MAIN PROPERTIES OF THE NEW HIGH T_C SUPERCONDUCTORS RELATED TO THERMODYNAMIC FLUCTUATIONS AND TO THE MAGNETISM OF COPPER.

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ABSTRACT - After a short analysis of classical superconductors, we discuss the atomic and electronic structure of the new superconductors. A strong atomic anisotropy with two-dimensional character can lead to thermodynamic fluctuations of the superconducting order parameter which do not favour the T_c value nor the superconducting properties. For the classical low-T_c superconductors there was always a strong decrease of T_c due to magnetic impurities; this occurs much less for these new high-T_c superconductors with Gd, Fe, Ni, impurities. The existence of antiferromagnetic (AF) Copper in the fundamental superconducting Copper-oxygen planes seems to favour this new type of superconductivity. The substitution of Cu^{++} by non-magnetic Zn^{++} lowers T_c very significantly. For that reason one can consider that the two dimensional (2D) AF of Cu^{++} can be at the origin of these new superconductors that have a much higher T_c. We will present some theoretical models on the correlation between this magnetism and the superconductivity. A fundamental result is that T_c disappears when the 2D AF disappears, even when the electrical conductivity becomes much larger.

I - INTRODUCTION

1 - The classical superconductors with $T_c \leq 23$ K.

The origin of this superconductivity was well explained by the BCS theory [1] that described the formation of conducting Cooper pairs due to an attractive interaction induced by the electron-phonon interaction with

$$T_c = 1.14\ \theta_D \exp(-\frac{1}{g})$$

$$g = \lambda - \mu^* \qquad (1)$$

(θ_D is the Debye temperature, λ is the attractive interaction parameter while μ^* corresponds to a repulsive Coulomb interaction between conduction electrons; of course the occurrence of superconductivity is related to $\lambda > \mu^*$).

Three problems of these classical superconductors may concern also the new superconductors:

- T_c goes always to zero for thin films i.e. for two dimensional (2D) systems, T_c is suppressed by thermodynamic fluctuations.
- For bulk materials T_c is strongly decreased by very low concentrations of magnetic impurities, a fact related to the destruction of Cooper pairs when a magnetic exchange interaction exists between the impurity spins and the two spins of the Cooper pairs of opposite

signs. The decrease in T_c can be well explained by the Ginzburg-Landau model i.e.:

$$\Delta F_S + F_{MS} = -\alpha\Psi^2 + \frac{\beta}{2}\Psi^4 + \gamma\Psi^2\langle\delta M^2\rangle. \quad (2)$$

Here $\Delta F_S = F_S - F_N$ is the free energy difference between the superconducting and normal states, Ψ^2 is the superconducting order parameter while $\alpha = \alpha_0(T_c - T)$.
Furthermore F_{MS} is related to the exchange interaction J between the Cooper pair spins and the localized impurity spins with $\gamma \equiv J^2$ while $\langle\delta M^2\rangle = 2\chi k_B T$ is the thermodynamic fluctuation of the impurity magnetization (χ is the paramagnetic susceptibility). The minimization of $\Delta F_S + F_{MS}$ leads to

$$T_c(\gamma) = T_c - \frac{\gamma}{\alpha_0}\langle\delta M^2\rangle \quad (3)$$

Most of the T_c values are suppressed by less than 1% of magnetic impurities. For ferromagnetic transitions below $T_c(\gamma)$, this $T_c(\gamma)$ also disappears; this is not entirely if an antiferromagnetic (AF) transition occurs.

