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Classification

Physics Abstracts

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Effects of electron irradiation on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superconductor

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Abstract. — We have irradiated $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals with electrons of energy E ranging from 0.1 to 2.4 MeV. From the measurements of the T_c reduction rate, $-\Delta\dot{T}_c = -d(\Delta T_c)/d(\Phi t)$ with Φt the irradiation fluence, we have clearly evidenced for the first time the influence of *both copper and oxygen defects* on the critical temperature. The analysis of the data, using calculated displacement cross-sections, allows us to extract the values of the displacement threshold energy for oxygen and copper atoms that we have evaluated as 10 and 15 eV respectively. We clearly show that irradiation effects have nothing to do with oxygen removal effects. However, the irradiation induced T_c decrease seems to be due to a pair breaking effect as suggested for Zn doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ samples. On this subject, we bring experimental evidence which shows that the « classical » Abrikosov-Gorkov depairing mechanism by magnetic impurities is not appropriate to explain either the irradiation effects or the Zn substitution ones.

1. Introduction.

Copper oxide superconductors have been found to be very sensitive to damage caused by radiation with highly energetic particles such as neutrons, ions or electrons [1-7]. In particular, the T_c reduction exceeds significantly that observed in conventional superconductors and even in A15 alloys under similar irradiation conditions. In the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) compound, it has been shown that the initial T_c reduction can be roughly correlated to the total number of atomic displacements by nuclear collisions irrespective of the irradiation type [8] — except for heavy energetic ions in which case electronic collisions dominate [9-11]. Transmission electron microscopy (TEM) has been used to study the defect structures induced by irradiation with electrons [12], neutrons [1, 13] and heavy ions [14]. In particular, these studies have allowed one to establish a clear correlation between the defects morphology and the pinning enhancement observed after neutrons [1] or high energy heavy ion irradiations [15, 16] — for a

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review of defect structures see [17] for instance. Nevertheless, as far as T_c is considered, the precise nature of the defects — and the underlying mechanism — responsible for the T_c degradation has not been elucidated yet.

In $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, it is well known that the oxygen content is the critical parameter governing T_c . As the oxygen atoms are the lightest of the structure and are loosely bound — at least along the chains — they can also be more easily displaced under irradiation, and it is tempting to attribute irradiation effects mainly to oxygen sublattice disordering. In order to investigate the validity of this assumption, we have irradiated at low temperature (20 K) YBCO single crystals by electrons of different energies, E . Indeed, electron irradiation remains one of the best controlled techniques to introduce point defects in a given matrix. By varying E , we can produce selective defects in the different sublattices, displacing first the lightest one, O, at low energy $E \leq 400$ keV, and further the heaviest ones, Cu and even Y and Ba, as the E value progressively increases from 0.1 to 2.4 MeV. We show unambiguously that T_c is sensitive to displacements of *both O and Cu atoms*. Moreover, we are able to determine with a good accuracy the displacement threshold energies for O and Cu atoms, i.e. the minimum energy which has to be transferred to an atom to produce a point defect, which is the necessary parameter to quantify irradiation disorder.

Obviously, to determine the mechanism which leads to such a drastic reduction of T_c by irradiation one needs a comprehensive understanding of the mechanism which induces superconductivity. Nevertheless different mechanisms can be put forward to try to understand this reduction of T_c . In these compounds it is well established that electronic properties and superconductivity are related to the hole content in the CuO_2 planes. Then one may envisage that disorder induces modifications of the charge transfer [18]. On the other hand, it has been suggested that the superconductivity damage under irradiation may result from a pair-breaking mechanism [19]. It is worth mentioning here that a connection between electron irradiation induced depression of T_c and formation of localized magnetic moments has already been evidenced by magnetic susceptibility measurements [7]. We show that our experimental results — namely Hall effect measurements on irradiated samples — are also better interpreted in the framework of a pair-breaking mechanism. On this subject we give strong experimental evidence to show that the « classical » Abrikosov-Gorkov depairing mechanism by magnetic impurities is not appropriate in this case.

