronic charges. Let us quote a few of them. $BaBi_xPb_{1-x}O_3$(Bi^{3+}, Bi^{5+}), $Pb_{1-x}In_xTe$ (In^{1+}, In^{3+}) $Pb_{1-x}Sn_xSe$ (Sn^{2+}, Sn^{4+}), $PbTl_xTe$ (Tl^{1+}, Tl^{3+}), Cs_2SbCl_6 (Sb^{3+}, Sb^{5+}). Some of these materials become superconducting in a given regime of x. The most notable candidate being $BaBi_xPb_{1-x}O_3$ and its more recent derivative $Ba_{0.6}K_{0.4}BiO_3$ which has a T_c as high as 30 K [8].

The simplest model which describes such a situation is the so-called extended negative U Hubbard model

$$H = \sum_{ij\sigma}{}' t_{ij} c^+_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \\ + \frac{1}{2} \sum_{ij\sigma\sigma'}{}' W_{ij} n_{i\sigma} n_{j\sigma'} - \mu \sum_{i\sigma} n_{i\sigma} \quad (2.1)$$

where the repulsive Coulomb interaction W_{ij} acting between electrons at two different sites i and j may be screened or unscreened. $c^{(+)}_{i\sigma}$ denote the creation (annihilation operators) for electrons at site i, having spin σ and $n_{i\sigma}$ their respective number operators. The hopping integral is denoted by t and the effective attractive onsite potential acting between two electrons of opposite spin is given by $U\,(<0)$. The chemical potential is denoted by μ. Depending on the relative value of $|U|/t$ one moves from one extreme limit: a BCS type state for $|U|/t \ll 1$ to another extreme limit: a Bose condensed state of a lattice gas of hardcore charged bosons for $|U|/t \gg 1$.

Let us focus on the latter since it is this limit which contains entirely new features of superconductivity. If we suppose that $|U|/t$ is big enough such that all electrons exist exclusively in form of onsite pairs one obtains by applying the standard degenerate perturbation theory the following effective Hamiltonian [4, 5]

$$\bar{H} = -\frac{1}{2} \sum_{ij}{}' J_{ij} \left(\rho^+_i \rho^-_j + H.c\right) + \sum_{ij}{}' K_{ij} \rho^z_i \rho^z_j \\ - B \sum_i (2\rho^z_i + 1) - \frac{N}{4} (J_0 + 2W_0) \quad (2.2)$$

where

$$J_{ij} = \frac{2t^2_{ij}}{|U|}, \quad K_{ij} = J_{ij} + 2W_{ij}, \quad B = \mu + \frac{1}{2}|U| - W_0 \quad (2.3)$$

and $J_0 = \sum_j J_{ij}$, $W_0 = \sum_j W_{ij}$. The charge operators

$$\rho^+_i = \left(\rho^-_i\right)^+ = c^+_{i\uparrow}\, c^+_{i\downarrow}\,, \quad \rho^z_i = \frac{1}{2}\left(n_{i\uparrow} + n_{i\downarrow} - 1\right) \quad (2.4)$$

having $s = 1/2$ Pauli statistics (fermions for identical sites and bosons for different sites) fully describe the hard core character of the onsite pairs.

The superconducting groundstate of the effective onsite pair Hamiltonian (2.2) is given by

$$|\psi\rangle \sim \prod_i \left(v_i\, e^{i\phi_i/2} + u_i\, e^{-i\phi_i/2}\, \rho^+_i\right)|0\rangle$$

which is the real space equivalent of the BCS state [10]. The v_i and u_i measure the relative occupation of a local pair at site i $\left(u^2_i + v^2_i = 1\right)$ and ϕ_i refers to the phase of the onsite pair wave function at site i. Depending on the relative values of J_{ij} and K_{ij} as well as the density of onsite pairs $n = (1/N)\sum_{i\sigma}\langle n_{i\sigma}\rangle$, the groundstate is either a homogeneous superconducting state with a homogeneous distribution of local pairs (v_i, u_i independent on i) and a globally phaselocked state (ϕ_i independent on i) or a mixed state where the concentration of onsite pairs spatially varies in a commensurate or incommensurate fashion together with a corresponding phase locked state [4, 5, 11]. This latter state has been called the mixed state and exhibits simultaneously superconductivity and CDW. The finite temperature phase diagram has been worked out for this problem within a self-consistent mean field scheme [5a] and is shown in figure 1. Similarly to superfluid ^{4}He II, the low lying excitations for the onsite pair problem equation (2.1) are collective oscillations of the local phases of the onsite pair wave functions. In a magnetic analogy they correspond to magnons of the pseudo spin operators (2.3). In the homogeneous superconducting state their dispersion is $\omega_k = s(T)|k| + \alpha k^2$ for small k where

