A Model to Study Microscopic Mechanisms in High- T_c Superconductors

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1. Introduction

Superconductivity is a very curious phenomenon characterized by a phase transition at a critical temperature (T_c) in which the conducting phase is in equilibrium with the superconducting phase. The most important properties of the superconducting phase are: zero resistance, ideal diamagnetism (Meissner effect), magnetic flux quantization and persistent current in superconducting rings, cylinders or coils. On the other hand, many effects are found in superconducting constrictions as well as in junctions between two superconductors or in junctions between a superconductor and a conductor. These effects are known as “Josephson effects”: (1) It is possible to occur tunneling of Cooper pairs across a thin insulator between two superconductors and thus a superconducting current may be maintained across the junction; (2) when we apply an electric field gradient across a Josephson junction an electromagnetic wave may be produced, (3) when a beam of electromagnetic waves is incident over a Josephson junction a variable electric potential difference may be produced.

Due to all the effects mentioned above, superconducting devices may be projected for an enormous number of practical applications. Superconducting wires can be used for power transmission and in other applications when zero resistance is required. A possible application of magnetic levitation is the production of frictionless bearings that could be used to project electric generators and motors. Persistent currents can be used in superconducting magnets and in SMES (superconducting magnetic energy storage). Devices based on the Josephson effects are actually been used in very sensitive magnetometers and appropriate devices based on these effects may give rise to a new generation of faster computers. Superconducting magnets are been used in particle accelerators and may also be used to levitate trains. Many of these devices are successfully been used and new devices are been developed. However, the actual use of these superconducting devices is limited by the fact that they must be cooled to low temperatures to become superconducting. Currently, the highest T_c is approximately equal to 135 K at 1 atm (Schilling & Cantoni, 1993). The discovery of a room temperature superconductor should trigger a great technological revolution. A book with a discussion about room temperature superconductivity is available (Mourachkine, 2004). The knowledge of the microscopic mechanisms of oxide superconductors should be a theoretical guide in the researches to synthesize a room temperature superconductor. However, up to the present time, the microscopic mechanisms of high- T_c superconductivity are unclear. In the present chapter we study microscopic mechanisms in high- T_c superconductors.

According to the type of charge carriers, superconductors can be classified in two types: n-type superconductors, when the charge carriers are Cooper pairs of electrons and p-type superconductors, when the charge carriers are Cooper pairs of holes.

We know that BCS theory (Bardeen et al., 1957) explains the microscopic mechanisms of superconductivity in metals. These materials are clearly n-type superconductors. According to BCS theory, electrons in a metallic superconductor are paired by exchanging phonons.

Microscopic mechanisms in some types of non-metallic superconductors, like MgB_2 (Nagamatsu et al., 2001), probably may be explained by BCS theory. However, according to many researchers (De Jongh, 1988; Emin, 1991; Hirsch, 1991; Ranninger, 1994), BCS theory is not appropriate to be applied to explain the mechanisms of superconductivity in oxide superconductors. Nevertheless, other models relying on a BCS-like picture replace the phonons by another bosons, such as: plasmons, excitons and magnons, as the mediators causing the attractive interaction between a pair of electrons and many authors claim that superconductivity in the oxide superconductors can be explained by the conventional BCS theory or BCS-like theories (Canright & Vignale, 1989; Prelovsek, 1988; Tachiki & Takahashi, 1988; Takada, 1993). In this chapter we discuss this controversy. That is, we discuss the microscopic mechanisms to explain the condensation of the superconductor state of oxide superconductors. This discussion may be useful to study all types of oxide superconductors, that is, oxide superconductors containing copper, as well as oxide superconductors that do not contain copper. However, the main objective of this chapter is to discuss the role of double valence fluctuations in p-type oxide superconductors. In a previous work (Luiz, 2008) we have suggested a simple phenomenological model useful to calculate the optimal doping of p-type high- T_c oxide superconductors. In this chapter we study possible microscopic mechanisms in high- T_c superconductors in order to give theoretical support for that simple model.

2. Oxide superconductors

It is well known that there are metallic superconductors and non-metallic superconductors. Oxide superconductors are the most important non-metallic superconductors. An interesting review about oxide superconductors is found in the references (Cava, 2000). The history of oxide superconductors begins in 1933 with the synthesis of the superconductor NbO ; with $T_c = 1.5$ K (Sleight, 1995). In 1975 it was discovered the oxide superconductor $\text{BaPb}_{0.7}\text{Bi}_{0.3}\text{O}_3$ (Sleight et al., 1975) with $T_c = 13$ K. In 1986, the oxide superconductor $\text{Ba}_{0.15}\text{La}_{1.85}\text{CuO}_4$ with $T_c = 30$ K has been discovered (Bednorz & Müller, 1986). The expression “high- T_c superconductors” has been generally used in the literature to denote superconductors with critical temperatures higher than 30 K. After this famous discovery many cuprate high- T_c superconductors have been synthesized. The cuprate superconductor $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+x}$ (Hg-1223) has the highest critical temperature ($T_c = 135$ K) at 1 atm (Schilling & Cantoni, 1993). In 2008, a new type of high- T_c superconductor containing iron (without copper) has been discovered (Yang et al., 2008). In Table 1, we list in chronological order the most important discoveries of superconductors containing oxygen. In Table 1, T_c is expressed in Kelvin and x is a variable atomic fraction of the doping element.