2. Experimental.

2.1 SAMPLES. — All but the 100 keV irradiations were performed on single crystals of YBCO which were grown using the standard flux method. Samples were plates of about $1 \times 1 \times 0.02$ mm³. Electrical resistivity measurements were performed using the standard Van der Pauw method for all the samples [20]. Good electrical contacts (contact resistance of about 0.1 Ω) were made by sticking Pt wires with silver paste and then performing an annealing at 300 °C for one hour. The critical temperature was determined at the middle point of the resistivity transition. Before irradiation, T_c for all the samples was around 92 K, which shows the good quality of the crystals. Samples SI, SII and SIII were irradiated at different energies, while samples HI and HII were irradiated at 0.4 and 2.4 MeV respectively.

The irradiation at 100 keV was carried out on two good quality thin films, TI and TII, which were laser ablated onto a (100) SrTiO_3 substrate [21]. Typical dimensions of the samples were $2 \times 2 \times 0.001$ mm³. In this case, good contacts were performed by attaching Au wires using a In sonosolder. T_c before irradiation was about 89 K for both samples.

2.2 ELECTRON IRRADIATIONS. — All but the 100 keV irradiations were performed in the VINKAC low temperature facility on the Van de Graaff electron accelerator at the LSI

Palaiseau [22]. During irradiation, the samples are immersed in liquid H_2 (20 K) and the irradiation flux is limited to $2 \times 10^{14} \text{ e}^-/\text{cm}^2 \text{ s}$ in order to avoid heating. After a given fluence, the samples are maintained at 95 K for 10 min in an *in situ* furnace [23] and transition curves are recorded. During this stage, the temperature stability is estimated to be 50 mK.

The 100 keV irradiations were performed in a modified transmission electron microscope column well described elsewhere [24]. The samples are irradiated under vacuum and the irradiation flux is limited to $5 \times 10^{14} \text{ e}^-/\text{cm}^2 \text{ s}$ in order to maintain the temperature below 100 K. After given fluences, the transition curves are recorded using a Lake Shore 330 temperature controller, which ensures a temperature stability of about 20 mK.

It is worth mentioning here that for all the energies, the electron path was large compared to the sample thickness thus ensuring a homogeneous damage throughout the sample. Moreover, all irradiations were performed along the *c* axis.

2.3 ISOCHRONAL ANNEALING, HALL EFFECT MEASUREMENTS. — Annealing measurements were made on samples HI and HII after low temperature irradiation at 0.4 and 2.4 MeV respectively, using the *in situ* furnace mentioned above. The annealing steps were ten minutes long and 10 K apart between 100 and 360 K. The fluences of each irradiation were calculated in order to produce the same T_c reduction of about 15 K for each sample. The precise values of T_{ci} (T_c before irradiation), T_{cf} (T_c after irradiation), T_{cfa} (T_c after annealing at 360 K) and the fluences used are summarized in table I for both samples.

After the annealing at 360 K, the samples were moved into a cryostat provided with a superconducting coil in order to measure the Hall effect in the normal state at $B = 8 \text{ T}$. The Hall resistivity was determined by measuring the odd voltage contribution after exchanging the voltage and current probes at the diagonals of the samples. The longitudinal resistivity was obtained using the standard Van der Pauw technique. From these measurements, we are able to determine the longitudinal σ_{xx} and Hall σ_{xy} conductivity variations between 100 and 300 K.

Table I. — Values of T_{ci} , T_{cf} , T_{cfa} , E and Φt for samples HI and HII irradiated and annealed.