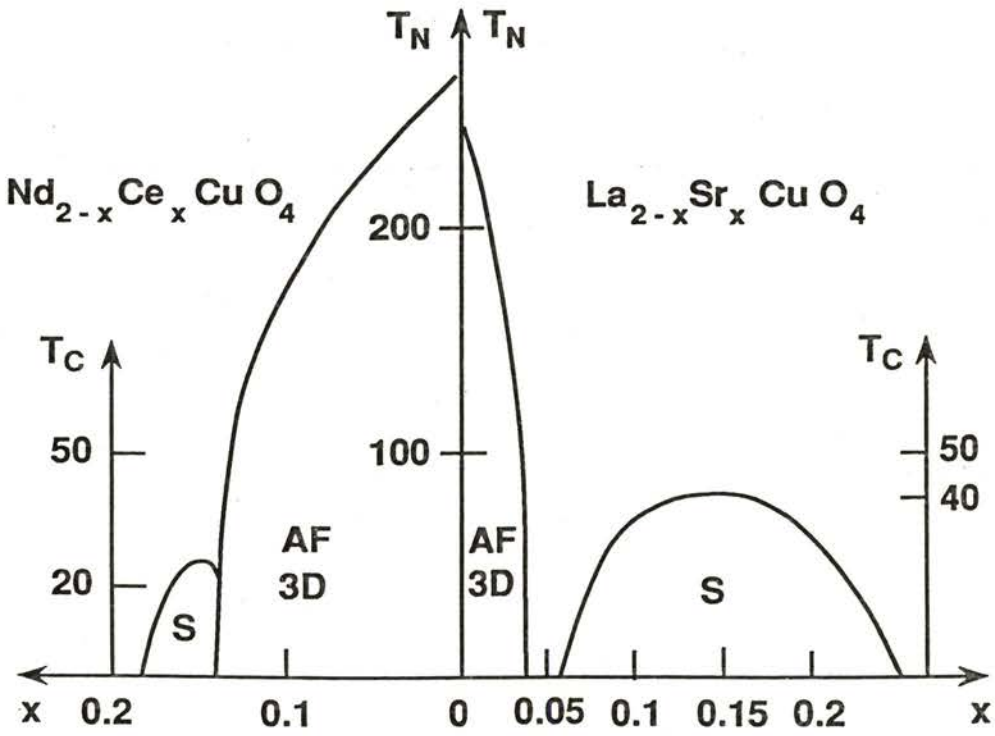


Fig 1: x dependence, related to the conduction carrier density, of the magnetic and superconducting transition temperatures T_N, T_c for $La_{2-x}Sr_xCuO_4$ and $Nd_{2-x}Ce_xCuO_4$.

- A further problem which can be important for the new superconductors, concerns the situation where two superconducting films of thickness d_S are separated but it contact with an intermediate normal metallic or even insulating film of thickness d_N: a weak superconducting order parameter can be induced in this intermediate film by the so called proximity or Josephson effects; it leads also to a decrease in T_C that will depend on the values of d_S and d_N.

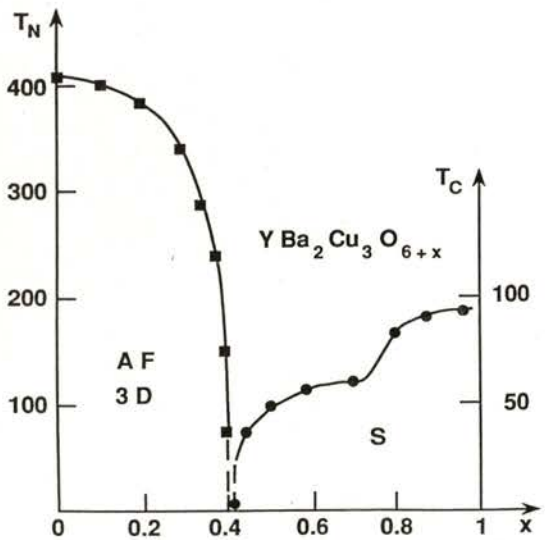


Fig 2. x dependence, related to the conduction carrier density, of the magnetic and superconducting transition temperatures of the 123 compounds

2 - The new high-T_C superconductors.

a) - The new copper oxide compounds. There are mainly four types of materials [2]:

-$La_{2-x}Sr_xCuO_4$ ($T_C \leq 39$ K) and $Nd_{2-x}Ce_xCuO_4$ ($T_C \leq 42$ K); for $x = 0$ we have insulators with AF transitions of copper. The three dimension (3D) AF disappears when $x \geq 0.03$ while superconductivity appears for $x \geq 0.07$ (Fig 1); $T_C(x)$ increases as a function of x up to $x \cong 0.15$ and then decreases to zero for $x \cong 0.25$.

- the 123 compounds $YBa_2Cu_3O_{6+x}$ ($T_C \leq 93$K) which are also insulators with (3D) AF for $x = 0$; $T_C(x)$ appears and increases beyond $x \geq 0.4$ up to $x = 0.6$ with a second increase up to $x \cong 0.9$ (Fig. 2) while the (3D) AF disappears near $x \cong 0.40$.

- the 2201, 2212 and 2223 compounds of Bi and Tl i.e. $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$ and $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$ with n = 1, 2, 3. T_C is maximum for n=3, and reaches 110 K for the Tl compounds. As a function of oxygen concentration, T_C goes through a maximum at 2n+4, a situation similar to the first compound (see Fig 1).

b) - Atomic structure effects: a very particular situation arises related to the fundamental CuO_2 planes. For $La_{2-x}Sr_xCuO_4$ and the 2201 compounds there is one CuO_2 plane per unit cell while for the 123 and 2212 compounds there are two and for the 2223 compounds there are three such planes per unit cell (Fig. 3). But inside the unit cell we can distinguish less metallic and less superconducting planes (Fig. 3). For that reason along the axis perpendicular to the a, b axis of the planes there can be proximity or Josephson effects for the less superconducting

planes but also more important thermodynamic fluctuations for the fundamental superconducting CuO_2 planes. There is also a large ratio between the two electrical resistivities ρ_c and ρ_{ab} [3] which clearly indicates the large anisotropy of the atomic and electronic structures.

c) - Electronic structure effects. Here we have two fundamental models.

- By considering that these compounds are ordinary metals [4] [5] (with $U < T$, where U corresponds to the Coulomb energy for two electrons on the sameatom while T corresponds to the conduction band width) one can relate the conduction band to the $Cu^{++} = 3d^9$ state or to antibonding and bonding bands due to the oxygen-copper hybridization. For such a half filled band with a 2D atomic structure, the Fermi surface corresponds exactly to a square which leads to a large Van Hove electronic density of states (Fig 4A). This modifies the BCS model in which N(e) was considered nearly constant for $\varepsilon = \varepsilon_F \pm k_B \theta_D$: very high values of T_c can be expected even if λ is small, because θ_D is replaced by a Fermi like temperature T_F.