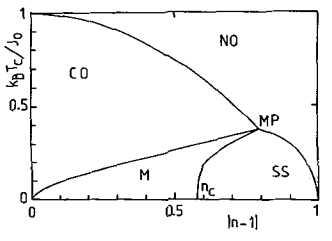

Fig. 1. – Typical phase diagram of the onsite pair system as a function of T and the electron concentration. MP is the multicritical point at which the following four phases meet: SS (homogeneous superconducting), M (mixed CDW-superconducting), CO (CDW) and NO (non-ordered onsite pairs). n_c denotes the zero temperature critical concentration separating M and SS.

the temperature dependent sound velocity $s(T) \sim (J_0(J_0+K_0))^{1/2}\langle\rho^x\rangle_T$, $\langle\rho^x\rangle_T = \frac{1}{2}\langle\rho^+ + \rho^-\rangle_T$ being the superconducting order parameter. As one increases the concentration of onsite pairs (going with n from 0 to 1) at zero temperature, $s(0)$ increases while ω_{k_B} (the frequency at the Brillouin zone) decreases and finally tends to zero at the critical concentration n_c separating the homogeneous superconducting from the mixed phase. In the mixed phase there are two branches of this collective oscillation, a soundwave like one relevant for the superfluidity and an optical branch relevant for the CDW. As the temperature increases the acoustic branch tends to zero ($s(T) \to 0$) at $T = T_c$ where superconductivity ceases to exist. Such a drastic effect onto the sound velocity arises from the fact that these onsite pairs are on a lattice and Umklapp process destroy the momentum of charge fluctuations.

In order to better appreciate the differences between real space pair and BCS superconductivity let us examine the extended negative U Hubbard problem (Eq. (2.1)) within a mean field approach [5b]. This permits us to follow the transition between weak and strong onsite pairing whereby the weak coupling limit in its mean field approach largely reflects the physics of a BCS superconductor. It has been shown [5, 12] that the groundstate for this model perfectly corresponds to a BCS variational groundstate which smoothly interpolates between weak and strong onsite attraction. Nonetheless concerning the excitations, they differ considerably in the two extreme limits. We schematically draw in figure 2 T_c/t for the model equation (2.1) as a function of $|U|/t$ in the absence of interpair interaction ($W_{ij} \equiv 0$) and for a concentration of electrons $n \cong 1$. Such a changeover of T_c from BCS to local pair superconductivity is valid for any n. We also plot the temperature for pairbreaking T_p/t and notice that upon increasing $|U|/t$ it first closely follows T_c/t and then starts increasing linearly with $|U|/t$ which simply signifies that in the strong coupling limit $|U|$ represents the binding energy

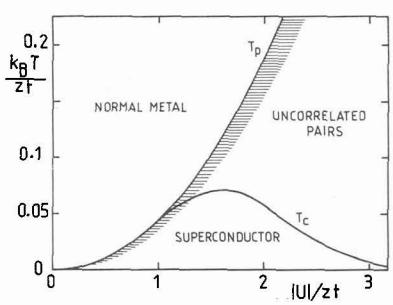

Fig. 2. – Schematic phase diagram of the negative U Hubbard model for $n \sim 1$. T_c denotes the superconducting critical temperature and T_p the pair breaking temperature.