The most relevant differences between the properties of oxide high- T_c superconductors and the properties of metallic superconductors can be summarized in the following points:

- a. All metallic superconductors are isotropic (the so-called “S-wave superconductivity”). All high- T_c oxide superconductors are characterized by a very large anisotropy

- manifesting itself in their layered structures with planes (a, b) perpendicular to the principal crystallographic axis (c-axis).
- In a metallic superconductor the coherence length is isotropic and is of the order of 10^{-4} cm. In high- T_c superconductors, the coherence length is anisotropic and of the order of angstroms. For example, in the system Bi-Sr-Ca-Cu-O, the coherence length is approximately equal to 1 angstrom (10^{-10} cm) along the c-axis and approximately equal to 40 angstroms in the transverse direction (Davydov, 1990).
 - In high- T_c superconductors, the dependence of T_c on the concentration of charge carriers has nonmonotonic character, that is, T_c does not rise monotonically with the rise of the carrier concentration. In a metallic superconductor, T_c rises monotonically with the rise of the carrier concentration.
 - In a metallic superconductor, the energy gap can be predicted by BCS theory. However, the energy gap of oxide superconductors seems to be anisotropic and probably cannot be predicted by BCS theory.

The isotopic effect, predicted by BCS theory, is a fundamental characteristic of a metallic superconductor. However, the isotopic effect is not clearly observed in oxide superconductors.

Superconductor	Year	T_c	Reference
(1) NbO	1933	1.5	Sleight, 1995
(2) K_xWO_3	1967	6.0	Remeika et al., 1967
(3) $LiTi_{2+x}O_4$	1973	1.2	Johnston et al., 1973
(4) $BaPb_{1-x}Bi_xO_3$	1975	13	Sleight et al., 1975
(5) $La_{2-x}Ba_xCuO_4$	1986	30	Bednorz & Müller, 1986
(6) $YBa_2Cu_3O_{7-x}$	1987	90	Wu et al., 1987
(7) $Ba_{1-x}K_xBiO_3$	1988	30	Cava et al., 1988
(8) $BiSrCaCu_2O_{6+x}$	1988	105	Maeda et al., 1988
(9) $Tl_2Ba_2Ca_2Cu_3O_{9+x}$	1988	110	Shimakawa et al., 1988
(10) $HgBa_2Ca_2Cu_3O_{8+x}$	1993	130	Schilling & Cantoni, 1993
(11) $NdFeAsO_{1-x}$	2008	54	Yang et al., 2008

Table 1. Superconductors containing oxygen in chronological order

3. Double charge fluctuations

In Table 2, we show the electron configurations and the stable oxidation states of the most relevant metals that are used in the synthesis of the oxide superconductors listed in Table 1. The stable oxidation states reported in Table 2 have been summarized according to tables described in a textbook (Lee, 1991). In Table 2, the symbol [Ar] means the electron configuration of Ar, the symbol [Xe] means the electron configuration of Xe and the symbol [Kr] means the electron configuration of Kr. In Table 2 unstable oxidation states are not described.

Using Table 2 and considering the oxide superconductors listed in Table 1, we can verify that: in the superconductor (1) Nb may have the oxidation states Nb(+III) and Nb(+V); in the bronze superconductor (2) W may have the oxidation states W(+IV) and W(+VI); in the superconductor (3) Ti may have the oxidation states Ti(+II) and Ti(+IV); in the

superconductor (4) Pb may have the oxidation states: Pb(+II) and Pb(+IV) and Bi may have the oxidation states Bi(+III) and Bi(+V); in the copper oxide superconductors (5), (6), (8), (9) and (10) Cu may have the oxidation states Cu(+I) and Cu(+III).

<i>Metal</i>	<i>Electron configurations</i>	<i>Oxidation states</i>
As	[Ar]3d ¹⁰ 4s ² 4p ³	+III, +V
Bi	[Xe]4f ¹⁴ 5d ¹⁰ 6s ² 6p ³	+III, +V
Cu	[Ar]3d ¹⁰ 4s ¹	+I, +II, +III
Fe	[Ar]3d ⁶ 4s ²	+II, +III, +IV, +V
Nb	[Kr]4d ³ 5s ²	+III, +V
Pb	[Xe]4f ¹⁴ 5d ¹⁰ 6s ² 6p ²	+II, +IV
Ti	[Ar]3d ² 4s ²	+II, +III, +IV
Tl	[Xe]4f ¹⁴ 5d ¹⁰ 6s ² 6p ¹	+I, +III
W	[Xe]4f ¹⁴ 5d ⁴ 6s ²	+IV, +V, +VI

Table 2. Electron configurations and oxidation states of some metals

Note also that in the superconductor (7) (without copper), Bi may have the oxidation states Bi(+III) and Bi(+V). In the superconductor (11), an example of the recent discovery of iron-based superconductors (Yang et al., 2008), we can verify that Fe may have the oxidation states Fe(+II) and Fe(+IV) and As may have the oxidation states As(+III) and As(+V).