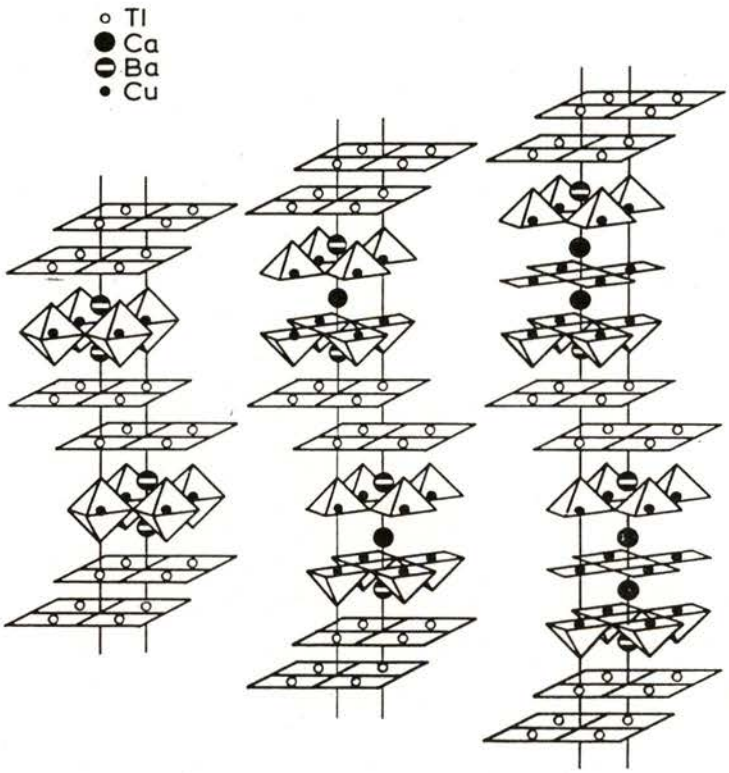


Fig 3. Atomic structure of $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$ with n=1,2,3

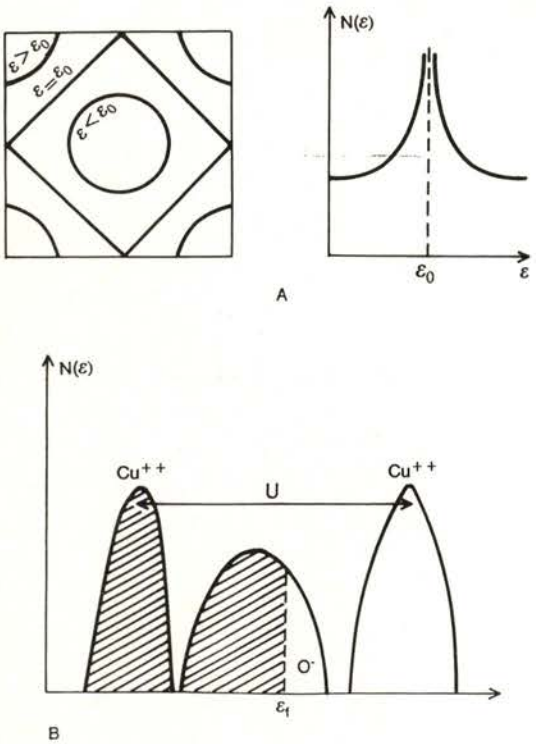


Fig 4. Fermi surface and density of state for a 2D metal for which ε_0 corresponds to a half filled band (A) ; if the Coulomb interaction U is larger than the band width, the half filled band of Cu^{++} is split into a filled and empty bandwidth, a possible intermediate oxygen band (B).

- For the Hubbard-Mott model [6] [7], for which U » T, the half filled band of Cu^{++} is split into two bands separated by the energy U: the low Hubbard band which is entirely filled by one electron/atom and the high empty Hubbard band. In the case of x = 0 i.e. $La_2CuO_4=La_2^{+++}Cu^{++}O_4^{--}$ we have an insulator with even an obligatory AF of the localized Cu^{++} spin electrons in the low Hubbard band (for which $J_{dd} \equiv T^2/U$). For $x \neq 0$ i.e.:
$La_{2-x}Sr_xCuO_4 = La_{2-x}^{+++} Sr_x^{++}Cu^{++} O_{4-x}^{--}O_x^-$,
the conductivity can be related to the oxygen band between the two Hubbard bands of Cu(Fig 4B); here the O^{--} corresponds to an insulating state (the filled oxygen band) while O^- corresponds to a conducting oxygen hole which is favoured by the positive Hall effect. The electronic structure is obviously different inside the CuO_2 planes and outside these planes in the c direction, where the conducting resistivity ρ_c is due essentially to the apical oxygen O^- outside the CuO_2 planes (the BaO planes for the 123 compounds). The destruction of the (3D) AF can be related also to the mobility of the O^- holes which can be perturbed by this AF if there is an $O^- - Cu^{++}$ exchange interaction.