of an isolated pair. This difference in the nature of excitations in the two extreme limits are clearly reflected in the thermodynamic and electrodynamic properties of these superconductors. In the weak coupling limit the specific heat and T_c are determined by the single particle excitations across the gap yielding

$$k_B T_c = 2D \times 0.57 \cdot \sqrt{n(2-n)} \exp(-2D/|U|)$$

and the characteristic BCS ratio $\Delta / k_B T_c = 3.52$ if one assumes a square density of states. Δ denotes the zero temperature energy gap and $D(=zt)$ the half bandwidth [5b]. The specific heat follows the BCS type behaviour $C_v \sim \exp(-\Delta/T)$. In the strong coupling limit these quantities are controlled by the sound wave like excitations yielding $k_B T_c = 3.31\left(na^{-3}\right)^{2/3}/m^*$ in the low density limit ($n \ll 1$) where the effective mass m^* of the onsite pairs is given by $m^* = 3\, J_0/a^2$ and a denotes the lattice constant. The numerical factor in the expression for $k_B T_c$ depends slightly on the crystal structure (3.31 corresponds to the simple cubic structure). In the high density limit $|n-1| \ll 1$ we have

$$k_B T_c = (J_0/2)\left[c^{-1} - \frac{1}{3}(n-1)^2\right]$$

where c denotes the Watson integral and for simple cubic structure $c = 1.5164$. The gap being of the order of $|U|$ in this limit leads to very large values of $\Delta / k_B T_c$. The specific heat being determined by the low lying sound wave like excitations behaves like $C_v \sim T^d$ for $T \to 0$ where d is the dimensionality of the system [14].

There is a changeover in T_c from a 3d Bosegas with anisotropic mass ($T_c \propto n^{2/3}\left(m_\perp/m_\parallel\right)^{1/3}/2m_\perp a^2$ for $1 < m_\parallel/m_\perp \lesssim 10^2$) to a quasi 2d Bose gas on a lattice with $T_c \sim \pi n/2m_\perp a^2$ for $m_\parallel/m_\perp \gg 1$. $m_\perp$ and $m_\parallel$ denote the mass of the Bosons in the basal plane and the c direction respectively.

Other essential differences between these two limits are linked to the effective size of the pairs or more precisely the range of interpair interaction. This has a noticeable effect on the width of the critical regime. In the BCS case this regime is very narrow, true critical behaviour will be unaccessible and hence a classical Ginzburg-Landau behaviour applies. In the opposite case of real space pairs the mean field approach looses its applicability near T_c and true critical behaviour occurs which will be that of a quantum $S = 1/2$ $X-Y$ model. This suggest [13] variations of the coherence length $\xi \sim \tau^{-\nu}$, the number of superconducting electrons $n_s \sim |\tau|^{2\beta}$, the Ginzburg-Landau ratio $\kappa \sim |\tau|^{-\bar{m}}$ and the fluctuation component of the resistivity $\sigma_s \sim \tau^{-s}$ as follows: $\nu = 1/2$ (2/3), $\beta = 1/2$ (1/3), $\bar{m} = 0$ (1/3) and $s = 1/2$ (1/3) for the weak (strong) coupling regime (in $d = 3$). τ denotes $(T - T_c)/T_c$. The specific heat is discontinuous in the

weak coupling regime and varies as $\tau^{-\alpha}$ $\left(\alpha \lesssim 10^{-2}\right)$ or $\ell n\,|\tau|$ in the strong coupling regime.

The Meissner effect and upper critical field H_{c2} are very sensitive to the size of the pairs. In the strong coupling limit this leads to very high values for H_{c2} at $T = 0$ and a positive curvature for H_{c2} near T_c [14]. It moreover leads to a penetration depth λ_H which is by a factor $(m^*/2m)^{1/2}$ bigger than the typical London penetration depth for the weak coupling regime [4].

3. Superconductivity of intersite real space pairs

Apart from onsite electron pairing there exist a variety of systems showing intersite pairing where two electrons pair up on well defined diatomic units in a crystalline lattice. These diatomic units show charge fluctuations differing by two electron charges. The most representative examples for such systems are $Ti_{4-x}V_xO_7$, $Li_{1+x}Ti_{2-x}O_4$ ($Ti^{3+} - Ti^{3+}$, $Ti^{4+} - Ti^{4+}$) and $Na_xV_2O_5$ ($V^{4+} - V^{4+}$, $V^{5+} - V^{5+}$). Depending on x these materials exhibit either a CDW ordered state where diatomic sites are alternatively occupied and unoccupied by intersite pairs or a state of statically disordered pairs. Above a certain temperature these pairs are dynamically disordered and show thermally activated hopping. At still higher temperatures these pairs eventually break up into individual electrons showing metallic behaviour. So far no superconductivity was observed in these materials (except for $Li_{1+x}Ti_{2-x}O_4$). The reason for that is that all these materials have a concentration of intersite pairs (per diatomic unit) which is close to 1. Following the arguments of the previous section and considering those intersite pairs as onsite pairs on diatomic sites we notice that close to $n = 1$ the system tends to order in a CDW state rather than a homogeneous superconducting phase. Any attempt to deviate from $n = 1$ by doping introduces impurity states in the crystalline matrix of the intersite pairs which would destroy any potential superconducting state for $n \neq 1$.