Observe that in most high- T_c superconductors there are alkaline earth metals (such as Ca, Sr, and Ba). We know that the electron configuration of an alkaline earth metal is given by [noble gas] ns^2 , where n is the number of the row in the periodic table. Thus, an alkaline earth atom may lose two paired external electrons (ns^2). According to Table 1, among the high- T_c oxide superconductors, $HgBa_2Ca_2Cu_3O_{8+x}$ (Hg-1223) has the highest critical temperature ($T_c = 135$ K) at 1 atm (Schilling & Cantoni, 1993). According to the tables in the textbook (Lee, 1991), the electron configuration of Hg is given by: [Xe] 4f¹⁴ 5d¹⁰ 6s². Because all electrons are paired in a Hg atom, it is possible that an Hg atom may lose two paired electrons at the external level (6s²). Therefore, alkaline earth metals atoms (such as Ca, Sr, and Ba) as well as Hg atoms may lose two paired electrons at the external level.

According to a number of authors the probable existence of double charge fluctuations in oxide superconductors is very likely (Callaway et al., 1987; Foltin, 1988; Ganguly & Hegde, 1988; Varma, 1988). Spectroscopic experiments (Ganguly & Hegde, 1988), indicate that double charge fluctuations is a necessary, but not sufficient, criterion for superconductivity. We argue that these charge fluctuations should involve paired electrons hopping from ions (or atoms) in order to occupy empty levels. That is, our basic phenomenological hypothesis is that the electrons involved in the hopping mechanisms might be paired electrons coming from neighboring ions or neighboring atoms.

Possible microscopic mechanisms for double charge fluctuations are: (1) hopping mechanisms (Foltin, 1989; Wheatley et al., 1988), (2) tunneling mechanisms (Kamimura, 1987), and (3) bipolaronic mechanisms (Alexandrov, 1999).

The discovery of Fe-based high- T_c superconductors (Yang et al., 2008) has reopened the hypothesis of spin fluctuations for the microscopic mechanisms of high- T_c superconductivity. However, it is interesting to note that Fe may have the oxidation states Fe(+II) and Fe(+IV). Thus, the conjecture of double charge fluctuations cannot be ruled out

in the study of the microscopic mechanisms in all Fe-based high- T_c superconductors. It is worthwhile to study the competition between double charge fluctuations and spin fluctuations in order to identify which phenomenon is more appropriate to investigate the microscopic mechanisms in the condensation of the superconducting state of Fe-based materials.

4. Valence skip

What is valence skip? About fifteen elements in the periodic table skip certain valences in all components they form. For example, according to Table 2, the stable oxidation states of bismuth are Bi(+III) and Bi(+V). The oxidation state Bi(+IV) is not stable. If the state Bi(+IV) is formed, occurs immediately a disproportionation, according to the reaction: $2\text{Bi}(+IV) = \text{Bi}(+III) + \text{Bi}(+V)$. In the compound BaBiO_3 , the formal valence Bi(+IV) is understood as an equilibrium situation involving a mixture of equal amounts of the ions Bi(+III) and Bi(+V). Observing Table 2, other important examples of elements with valence skip are As, Pb and Tl. In (Varma, 1988) there is a discussion about the microscopic physics behind the phenomenon of valence skip.

Elements with valence skip, like Bi and Pb, are the most appropriate elements to study the hypothesis of double charge fluctuations. It has been stressed that all elements with valence skip may be used in the synthesis of superconductors (Varma, 1988).

5. D-wave superconductivity

Let us consider copper oxide high- T_c superconductors. We assume that the copper-oxygen planes are parallel to the plane x, y and that the z -axis is parallel to the crystallographic axis (c -axis).

In a weak crystal field, according to Hund's rule, the ion Cu(+III) is paramagnetic because the levels $3d(x^2 - y^2)$ and $3d(z^2)$ are half-filled. However, it has been shown that in a strong crystal field this ion becomes diamagnetic (McMurry & Fay, 1998). We know that the electron configuration of the ion Cu(+III) is $[\text{Ar}]3d^8$. Considering a strong crystal field, the ion Cu(+III) may give rise to a square planar complex. On the other hand, considering a strong crystal field, the spin-pairing energy P is smaller than the splitting energy Δ (McMurry & Fay, 1998). Thus, in this case, all electrons in a Cu(+III) ion should be paired and this ion should be diamagnetic. This hypothesis is consistent with a correlation between crystal field splitting and high transition temperatures (Zuotao, 1991).