II - MAIN RESULTS RELATED TO THERMODYNAMIC FLUCTUATIONS AND MAGNETISM.

1 - Thermodynamic fluctuations due to the anisotropic structure:
An important feature arises from the 2D character of the CuO_2 planes and the amplitude of their coupling. To evaluate the thermal fluctuations we use the G.L. model i.e. [8] [9]:

$$F_S - F_N = -\alpha\Psi^2 + \frac{\beta}{2}\Psi^4 + \frac{\hbar^2}{2m_c}|\nabla\Psi|^2 \quad (4)$$

(with $\alpha = \alpha_o (T_{co}-T)$ where T_{co} is the mean field transition temperature, m_c is the mass of a Cooper pair i.e. $m_c=2m^*$). Concerning the thermal fluctuations $\langle\delta\Psi^2\rangle = \Sigma_q \langle\delta\Psi_q^2\rangle$ one obtains:

$$\Psi^2 = \frac{\alpha}{\beta} - \langle\delta\Psi^2\rangle$$

$$T_c = T_{co} - \frac{\beta}{\alpha_o}\langle\delta\Psi^2\rangle = \frac{T_{co}}{1+x} \quad (5)$$

$$\langle\delta\Psi\rangle^2 \cong k_B T \frac{2m_c}{\hbar}\Sigma_q \frac{1}{q^2}$$

The term q^2 is only valid for an isotropic structure, while for anisotropic structures one has to replace it by $(q_{\|}^2 + (\xi_{\perp}^2/\xi_{\|}^2)\, q_{\perp}^2)$ where $q_{\|}$ and $q_{\perp}$ are wave vectors parallel and perpendicular to the CuO_2 planes while $\xi_{\|}$ and $\xi_{\perp}$ are the corresponding superconducting coherence lengths. For an isotropic 3D atomic structure one obtains $x \ll 1$ while for anisotropic structures one can obtain $x \gg 1$ with

$$x = \frac{2k_B T_{co}}{n}\,\frac{m^*(\mathrm{Log}\,2 + \Pi/2)}{\Pi\xi_{\perp}\hbar} \quad (6)$$

(n is the density per unit volume of the conducting electrons or holes). Large values of x are obtained for large values of T_{co} and m^* and for low values of n and $\xi_{\perp}$. An important result is that for $x \gg 1$:

$$k_B T_c = \left(\frac{\Phi_o}{2\Pi}\right)^2 \frac{\xi_{\perp}\,\lambda_L^{-2}}{2(\mathrm{Log}\,2 + \Pi/2)} \quad (7)$$

This result can can easily predict values of T_c between 10 and 100 K. The fact that the initial increase in T_c is always proportional to the inverse London penetration depth, namely λ_L^{-2}, has been observed by reference [10]: the theoretical value of $dT_c/d\,\lambda_L^{-2} = 0.5\ 10^{-7}$ is very close to the experimental value of $0.4\ 10^{-7}$. For that reason one is obliged to consider that values of $T_{co} \gg 100$ K are related to the true electronic structure of the CuO_2 planes and the attractive interactions of Cooper pairs. This very large value of T_{co} is decreased by thermodynamic fluctuations so that we expect maxima in $T_c \cong 100$ K. The value of T_c will be sensitive also to proximity effects because between the superconducting planes there are metallic but non-superconducting planes for which a weaker superconducting order parameter is induced; this necessarily decreases T_c such that $T_c(\text{proximity}) = T_c(1-Y)$ with Y proportional to d_S^{-2}, to $\xi_{\perp}^{-4}$ and to d_N^2: for that

reason Y can decrease if d_S increases due to the occurrence of more superconducting planes per unit cell; but Y can increase if $\xi_\perp$ decreases or if d_N increases (d_N and d_S are thickness of the non-superconducting and superconducting planes respectively). Nevertheless for n>3, T_c never increases even if d_S decreases because the electronic structure of all the CuO_2 planes are not exactly similar.

A fundamental experimental result for the thermodynamic fluctuations and the proximity effect concerns the application of magnetic fields $H < H_{c2}(T)$ This often leads to a large increase in the resistive transition (Fig 5) which is related not only to the vortex flux flow but also to an increase of the thermodynamic fluctuations and the proximity effect. In the case of the proximity effect it was shown [11] that $\xi_\perp(H) < \xi_\perp(0)$ so that $T_c(H) < T_c(0)$. But this field H has a non homogeneous repartition inside the sample so that the transition width D T_c (Fig 5) can be related to $T_c(0)$-$T_c(H)$ because of a non percolating situation. Experimentally evidence arises from magnetoresistance measurements which are the same above and below $T_c(0)$ [12], and the existence of a gap D (T) which goes to zero at $T_c(H)$ [13] and therefore. cannot be related to $H \cong H_{c2}$.