Sample	Electron energy (MeV)	Fluence ($10^{18} \text{ e}^-/\text{cm}^2$)	T_{ci} (K) (T_c before irradiation)	T_{cf} (K) (T_c after irradiation)	T_{cfa} (K) (T_c after annealing)
HI	0.42	148	92.8	76.3	86.7
HII	2.42	28	92.7	77.2	85.7

3. Results.

3.1 CRITICAL TEMPERATURE DECREASE RATE. — Samples SI, SII and SIII were irradiated at different energies E ranging from 0.1 to 2.4 MeV. For each value of E , we find a linear decrease of the critical temperature as a function of the electron fluence allowing us to determine the T_c decrease rate unambiguously as $-\Delta\dot{T}_c = -\Delta T_c/\Delta\Phi t$ where Φt is the irradiation fluence. In figure 1 we have plotted the variations of T_c with fluence for sample SIII. The sample was initially irradiated with electrons of 2.42 MeV and then successively irradiated with electrons of 0.55 (open circles), 2.42 (full circles), 0.55, 2.42, 1.2 (open triangles) and 2.42 MeV. It is important to note that after an irradiation at low (high) energy,

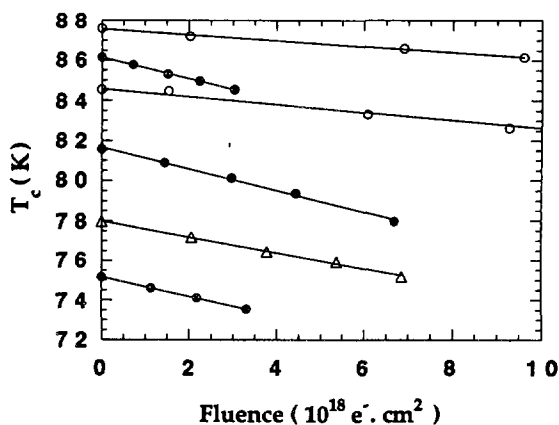


Fig. 1. — T_c dependence on electron fluence, Φt , for sample SIII initially irradiated with electrons of 2.42 MeV, and successively irradiated with electrons of 0.55 (open circles), 2.42 (full circles), 0.55, 2.42, 1.2 (open triangles) and 2.42 MeV.

the value of $\Delta \dot{T}_c$ at high (low) energy remains the same as in as-grown samples. This clearly shows that the physical parameter which determines the damage rate is not affected by irradiation within the energy and fluence range investigated, and allows us to be sure that $\Delta \dot{T}_c$ values at a given energy do not depend on previous irradiation damage.

Values of $-\Delta \dot{T}_c$ are plotted as a function of E in figure 2. The linear dependence between ΔT_c and Φt observed within the whole energy range investigated means that the created defects are uncorrelated. One can then write $-\Delta \dot{T}_c$ as a linear combination of the displacement cross-sections in the different sublattices :

$$-\Delta \dot{T}_c = \sum a_i \sigma'_d(E, E_d^i) \quad (1)$$

where σ'_d and E_d^i are respectively the cross-section and the displacement threshold energy for species i .

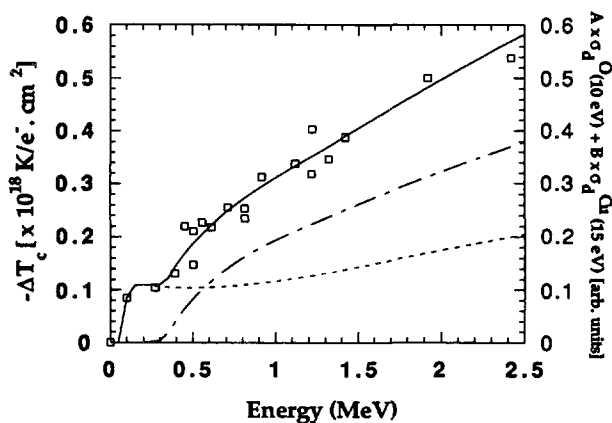


Fig. 2. — T_c decrease rate, $-\Delta \dot{T}_c$, as a function of the electrons energy E : open squares, experimental values ; solid line, fitting curve using calculated displacement cross-sections. The dotted line represents the contribution of the oxygen atoms ($E_d^O = 10$ eV) and the dashed line the contribution of the copper ones ($E_d^{Cu} = 15$ eV).