If there are paired electrons in the nearest neighbors of the Cu(+III) ions, two neighboring paired electrons can be attracted by Coulomb interactions and eventually may occupy double empty energy levels. Because the orbital $3d(z^2)$ becomes filled, the electrons coming from $+z$ and $-z$ directions are strongly repelled. On the other side, electrons coming from the directions $+x, -x, +y, -y$ are not repelled and, eventually, may jump to occupy the empty levels. These electrons are obviously d -electrons and these jumps may give rise to a collective wave function of d -electrons. This hypothesis is consistent with the so-called assumption of d -wave superconductivity. The probable existence of d -wave superconductivity in oxide superconductors is supported by a great number of experiments (Leggett, 1994; Scalapino, 1995; Shen & Dessau, 1995; Tanaka, 1994). This picture leads to the conclusion that the microscopic mechanism of the condensation of the superconducting state should be a Bose-Einstein condensation. In the next section we discuss the possibility of a direct Bose-Einstein condensation in oxide superconductors.

6. Bose-Einstein Condensation (BEC)

An important question concerning the microscopic mechanisms of high- T_c superconductivity is: how the electrons are paired to form the Cooper pairs that are necessary for the condensation of the superconducting state? An answer to this question might be provided by the following hypothesis: the superconducting state arises from a Bose-Einstein condensation (BEC) of existing paired electrons that jump to occupy the double empty 3d levels mentioned in the previous section. Because these electrons were previously paired in atoms or in ions, it is not necessary to assume external interactions (with phonons or other bosons) to account for the pairing energy P of these paired electrons. Another question to analyze is: are these paired electrons in the spin-singlet or in the triplet state? By our hypothesis, these electrons were just paired in atoms or in ions; therefore, these existing pairs are in the spin-singlet state. This singlet state hypothesis is confirmed by Knight shift experiments (Scalapino, 1995).

Is BEC possible in oxide superconductors? According to (Chakraverty et al., 1998) BEC is impossible in oxide superconductors. However, we want to show that BEC in oxide superconductors cannot be ruled out.

Initially it is convenient to clarify some concepts regarding BEC. It is well known that a collection of particles (bosons) that follows the counting rule of Bose-Einstein statistics might at the proper temperature and density suddenly populate the collections ground state in observably large numbers (Silvera, 1997). The average de Broglie wavelength λ_{dB} which is a quantum measurement of delocalization of a particle, must satisfy this condition. We know that $\lambda_{dB} = h/p$, where h is Planck's constant and p is the momentum spread or momentum uncertainty of the wave packet. In the other extreme, for particles in the zero momentum eigenstate, the delocalization is infinite; i.e., the packet is spread over the entire volume V occupied by the system. It is generally accepted that BEC occurs when the interparticle separation is of the order of the delocalization λ_{dB} (Silvera, 1997).

The thermal de Broglie wavelength λ_{dB} is a measure of the thermodynamic uncertainty in the localization of a particle of mass M with the average thermal momentum. Thus, λ_{dB} is given by

$$\lambda_{dB} = h/[3MkT]^{1/2} \quad (1)$$

where k is Boltzmann's constant. Equation (1) shows that at a certain low temperature T or/and for a small mass M , λ_{dB} may be spread over great distances. In order to determine the critical temperature T_c at which the addition of more particles leads to BEC it is sufficient to calculate a certain critical density $n = N/V$, where N is the number of bosons. This calculation is performed using Bose-Einstein statistics; according to (Silvera, 1997) and considering $M = 2m^*$, where m^* is the effective mass of the electron, we obtain

$$T_c = 3.31h^2n^{2/3}/(4\pi^2kM) \quad (2)$$

The first application of BEC theory to explain ^4He superfluidity was realized in 1938 (London, 1938). In an important paper (Blatt, 1962), the BEC approach has been extended to give the same results predicted by BCS theory. Thus, it is reasonable to conclude that the conventional n-type superconductivity in metals (explained by BCS theory) is a special case that can also be considered as a phenomenon of BEC of Cooper pairs.

There are three possibilities of occurrence of BEC: (a) BEC involving just bosons, (b) BEC involving just fermions, and (c) BEC involving bosons and fermions simultaneously. In (a)

there is a direct BEC without the need of an interaction to bind the bosons. However, in the cases (b) and (c) BEC is possible only indirectly in two steps: in the first step it occurs the binding between pairs of fermions giving rise to bosons and, in the second step, BEC of these bosons may occur.

Because liquid ^4He is a system of bosons, the condensation of ^4He is a BEC of type (a). Superfluidity of ^3He (Lee, 1997) is an example of BEC of type (b). Because liquid ^3He is a system of fermions, in order to occur BEC, two particles must be binded to form a boson and, in the next step, a BEC of these bosons may occur. Another example of BEC of type (b) is the phenomenon of superconductivity in metals and alloys. On the other hand, BEC theory has been successfully applied in dilute atomic gases (Dalfovo et al., 1999), opening new applications involving the BEC concept.