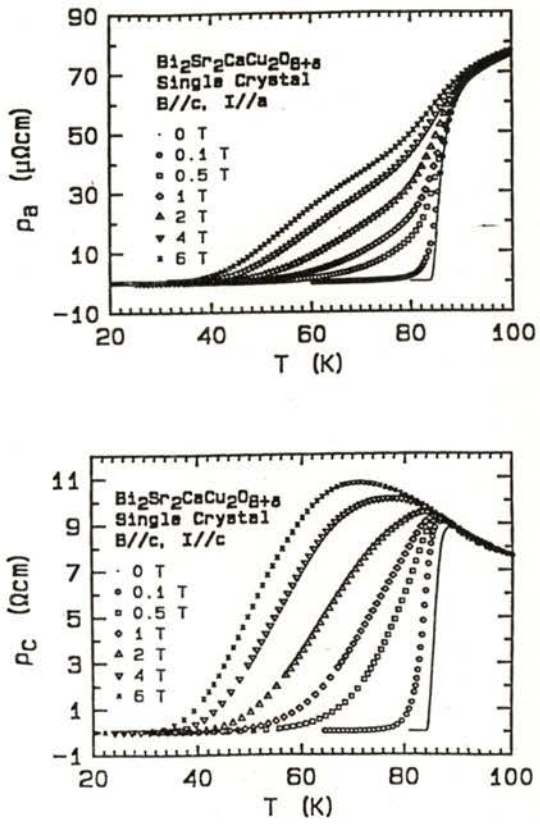


Fig 5. Effects of magnetic fields on the transition width of the electrical resistivity.

2 - AF Magnetism of Copper:

a) - (3D) AF: There is no coexistence of the antiferromagnetic transition $T_N(3D,x)$ and the superconducting transition $T_c(x)$ because $T_N(3D,x)$ decreases strongly with x while $T_c(x)$ only appears a bit later (Fig 1, 2). The value of $T_N(3D,x)$ is related to two exchange interactions of $3d^9\,Cu^{++}$ spins i.e. $J_{\parallel} = J_{dd}$ inside the

CuO_2 planes and $J_\perp \ll J_\parallel$ for Cu^{++} spins between different CuO_2 planes along the c axis. For $x \cong 0$ it leads to:

$$T_N(3D) \cong \frac{J_\parallel}{1 + \mathrm{Log}\frac{J_\parallel}{J_\perp}} \qquad (8)$$

while for $x \neq 0$, $T_N(3D,x)$ decreases due to the interaction with O^- holes whose density increases with x. Nevertheless for $Nd_{2-x}Ce_xCuO_4$ (Fig 1) and $YBa_2Cu_3O_{6+x}$(Fig 2) there is a common border between $T_N(3D,x)$ and $T_c(x)$, an effect which can be explained by the Ginzburg-Landau model [14] for which

$$\Delta F_S = -\alpha_1 \Psi^2 + \frac{\beta_1}{2}\Psi^4$$

$$\Delta F_{N(AF)} = -\alpha_2 M^2 + \frac{\beta_2}{2} M^4 \qquad (9)$$

$$F_{NS} = \gamma M^2 \Psi^2$$

with $\alpha_1 = \alpha_{10}(T_{cs}^0 - T)$ and $\alpha_2 = \alpha_{20}(T_{cs}^0 - T)$.
M is the AF order parameter, while $\gamma > 0$ is related to the opposition of the two order parameters, and Ψ is the same as in formula (2). In the case of $\gamma = 0$, T_c (x) and $T_N(3D,x)$ coexist (Fig 6). Then one can show that for $\gamma^2 < \beta_1\beta_2$, the coexistence continues to hold for $T_c(\gamma,x)$ and $T_N(\gamma,x)$ but their values decrease with γ. But for $\gamma^2 > \beta_1\beta_2$ there can be no coexistence but there can be a common border if the two condensation energies $-\alpha_1^2/2\beta_1$, $-\alpha_2^2/2\beta_2$ are not very different (Fig 6). For $La_{2-x}Sr_xCuO_4$(Fig 1), there is no common border which cannot be explained by γ and which is related to the interaction of Cu^{++} and O^- holes. This non coexistence where x represents the density of O^- holes, can be explained by the fact that when decreasing x one approaches the metal-insulator transition with a weak mobility of the O^- holes responsible of the superconductivity: this can favour the repulsive Coulomb interaction between O^- holes and $T_c(x)$ can disappear if $\mu^* > \lambda$. We should also note also that the attractive interaction λ can decrease if x decreases.