The best fit for the experimental results using equation (1) is also reported in figure 2. The cross-sections have been calculated using an analytical method developed by D. Lesueur [25]. It is worth mentioning that in this calculation the number of displaced j atoms n_{kj} by a primary k are taken into account. Here, the fitting parameters are only the two displacement threshold energies, E_d^O and E_d^{Cu} . Indeed, the low energy ($E < 0.4$ MeV) experimental values, can be fitted by considering the sole oxygen displacements : for $E_d^O = 10 \pm 1$ eV, we found a linear dependence between $-\Delta T_c$ and $\sigma_d^O(E, E_d^O)$ with a slope $A = (3.2 \pm 0.4) \times 10^3$ K. For higher electron energies, displacements in the copper sublattice must be considered resulting in the following expression :

$$-\Delta T_c = A\sigma_d^O(E, E_d^O) + B\sigma_d^{Cu}(E, E_d^{Cu}) \quad (2)$$

with $A = (3.2 \pm 0.4) \times 10^3$ K as aforementioned while $B = (8 \pm 0.7) \times 10^3$ K and $E_d^{Cu} = 15 \pm 1$ eV were determined in a similar way as A and E_d^O . In this expression σ_d^O and σ_d^{Cu} are displacement cross-sections normalized to the total number of atoms, which means that $\sigma_d^i \times \Phi t$ represents the displacement per atom — d.p.a. — for the i species, i.e. the ratio between the total number of i atoms displaced and the total number of atoms. Then, we see for instance that 1 % of d.p.a. for O produces a T_c reduction of 32 ± 4 K and 80 ± 7 K if we consider Cu atoms.

3.2 CRITICAL TEMPERATURE RECOVERY AFTER ANNEALING. — In order to study the differences between low and high energy electron irradiation induced defects, we have irradiated samples HI and HII respectively at 0.4 and 2.4 MeV and then studied the T_c recovery by isochronal annealing between 100 and 360 K. In figure 3a we have represented the T_c recovery rate $(T_c - T_{ci})/(T_{cf} - T_{ci})$ as a function of the annealing temperature. For HI we can clearly distinguish two recovery stages at 180 and 350 K. These two stages are also present in the high energy irradiated sample although they are less sharp than in the previous case. At 0.4 MeV the analysis presented above shows that almost only oxygen atoms are displaced, and therefore the two recovery stages at 180 and 350 K could be associated with two different recombination processes of oxygen defects. We can make a rough estimation of the amount of copper defect recombination in the 2.4 MeV irradiated sample by simply subtracting the contribution of oxygen defects assuming that formula (2) remains valid during annealing. Such recombination processes seem to occur within a broad temperature range between 200 and 300 K (cf. Fig. 3b).

3.3 HALL EFFECT MEASUREMENTS IN THE NORMAL STATE. — After irradiation and annealing, the two samples HI and HII present a T_c depression of about 6 K. The Hall constant, measured before and after irradiation and annealing, is not affected by this T_c depression. In fact, previous results obtained after ion or neutron irradiation [5, 26] show that the Hall coefficient is relatively insensitive to irradiation disorder even at much higher fluences. The same result is also observed when disorder is introduced by Zn substitution on copper sites [27]. It has been suggested by Anderson [28] that a better analysis of Hall data can be performed by studying the cotangent of the Hall angle Θ_H , given by $\cotg(\Theta_H) = \sigma_{xx}/\sigma_{xy}$ which is expected to increase linearly with the square of temperature :

$$\cotg(\Theta_H) = E \times T^2 + F .$$

These curves are plotted in figure 4, for sample HI before and after irradiation. We observe that this relation holds even after irradiation, the slope E being independent of the damage introduced and the coefficient F increasing as T_c decreases. Our goal is not to give