In order to create favorable conditions for intersite pairs we ought to look for systems where the dopant cations lie outside the matrix of the intersite pairs and also where there are no predestined diatomic sites favouring one valence state *via* another. The new high T_c cuprates have such properties considering that the intersite pairs (of holes) form on adjacent sites of oxygen ions fluctuating between the two valence states $\left(O^{2-} - O^{2-},\ O^{1-} - O^{1-}\right)$.

Let us in the following consider the superconducting properties of a system with intersite pairing on the basis of the Hamiltonian (2.1) for which $U > 0$ and $W_{ij} \equiv W < 0$ for i, j being nearest neighbour sites [6]. We are now confronted with a problem which contains both, onsite Hubbard U repulsion giving rise to magnetic correlations and intersite attraction giving rise to superconductivity. In general it is no longer possible (even in the strong attraction limit) to re-write this problem in terms of a hardcore charged Bose gas. The exception is the extremely dilute case. We therefore have to content ourselves with, as a first approximation, the mean field analyses. This was done [6] for the weak correlation limit $U < 2zt$ as well as for the strong correlation limit $U \gg t$. The latter has obvious connections with the RVB proposals [7]. As we shall see this gives already rise to an extremely rich phase diagram involving different anisotropic superconducting states, as well as a spin density wave state (SDW) in a weak correlation regime $U < 2zt$.

Depending on whether we consider singlet or triplet intersite pairing we have to define the two following gap functions

$$\Delta_s\left(\underline{k}\right) = \frac{1}{N}\sum_{\underline{q}} V^s_{\underline{k}\,\underline{q}} \left\langle c_{-\underline{q}\downarrow}\, c_{\underline{q}\uparrow}\right\rangle ,$$

$$\Delta^\sigma_t\left(\underline{k}\right) = \frac{1}{N}\sum_{\underline{q}} V^t_{\underline{k}\,\underline{q}} \left\langle c_{-\underline{q}\,\sigma}\, c_{\underline{q}\,\sigma}\right\rangle \qquad (3.1)$$

where

$$V^s_{\underline{k}\,\underline{q}} = -U - W_{\underline{k}-\underline{q}}\ , \quad V^t_{\underline{k}\,\underline{q}} = \frac{1}{2}\left(W_{\underline{k}+\underline{q}} - W_{\underline{k}-\underline{q}}\right), \qquad (3.2)$$

W_k denoting the Fourier transform of W_{ij}. These gap functions have to be compatible with the lattice symmetry imposing the following Ansatz which decomposes them into a linear combination of orthonormal contributions

$$\Delta_s\left(\underline{k}\right) = \Delta_0 + \Delta_\gamma\ \gamma_k + \Delta_\eta\ \eta_k$$

$$\Delta_t\left(\underline{k}\right) = \Delta^n_x \sin k_x + \Delta^n_y \sin k_y \qquad (3.3)$$

with

$$\gamma_k = 2\left(\cos k_x + \cos k_y\right) \text{ and } \eta_k = 2\left(\cos k_x - \cos k_y\right).$$

At $T = T_c$ one has separate gap equations for Δ_0, Δ_γ, Δ_η and $\Delta^p_{x,y}$. The numerical solutions for T_c determining the onset of pure pairings are plotted in figure 3 where besides the various superconducting transition temperatures for s wave pairing (Δ_0, Δ_γ) and d (Δ_η) wave pairing as well as $p\left(\Delta^p_{x,y}\right)$ wave pairing we also plot the transition temperature for SDW described by the order parameter

$$m_{\mathbf{Q}} = \frac{1}{2N}\sum_{\underline{k}\,\sigma} \sigma \left\langle c^+_{\underline{k}\sigma} c_{\underline{k}+\mathbf{Q},\sigma}\right\rangle \quad \text{for} \quad \mathbf{Q} = \left[\frac{\pi}{a}, \frac{\pi}{a}\right].$$