We study now the possibility of occurrence of a Bose-Einstein condensation in an oxide material. If possible, this phenomenon should be a BEC of type (c) mentioned above, that is, the mechanism should involve bosons and fermions simultaneously. In order to verify if BEC is possible in oxide superconductors, it is sufficient to calculate the order of magnitude of the critical temperature T_c using Equation (2). According to Table 1 in the reference (De Jongh, 1988), in a p-type superconductor, the order of magnitude of the carrier density is $n = 10^{21}/\text{cm}^3$. Considering an effective mass $m^* = 12m$, where m is the rest mass of the electron, we obtain by Equation (2) the following approximated value: $T_c = 100$ K. This order of magnitude is reasonable because Equation (2) is based on an isotropic hypothesis. However, oxide superconductors are not isotropic. But the crude calculation based on Equation (2) is sufficient to show that BEC in oxide superconductors cannot be ruled out. A more appropriate formula to calculate T_c (supposing BEC) has been derived in (Alexandrov & Edwards, 2000).

7. Energy gap of High- T_c oxide superconductors

The energy gap is a controversial property of oxide superconductors. In order to study this fascinating subject it is instructive to clarify some concepts regarding the energy gap of superconductors. The excited states of a system may be decomposed in a superposition of elementary excitations, the so-called normal modes of excitations. The elementary excitations can be of two general classes (a) excitations of Bose-Einstein type, which can be created or destroyed individually, and (b) excitations of the Fermi-Dirac type, which can only be created or destroyed in pairs. In this case, the excitations are called quasiparticles and quasiholes, so that, in any state of the system, the number of quasiparticles is equal to the number of quasiholes.

The so-called "energy spectrum" is a function $E(k)$ where k is the modulus of the wave propagator vector $\mathbf{k}$. It is said that a given class of excitations has an energy gap E_g when the minimum value of $E(k)$ is not zero. In the case of Bose-Einstein excitations there two possible $E(k)$ curves: (a) an energy spectrum with an energy gap E_g that may represent an energy spectrum of plasma oscillations (plasmons) and (b) an energy spectrum without an energy gap that may represent an energy spectrum of lattice vibrations (phonons). The energy spectrum in the phenomenon of ^4He superfluidity is an example of the last case.

For Fermi-Dirac excitations in systems involving electrons, there are three possibilities: (1) In the case of a normal metal, an infinitesimal amount of energy is sufficient to excite a quasielectron and quasihole pair, in this case, the energy gap is equal to zero, (2) in a metallic superconductor, the minimum energy of excitation, always considering $k = k_F$, (that

is, on the Fermi surface), changes from zero to a certain value Δ (the energy gap parameter), and (3) in a semiconductor, the energy gap is the minimum energy necessary to excite a quasielectron and quasihole pair.

In the case of the Fermi excitations in metallic superconductors, the existence of an energy gap is always related to the binding energy of the fermions pairs. In this case, the energy gap is always temperature-dependent. At $T = 0$ K, the energy gap parameter Δ has a maximum value because all Cooper pairs are in the ground state. If the temperature is raised above absolute zero, pairs are broken up by thermal agitation and the energy gap begins to decrease. As the temperature rises, the number of quasiparticles increases and the energy gap continues to fall, until, finally, at $T = T_c$, the energy gap is equal to zero.

We discuss now the energy gap of oxide superconductors. By our phenomenological model, it is not necessary to assume mechanisms of pair binding involving virtual phonons. Thus, the above mentioned excitations should not exist. To break an existing pair it is necessary to give energy greater than P , where P is the pairing energy of the existing pair. Thermal energies, considering $T < T_c$, should not be pair breaking. Thus, as the temperature raises from $T = 0$ K until $T = T_c$, the above-mentioned excitations does not exist. Therefore, we conclude that the energy gap of oxide superconductors should be temperature-independent. This conclusion has also been obtained by another author, using a different approach (Alexandrov, 1998).

There are various experimental procedures to measure the energy gap: (a) specific heat measurements, (b) absorption of electromagnetic waves, (c) ultrasonic attenuation and (d) tunneling measurements using Josephson junctions. We discuss only the experimental procedures (b) and (d).

In metallic superconductors, the most direct energy gap measurement comes from tunneling measurements with Josephson junctions (SIS junctions). It is well known that a SIS junction is obtained with an insulator film between two superconductors. For metallic superconductors the width of the insulator film should be of the order of 10^4 angstroms. However, for high- T_c oxide superconductors, where the coherence length is of the order of the distance between two atoms, the order of magnitude of the width of the insulator film should be about one angstrom, and the production of this film should be a very difficult task. Therefore, tunneling measurements with SIS junctions are not very appropriate to measure the energy gap of high- T_c oxide superconductors. Thus, it seems that experimental methods such as optical spectroscopy and neutron scattering provide a better route for the investigation of the energy gap of high- T_c oxide superconductors.