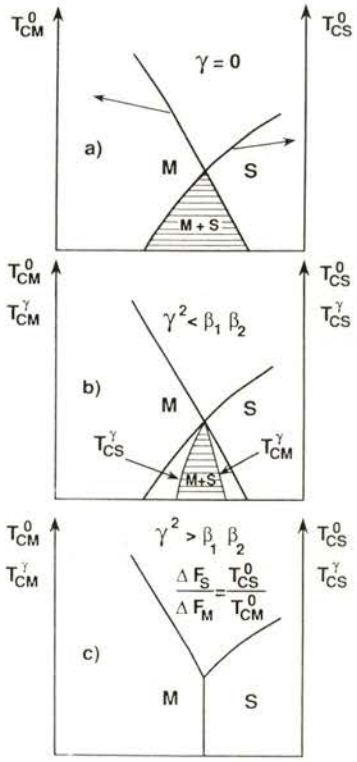
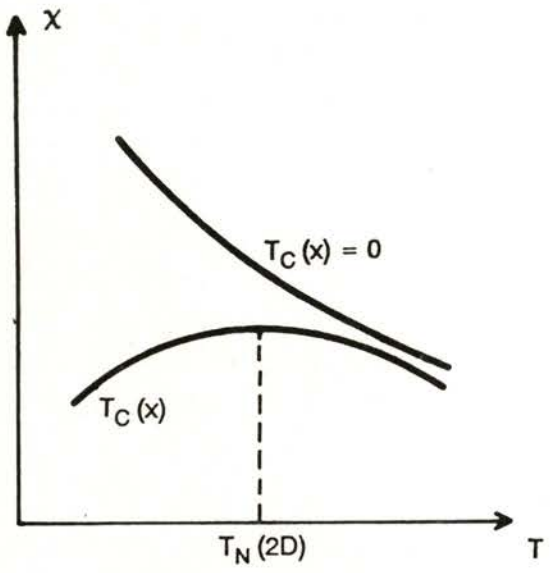


Fig 6. Variation as a function of x of the superconducting (S) and magnetic (M) transition for (a) $\gamma = 0$, for (b) $\gamma^2 < \beta_1\beta_2$ and for (c) $\gamma^2 > \beta_1\beta_2$ with $(\Delta F_M/\Delta F_S) \cong (T_{CM}^o/T_{CS}^o)$



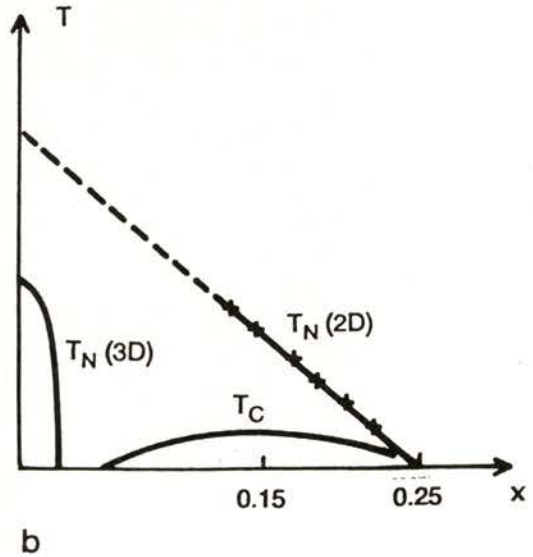


Fig 7. For $La_{2-x}Sr_xCuO_4$, the magnetic susceptibility above $T_c(x) \neq 0$ shows a 2D Néel temperature (a) whose values tends to zero like $T_c(x)$ for $x > 0.25$ (b).

b) - (2D) AF: of course even for $x \cong 0$ there appears above $T_N(3D)$ a two dimensional AF in the CuO_2 planes for which the susceptibility continues to increase beyond 1000 K because $T_N(2D) \cong J_{||}$. Neutron scattering studies show that the coherence length of this (2D) AF decreases with T above $T_N(3D)$ [15].

But when as a function of x, $T_N(3D,x)$ disappears there is now well documented experimental evidence by neutron scattering [16] [17] studies and susceptibility measurements above T_c [18] that there is coexistence between the superconductivity and this (2D) AF in the CuO_2 planes for which there are also thermodynamic fluctuations and a short AF coherence length which decreases with x [16] [19] down to 4 Å for the 123 compounds when $x \cong 1$ i.e. $T_c \cong 92K$. The susceptibility measurements also shows AF Néel transitions of (2D) character with a very large maximum as a function of T (Fig. 7a). This favours of course the Hubbard model. A second fundamental effect observed for $La_{2-x}Sr_xCuO_4$ and Bi 2212 compounds concerns the fact that T_c and $T_N(2D)$ disappear together when x increases and they become more metallic (Fig. 7b) [18]. It is easy to understand that the good mobility of O^- holes does not favour the (3D) and (2D) AF of Cu^{++} if there is an exchange interaction J_{pd} between Cu^{++} spins and O^- spins. This situation suggests strongly that the origin of these higher T_c superconductors can be related also to

an attractive interaction between O^- holes or, even more likely, to the exchange of magnetic singlet-triplet excitations of the nearly singlet pair liquid of Cu^{++} namely the $Cu^{++}\uparrow Cu^{++}\downarrow$ which can replace the phonon excitation for the classical superconductors. Because this singlet-triplet excitation favours the O^- mobility one can obtain two attractive interactions i.e. (Fig 8):

$$\lambda = N(\xi_F)\frac{t^2}{J_{dd}} \quad [6]$$

$$\lambda = N(\xi_F)\frac{J_{pd}^2}{J_{dd}} \quad [20]$$

with $T_{co} \cong J_{dd}\, e^{-1/\lambda}$. Of course λ can be very large leading to large T_{co} values because $J_{dd} \cong 1500$ K. There are also theoretical models which indicate that the resistivity above T_c is more related to magnetic fluctuating excitations than to phonons, but one cannot entirely exclude the attractive interaction due to phonons and to magnetic excitations. It is also easy to show that λ due to magnetic excitations can decrease if the AF coherence length increases, because it does not favour completely the singlet-triplet excitation of the Cu^{++} singlet pair liquid if there are AF correlations between neighbouring singlets. For that reason it is evident that λ decreases when one approaches the metal-insulator transition which favours the (3D) AF, while λ decreases also when one approaches the more metallic situation which destroys the (2D) AF. All this favours the importance of the (2D) AF magnetic excitation.