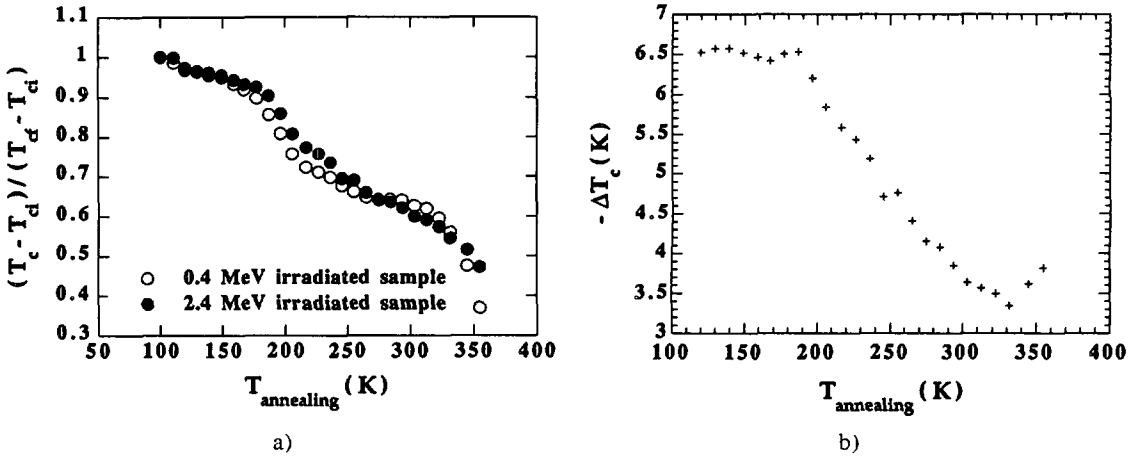


Fig. 3. — a) T_c recovery rate, $(T_c - T_{cl}) / (T_{cf} - T_{cl})$ (where T_{cl} and T_{cf} are respectively the T_c values before and after irradiation) as a function of the annealing temperature for samples HI (open circles) and HII (full circles); b) Contribution of copper defects recombination processes to the T_c recovery for sample HII irradiated by 2.4 MeV electrons.

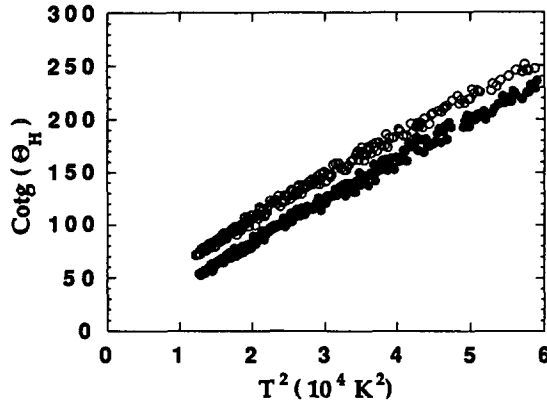


Fig. 4. — $\text{Cotg}(\Theta_H) = \sigma_{\parallel} / \sigma_{\perp}$ (with Θ_H the Hall angle) versus T^2 for sample HI before (full circles) and after irradiation (open circles) by 0.4 MeV electrons.

experimental support to Anderson's theory, but in this form, to compare our results with those of Chien *et al.* [27] obtained from a sequence of Zn doped samples. Indeed, when we plot the $\Delta T_c / T_{c0}$ dependence on F (see Fig. 5) we observe a very good agreement between their results and ours, which indicates that irradiation effects on T_c seem to be of the same kind as those produced by Zn doping.