We know that when a photon is absorbed by a metallic superconductor, we have:

$$h\nu = E_g + \hbar\omega \quad (3)$$

where h is Planck's constant, ν is the frequency of the photon and ω is the frequency of the phonon. However, according to recent experimental results of photon absorption (Carbotte et al., 1999; Munzar et al., 1999), it seems that for oxide superconductors, Equation (3) does not hold and, probably, the photon energy is used to break two paired electrons, according to the relation: $h\nu = 2E_g$.

By our phenomenological model, superconductivity is due to double charge fluctuations involving d-electrons and occurs in the a , b planes. Thus, we conclude that the energy gap of oxide superconductors should depend on the direction of the wave vector $\mathbf{k}$, and there are many experimental evidences supporting the hypothesis of an anisotropic energy gap (Beasley, 1991; Sun et al., 1994; Maitra & Taraphder, 1999).

8. The pseudogap of High- T_c oxide superconductors

What is the so-called “pseudogap”? In the previous section we have discussed the question of the energy gap of high- T_c oxide superconductors. We have emphasized that spectroscopic methods are very appropriate to investigate the energy gap of high- T_c oxide superconductors. Using spectroscopic methods and other techniques it has been verified the opening of a gap in the electronic spectrum above the critical temperature of high- T_c oxide superconductors (Timusk & Statt, 1999). This energy gap has been denoted by “pseudogap”. There is an interesting discussion in the literature about the origin of the pseudogap (Kugler et al., 2001). The existence of the pseudogap clearly reflects the possible presence of Cooper pairs above T_c and some experiments (Kugler et al., 2001), show that the order of magnitude of the pseudogap above T_c is approximately equal to the order of magnitude of the energy gap below T_c . If these experiments are confirmed, they should clearly indicate the presence of the same Cooper pairs above and below T_c . Thus, the hypothesis of Bose-Einstein condensation of Cooper pairs may be used to explain these experimental results (see Section 6). On the other hand, according to our phenomenological model, based on double charge fluctuations, these Cooper pairs should be paired electrons hopping from site to site to occupy double empty levels existing in neighboring ions or neighboring atoms (see Sections 3 and 5).

9. Oxygen doping of oxide superconductors

The most relevant doping procedures used for the synthesis of cuprate superconductors have been described in a review article (Rao et al., 1993). The first high- T_c oxide superconductor was the copper oxide $\text{Ba}_x\text{La}_{2-x}\text{CuO}_4$ (Bednorz & Muller, 1986). This superconductor is synthesized by doping the parent material La_2CuO_4 with Ba atoms. Soon after this discovery, it was realized (Schirber et al., 1988) that doping the parent material La_2CuO_4 with oxygen, without the introduction of any Ba atomic fraction x , it is also possible to synthesize the superconductor $\text{La}_2\text{CuO}_{4+x}$. Thus, in this case, we conclude that the introduction of oxygen is responsible for the doping mechanism of the parent material La_2CuO_4 (Schirber et al., 1988).

Oxide materials may become superconductors when a parent material is doped by the traditional doping mechanism with cation (or anion) substitution or by a doping mechanism based on oxygen non - stoichiometry (De Jongh, 1988). If a certain oxide contains a metal with mixed oxidation numbers, by increasing (or decreasing) the oxygen content, the metal may be oxidized (or reduced) in order to maintain charge neutrality. Therefore, the synthesis of p-type superconductors may be obtained by doping the parent materials with an excess of oxygen atoms and the synthesis of n-type superconductors may be obtained by doping the parent materials with a deficiency of oxygen atoms. One famous example of oxygen doping is provided by the family of p-type oxide superconductors Y-Ba-Cu-O. It is well known that $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, considering compositions x between $x = 0.5$ and $x = 0.9$ are superconductors with a maximum T_c with oxygen doping at a composition corresponding to $x = 0.9$. An important example of n-type superconductor is provided by the recent discovery of the superconductor GdFeAsO_{1-x} , a high- T_c superconductor with oxygen-deficiency; it has been shown that oxygen doping is a good and reliable procedure for the synthesis of a new family of iron-based high- T_c superconductors (Yang et al., 2008).

In the present chapter we shall study only oxygen doping of p-type oxide superconductors. It is well known that for p-type superconductors the optimal oxygen doping of high- T_c

oxide superconductors corresponds to a certain critical hole content. An under-doped superconductor is synthesized when the hole content is less than this critical value and an over-doped superconductor is synthesized when the hole content is greater than this critical value. The prediction of the optimal doping is an unresolved issue. In the next section we propose a simple model to estimate the optimal doping of p-type oxide superconductors.

10. Optimal oxygen doping of oxide superconductors

Our basic hypothesis is that the existence of double charge fluctuations involving paired electrons may be a key to study the microscopic mechanisms in oxide superconductors. The essential concept in this hypothesis is that the hopping mechanism involves two paired electrons, instead of the hopping of a single electron. Our hypothesis may be easily applied in the oxide superconductors containing Bi (without Cu) because, in this case, it is well known that Bi (III) and Bi (V) are the only stable oxidation states for the Bi ions. Thus, double charge fluctuations may occur between the ions Bi (III) and Bi (V).