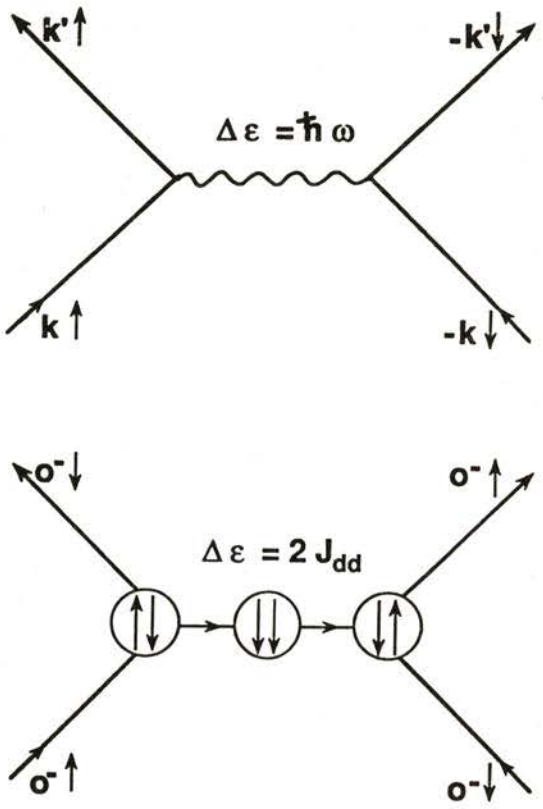


Fig 8. Attractive interaction between two electrons or two O^- holes through the exchange of a phonon energy $\Delta\varepsilon = \hbar\omega$ or a magnetic singlet-triplet energy $\Delta\varepsilon = 2\, J_{dd}$ between two Cu^{++} spins.

3 - Effects of magnetic and non magnetic impurities on T_c:

a) - Effects of Gd, Fe outside the CuO_2 planes :

- For $Y_{1-x}Gd_xBa_2Cu_3O_7$ there is no change of T_c even up to $x = 1$. This

means that there is no magnetic interaction between Gd and the conducting Cooper pairs in the CuO_2 planes. But for $Gd_{1+x}Ba_{2-x}Cu_3O_7$ the value of T_c decreases as a function of x (Fig 9) because Gd_x^{+++} is in the Ba^{++} plane implying that the decrease of T_c is more related to a change of the electronic structure due to the decrease of the O^- holes transformed into O^{--}.

- A similar effect is obtained by Fe^{+++} which goes mainly into the chain planes Cu(1)O outside the CuO_2 planes. For that reason there can be a strong decrease of T_c(Fig 9) for $YBa_2Cu_{3-y}Fe_yO_7$. There is experimental evidence that this effect is not related to magnetic interactions with the Cooper pairs but only to a change of the electronic structure. It has been shown that there is no change of T_c for $YBa_2Cu_{3-y}Fe_yO_{7+y}$ (Fig 9): Fe^{+++} which substitutes Cu^{++} can destroy O^- holes which are recreated by O_y. Furthermore [21] the T_c decrease due to Fe^{+++} can be compensated by the substituting Y^{+++} with Ca^{++}; in this case. the decrease of T_c is much smaller for $Y_{1-x}Ca_xBa_2Cu_{3-y}Fe_yO_7$ if $x \equiv y$ because Fe^{+++} decreases the O^- hole density while Ca^{++} increases it.