4. Discussion.

We have clearly shown that T_c is sensitive to displacements of oxygen and copper atoms with respective displacement threshold energies E_d of 10 and 15 eV. Determination of the oxygen threshold energy by electron microscopy leads to higher values of $E_d \approx 15$ eV with the conclusion that YBCO is insensitive to 100 keV electron irradiation [12, 29]. These results are in contradiction with ours. It is worth mentioning here that electron fluences and rates used in

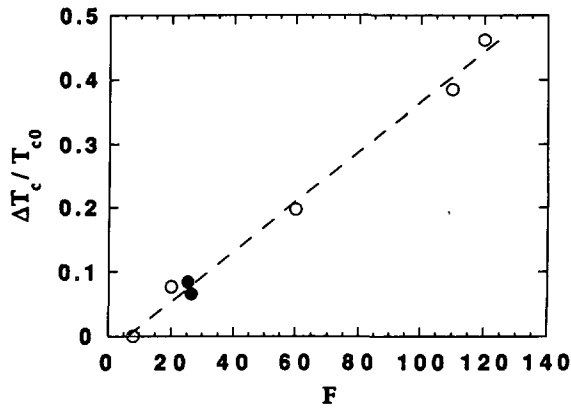


Fig. 5. — T_c reductions normalized to its initial value as a function of F (see text) for irradiated samples HI and HII (full circles) and Zn doped samples from [27] (open circles).

electron microscopy experiments are much higher than ours and so defects visible by this technique may be different from those responsible for the initial T_c decrease. In particular, Basu *et al.* [29] determined the oxygen displacement threshold energy from the changes in the splitting of the (1, 1, 0) diffraction spots in the [0, 0, 1] diffraction pattern induced by the redistribution of oxygen atoms in the CuO chain planes. However, we have verified that a sample of YBCO irradiated by 2.4 MeV electrons up to $T_c = 0$ is still orthorhombic [30], which indicates that the damage on T_c should precede any visible modification of orthorhombicity.

The key questions are : 1) from which sites the oxygen and copper atoms responsible for the T_c decrease are ejected ; 2) by which mechanism do the ejections induce the T_c decrease. Since we have to deal with a very complicated structure, it is very difficult to bring a clear-cut answer to the first question. Nevertheless, it is interesting to consider two particular cases : first, we may assume that only the displacements of the chain atoms induce a T_c reduction ; secondly, that only the displacements of the atoms belonging to the CuO_2 planes play a role in the T_c decrease.

The first assumption comes directly from the fact that chain atoms are weakly bound and then can be easily displaced. By now rewriting expression (2) as a function of the cross sections normalized to the chain atoms :

$$\sigma_d^{\text{Chain}} = (1/7 \sigma_d^{\text{O}} + 1/3 \sigma_d^{\text{Cu}}) \quad (3)$$

and noting that the experimental B/A involved in (2) is equal to $7/3$, we obtain the relation :

$$-\Delta \dot{T}_c = C \times \sigma_d^{\text{Chain}} \quad (4)$$

with $C = (23 \pm 3) 10^3$ K. This assumes that the probability for oxygen and copper atom displacements does not depend on their sites, and that only displacements in the chain affect T_c . Expression (4) leads to the conclusion that displacements of copper or oxygen chain atoms have the same effect on T_c . In other words, it seems that the important fact is to break the CuO chains disregarding the site where the break takes place.

For electron irradiation at low energies ($E < 0.35$ MeV), we found that only oxygen displacements affect T_c as observed in oxygen depletion experiments. But this analogy no longer holds in a quantitative comparison. Indeed from formula (4) 0.1 % d.p.a. leads to a T_c reduction of about 23 K. The same amount of oxygen removal from the chains leads to

0.4 K of T_c reduction for an initial fully oxygenated sample [31]. This indicates that the damaging mechanism induced by irradiation is totally different from the one caused by oxygen depletion, even if we suppose that irradiation only affects the chain structure. In the latter case, it is now well accepted that the T_c depression is due to a reduction of the hole concentration in the CuO_2 planes [31]. This effect is related to modifications in the band structure produced by short/long range ordering of the chain structure depending on the oxygen content. Such arrangements have been observed by TEM and, in particular, when the oxygen content equals 6.5 a new orthorhombic structure consisting of one over two chains fully oxygenated in the ab planes was observed [31]. It is worth mentioning here that calculations of the electronic structure of a fully oxygenated sample have shown that redistribution of oxygen atoms in the chains results in a drastic reduction of charge transfer and hole density in the CuO_2 plane even if the oxygen content remains constant. Such a mechanism, which has been proposed to explain effects of irradiation damage [18], predicts that the loss of superconductivity should be associated with a transition from the orthorhombic to a tetragonal structure. This situation was clearly not observed in electron irradiated samples as mentioned above [30].