To apply our hypothesis to a copper oxide superconductor (without Bi) it should be necessary to suppose the existence of Cu (I) because we are assuming double charge fluctuations between the states Cu(+I) and Cu(+III). In p-type Cu oxide superconductors, the existence of the oxidation state Cu(+III) is obvious by the consideration of charge neutrality. Thus, from an experimental point of view, it is very important to verify if the oxidation state Cu(+I) is present in the high- T_c Cu oxide superconductors. The probable existence of the states Cu(+I) and Cu(+III) has been verified in the works (Karppinen et al., 1993; Sarma & Rao, 1988).

It is generally believed that the microscopic mechanisms in a cuprate superconductor depends only on the ions Cu(+II) and Cu(+III). Let us suppose that the hopping mechanism involves just a single electron between Cu(+II) and Cu(+III); if this single charge fluctuation would be responsible for superconductivity, we should conclude that the enhancement of Cu(+III) ions should produce a continuous enhancement of the critical temperature T_c . However, it is well known that T_c decreases when the hole concentration is higher than a certain concentration (Zhang & Sato, 1993). This important property is the nonmonotonic dependence of T_c on the carrier concentration, a high- T_c characteristic feature mentioned in item (c) of Section 2. Thus, by this reasoning and considering the experimental results (Karppinen et al., 1993; Sarma & Rao, 1988), we can accept the presence of the mixed oxidation states Cu(+I), Cu(+II) and Cu(+III) in the copper oxide superconductors. On the other hand, this conjecture is well supported if we consider the copper disproportionation reaction (Raveau et al, 1988): $2\text{Cu}(+II) = \text{Cu}(+I) + \text{Cu}(+III)$.

What should be the optimal chemical doping of Cu oxide superconductors in order to obtain the maximum value of T_c ? Initially we suppose an equal probability for the distribution of the copper ions states Cu (I), Cu (II) and Cu (III); thus, the initial concentrations of these ions should be $(1/3)\text{Cu}(+I)$, $(1/3)\text{Cu}(+II)$ and $(1/3)\text{Cu}(+III)$. However, we may suppose that by oxidation reactions, $(1/3)\text{Cu}(+II)$ ions may be completely converted to $(1/3)\text{Cu}(+III)$ ions. In this case, the maximum concentration of the Cu(+III) ions should be: $(1/3) + (1/3) = (2/3)$. Thus, the optimal doping should correspond to the following maximum relative concentrations: $(1/3)\text{Cu}(+I)$ ions and $(2/3)\text{Cu}(+III)$ ions. That is, the optimal doping, should be obtained supposing the following ratio: $[(\text{Cu}(+III) \text{ ions})/(\text{Cu}(+I) \text{ ions})] = 2$.

We apply this hypothesis to estimate the optimal doping of the famous cuprate superconductor $\text{YBa}_2\text{Cu}_3\text{O}_x$, where x is a number to be calculated. Using the relative values:

(1/3) for Cu(+I) ions and (2/3) for Cu(+III) ions, we may write the formula unit: $YBa_2Cu_1(+I)Cu_2(+III)O_x$. Considering the oxidation states Y(+III), Ba(+II) and O(-II) and using the charge neutrality condition, we get:

$$3 + (2 \times 2) + (1 \times 1) + (2 \times 3) - 2x = 0 \quad (4)$$

From Equation (4) we obtain:

$$x = 7.0 \quad (5)$$

The result (5) is in good agreement with the result ($x = 6.9$) reported in (Kokkallaris et al., 1999).

In the next section, using the above method, we estimate the oxygen content for optimal doping of the most important p-type copper oxide superconductors.

11. Results

Using the simple model described in the previous section, we estimate the necessary oxygen content to obtain the optimal doping of the most relevant p-type cuprate superconductors. It is important to note that the experimental determination of the oxygen content is a very difficult task. In Table 3 we have selected a number of works containing this experimental information.

<i>Superconductor</i>	<i>Predicted</i>	<i>Measured</i>	<i>REFERENCE</i>
$Ba_{0.15}La_{1.85}CuO_x$	$x = 4.1$	$x = 4.0$	Bednorz & Müller, 1986
La_2CuO_x	$x = 4.2$	$x = 4.1$	Schirber et al., 1988
$YBa_2Cu_3O_x$	$x = 7.0$	$x = 6.9$	Kokkallaris et al., 1999
$YBa_2Cu_4O_x$	$x = 8.2$	$x = 8.0$	Rao et al., 1993
Sr_2CuO_x	$x = 3.17$	$x = 3.16$	Hiroi et al., 1993
$Bi_2Sr_2CaCu_2O_x$	$x = 8.3$	$x = 8.3$	Babaei et al., 2003
$Tl_2Ba_2CuO_x$	$x = 6.2$	$x = 6.0$	Rao et al., 1993
$TlCa_2Ba_2Cu_3O_x$	$x = 9.0$	$x = 9.0$	Martin et al., 1990
$HgBa_2Ca_2Cu_3O_x$	$x = 8.5$	$x = 8.4$	Hung et al., 1997

Table 3. Comparison between the values of oxygen content predicted by our simple model and the experimental values reported in the literature

According to our model, we have used for the copper ions the following relative values: (1/3) for Cu(+I) ions and (2/3) for Cu(+III) ions. For the other elements in Table 3, we have considered the following stable oxidation states: La(+III), Y(+III), Ba(+II), Sr(+II), Bi(+III), Ca(+II), Tl(+III), Hg(+II) and O(-II). We verify that the results predicted by the simple model proposed here are in good agreement with the experimental results listed in Table 3.