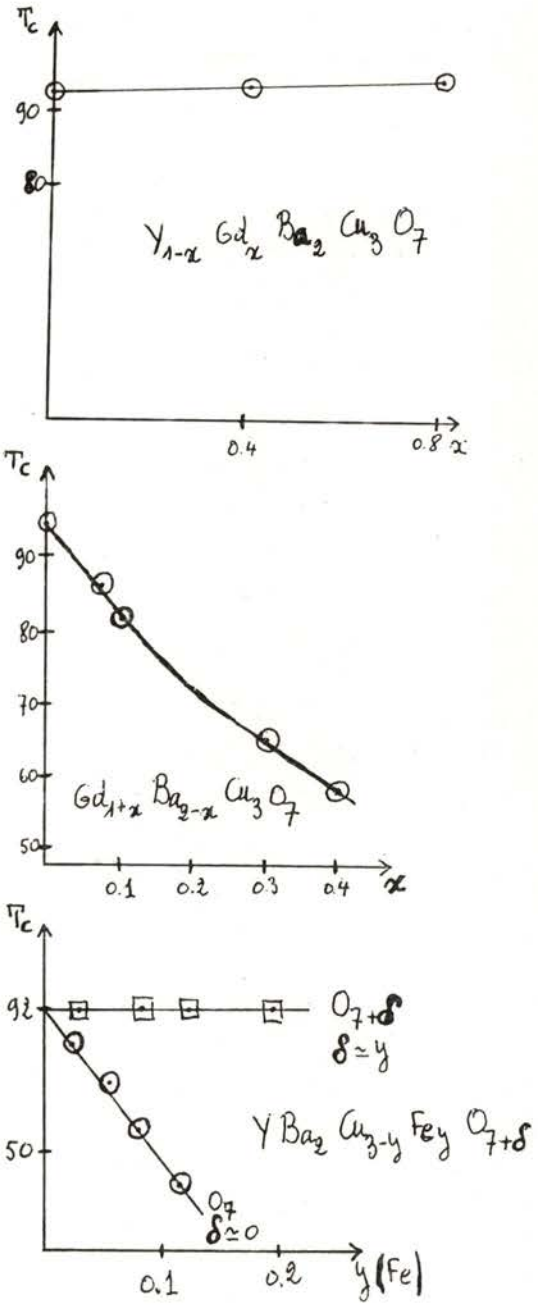


Fig 9. Variation of T_c for $Gd_{1+x}Ba_{2-x}Cu_3O_7$ and $YBa_2Cu_{3-y}Fe_yO_{7+\delta}$ as a function of x and y concentrations. But for $\delta \equiv y$, $T_c(y)$ does not change.

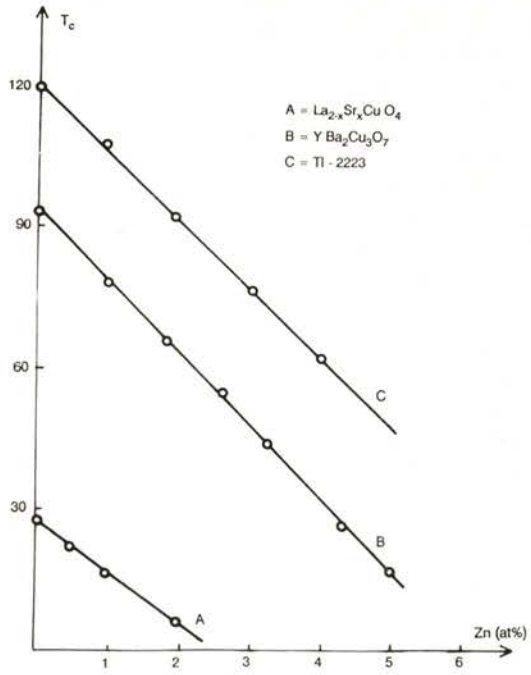


Fig 10. T_c dependence of several high T_c compounds by the substitution of Cu by Zn.

b) - Effects of Ni^{++} and Zn^{++} substituting Cu^{++} inside the CuO_2 planes:

A surprising result is that T_c decreases much more for non magnetic Zn; in fact $DT_c \cong 13$ K for 1% of Zn [22] (Fig 10). This effect can be explained because Zn changes the (3D) AF. T_N(3D) goes to zero rapidly for $YBa_2Cu_{3-y}Zn_yO_{6+x}$ (i.e at $x = 0.25$ for $y = 0.037$) which is very different from what is shown in Fig 2. But Zn can also destroy the (2D) AF if a copper singlet $\uparrow Cu^{++}Cu^{++}\downarrow$ is transformed into $\uparrow Cu^{++}Zn^{++}$. This means that there are three different reasons for the decreases of T_c: i) the attractive interaction related to a singlet-triplet excitation decreases because this singlet-triplet excitation disappears near Zn. ii) Zn^{++} creates also a local paramagnetic moment of Cu^{++} whose interaction with O^- hole pairs can destroy these pairs. iii) decreased mobility of the O^- holes due to the atomic disorder created by Zn: this can favour the repulsive Coulomb interaction between O^- holes i.e. μ^*. The Zn^{++} does not change the O^- hole density as shown by the Hall effect (only above 300 K) [23].

The effect of Ni^{++} on T_c may be less important because it does not suppress the magnetic singlet-triplet excitation and does not create isolated paramagnetic Cu^{++} moments. There can be nevertheless an atomic disorder with less mobility of the O^- holes and an increase of μ^*.

III - CONCLUSION

It has become increasingly evident that the magnetism of copper [but only the (2D) AF] has an enhancing effect on T_c which is apparently related to an attractive interaction due to singlet-triplet excitations. However electron-phonon attractive interactions could also

explain the negative effects due to thermal phonons near T_c as in the case of some classical superconductors. The (2D)-like atomic structure decreases the true very high mean field value T_{CO}, and favours (near T_c) the thermodynamic fluctuations of the superconducting order parameter and also the vortices. The effect of Zn has a fundamental effect on the decrease of T_c which can be related to i) the localization of the oxygen holes related to atomic disorder ii) the perturbation of the (2D) AF with occurrence of paramagnetic moments of Cu^{++}.

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