It is particularly interesting to observe that extended s wave pairing is predominant in the dilute limit $n \ll 1$,

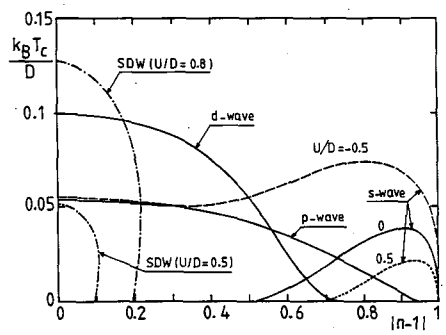

Fig. 3. – Phase diagram for the intersite pair system as a function of T and electron concentration n for the various anisotropic superconducting pairings and SDW for $|W| / D = 0.5$ and different values of U / D. $D = 4t$ is the half band width.

$(2 - n \ll 1)$ only and resembles much the behaviour of onsite pairs (Fig. 1) in the same regime. This is a first indication that in the dilute limit intersite pairs may behave as a superfluid of hardcore charged bosons. Upon increasing the density of electrons extended s-wave pairing first becomes unstable *versus* p-wave, then p-wave *versus* d-wave and finally d-wave pairing *versus* SDW. We notice also that for a large regime of parameters U, W and t the SDW state is only stable within a very narrow region around the half filled band case $n = 1$.

The quadratic lattice considered here has perfect Fermi surface nesting properties and the density of states displays the van Hove singularity if one considers nearest neighbour hopping only. If next nearest neighbour hopping is included the Fermi surface nesting is spoiled and the van Hove singularity moves away from $n = 1$. The resulting picture for $T_c(n)$ changes, the maximum of $T_c^{p,d}$ moving from $n = 1$ toward lower concentrations while that of s-wave pairing can shift to higher concentrations. The relative stability of different pairings can be essentially modified [6]. This may be significant as far as the pressure dependence of T_c is concerned; it can increase or decrease T_c depending on the concentration n.

Concerning the thermodynamic and electrodynamic properties of s wave superconductivity we expect a similar behaviour to that of onsite pairs, at least in the very dilute limit. It has been shown [15] that the gap equations for intersite pairing in the dilute limite reduce to the Schrödinger equation for a single pair which shows real bound states and for this reason we expect a Bose condensed state analogous to the one for onsite pairs in the equivalent limit.

4. Conclusions

We have shown on the basis of two representative cases – real space pairing with onsite and with intersite attractions – what sort of novel features can be expected for their superconductivity as compared to BCS. Our findings are independent of the particular mechanism giving rise to electron attraction. We find in particular that in the limit of small electron concentration the superconductivity is given by a Bose condensation for charged bosons with $T_c \sim n^{2/3}$ for d=3, λ-like specific heat anomaly, large upper critical fields and penetration depths and coherence length comparable to the lattice constant. The critical behaviour is that of a quantum $S = 1/2$ $X - Y$ model and the transition to the superconducting state of Kosterlitz-Thouless type is possible for $d = 2$. Many of these features have been observed in the recently discovered high T_c oxides (LaBaCuO and YBaCuO) and also in other oxides like $BaBi_{1-x}Pb_xO_3$ and $Ba_{0.6}K_{0.4}BiO_3$. This suggests that charged Boson superfluidity might be a unifying concept of superconductivity for the whole class of these oxides. If real-space pairing applies to these materials we believe that the primary pairing mechanism is related to the nature of cations which can exist in different valence states together with a high polarizability of certain anions. Such systems are expected to support large dynamical lattice deformations which were effectively seen [16]. A more general picture which might apply to this situation is one described by a mixture of Bosons (pairs of electrons) and fermions belonging to two different species of electrons and intracting with each other *via* a chargetransfer interaction [17]. The features of such a model are intermediate between a BCS and a real space pair superconductor.

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