In Table 3 we have considered only copper oxide superconductors. Our model may be extended to estimate the optimal doping of p-type oxide superconductors that do not contain Cu. For example, consider the material $Ba_{0.6}K_{0.4}BiO_x$, a famous superconductor without copper, with T_c approximately equal to 30 K (Cava et al., 1988). We shall suppose for the Bi ions the same proportionality assumed in the model just suggested for the cuprate superconductors, that is, for optimal doping, we assume that the relative concentrations

should be given by: $(1/3)\text{Bi}(+III)$ and $(2/3)\text{Bi}(+V)$. Considering the oxidation states $\text{Ba}(+II)$ and $\text{K}(+I)$ and using the charge neutrality condition, we have:

$$1.2 + 0.4 + (1/3)(3) + (2/3)(5) - 2x = 0 \quad (6)$$

From Equation (6) we obtain the result:

$$x = 2.95 \quad (7)$$

The result (7) is in good agreement with the value ($x = 3$) reported in the reference (Cava et al., 1988).

12. Discussion

We believe that the simple model proposed in this paper in the case of p-type oxide superconductors could also be extended to estimate the optimal doping of n-type oxide superconductors. However, in the case of n-type oxide superconductors, the reaction produced by oxygen doping is a reduction reaction instead of an oxidation reaction that occurs in p-type oxide superconductors. Since we have not found in the literature any experimental determination of the oxygen content in the case of n-type oxide superconductors we shall not discuss this issue here. This question will be addressed in a future work.

We have proposed a simple model to estimate the relative concentrations of the ions involved to estimate the oxygen content for optimal doping of p-type oxide superconductors. The predictions based on this model are in good agreement with experimental results reported in the literature (Table 3). However, we emphasize that this simple model is not a theoretical model, it is a phenomenological model. In Sections 5, 6, 7 and 8 we have discussed some questions in order to give theoretical support for this model.

13. Concluding remarks

In this chapter we have studied the most relevant questions about the microscopic mechanisms of superconductivity in oxide materials. Parts of our arguments may be found in the list of references in the next section. However, we believe that our ideas have been expressed in a clear form for the questions at hand.

Our conjectures can be used to explain some remarkable properties of high- T_c superconductors mentioned in Section 2: (a) the anisotropy is explained considering that the electrons involved in the hopping mechanisms are 3d-electrons (see Section 5); (b) the order of magnitude of the coherence length (the mean distance between two electron pairs) is in accordance with the order of magnitude of the distance between the electron clouds of two neighboring ions (see Section 2); (c) the nonmonotonic dependence of T_c on the carrier concentration is explained by the hypothesis of double charge fluctuations and optimal doping (Sections 3 and 11).

The theory of bipolaronic superconductivity (Alexandrov & Edwards, 2000) is similar to our phenomenological model. In the theory of bipolaronic superconductivity, bipolarons are formed supposing a mechanism to bind two polarons. However, by our hypothesis, it is not necessary to suppose the formation of bipolarons by the binding of two polarons. We have assumed that the preformed pairs are just pairs of electrons existing in the electronic configurations of the ions or atoms involved in double charge fluctuations. These pairs

should be, for example, lone pairs in atoms or ions or pairs of electrons in the electronic configurations obtained when Hund's rule is applied.

The simple model described here is not a theoretical model and cannot be used to account quantitatively for the microscopic mechanisms responsible for superconductivity in oxide materials. However, we believe that our assumptions are helpful to the investigations of the microscopic mechanisms in oxide superconductors. We expect that this simple model will also be useful to encourage further experimental and theoretical researches in superconducting materials. It is worthwhile to study the details of the role of double charge fluctuations in the microscopic mechanisms responsible for superconductivity in oxide materials.

Finally, we suggest some future researches. It is worthwhile to make experiments to verify if this model is correct. Supposing that this simple model works, it would be possible to calculate stoichiometric compositions in order to obtain the optimal doping in the researches to synthesize new oxide superconductors. It is well known that the most important method in semiconductor technology is obtained by ion implantation techniques. Similarly, we believe that ion implantation techniques probably will be important in superconductor technology as well. Thus, we hope that future researches based on ion implantation techniques could open a new route in the synthesis of high- T_c superconductors. These future researches, using ion implantation, should take advantage of the possibility of double charge hopping mechanisms, instead of single charge hopping mechanisms existing in the case of ion implantation in the semiconductor technology.

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