From Hall data, it is tempting to compare irradiation effects with Zn substitution on the Cu sites. Concerning these experiments, the precise site where the substitution occurs is still a controversial issue. Nevertheless, it seems that the main effect of Zn doping is the appearance of local magnetic moments in the CuO_2 planes associated with the disorder induced by the Zn atoms substituted on the CuO_2 planes. Wherever the substitution takes place, a nominal doping of 0.1 % will induce a T_c reduction of about 4 K [27, 32].

In order to compare irradiation and Zn doping effects, let us consider the situation in which T_c is mainly affected by displacements within the CuO_2 planes. Then, expressing the variations of ΔT_c as a function of $\sigma_d^{\text{Op}} = 4/7 \times \sigma_d^{\text{O}}$ and $\sigma_d^{\text{Cup}} = 2/3 \times \sigma_d^{\text{Cu}}$, the displacement cross sections for oxygen and copper atoms in the CuO_2 planes respectively, we obtain :

$$-\Delta T_c \approx D \times (\sigma_d^{\text{Op}} + 2 \times \sigma_d^{\text{Cup}}) \quad (5)$$

where $D \approx 6 \times 10^3$ K and the constant 2 which multiplies σ_d^{Cup} were deduced from the experimental values A and B in (2). According to formula (5), the displacement of a copper atom is twice more efficient than that of an oxygen atom. This can be understood since ejection of a copper atom will break the array of Cu-O-Cu atoms in both $[1, 0, 0]$ and $[0, 1, 0]$ directions, while ejection of an oxygen atom will break this arrangement only along one of these directions. Within this interpretation, 0.1 % d.p.a. Cu will induce a T_c reduction of about 12 K, which is comparable with the 4 K obtained for an equivalent amount of Zn substitution.

In any case, we have to remember that the two pictures proposed above are limiting cases that we have considered in an attempt to establish a quantitative comparison between irradiation effects on the one hand, and oxygen or Zn doping effects on the other hand. The real situation is certainly more complicated and it is possible that displacements of apical oxygen and even of yttrium or barium atoms could play a role concerning the T_c damage. Nevertheless, the contribution of the heaviest atoms should be of the second order within the energy range studied here. Moreover, it is worth mentioning that the apical oxygen is expected to participate to the charge transfer by providing a fourfold square planar coordination to the Cu chain atoms when the chains are present [18]. In this context, the major contribution of an apical oxygen displacement should be related to a degradation of the charge transfer which we have shown to be a minor effect in the case of electron irradiations.

Since the presence of magnetic moments has been observed in irradiated samples [7] as well as in the Zn substituted ones [32 for instance], it seems natural to consider a pair-breaking mechanism due to the breaking of the time reversal symmetry. For this purpose, the classical

Abrikosov-Gorkov mechanism has been evoked [19]. Within this theory, a BCS point-like interaction between carriers is considered. Furthermore, it is assumed that the time reversal correlation function, $\langle K(t), K(0) \rangle$, decays exponentially in time leading to an ergodic situation. Namely we have :

$$\langle K(t), K(0) \rangle \propto \exp(-t/\tau_k) \quad (6)$$

Then, the T_c reduction rate is found to be proportional to the inverse relaxation time τ_k , which in the presence of uncorrelated magnetic impurities leads to [33] :

$$\Delta T_c \propto J^2 \times N(0) \times n_m \quad (7)$$

where J is the exchange integral between carriers and magnetic impurities, n_m is the magnetic impurities concentration, and $N(0)$ is the density of states at the Fermi level.

From formula (7), the T_c decrease must be linear on fluence and the T_c damage rate proportional to $N(0)$. Thus, one should expect a poorly oxygenated sample to be less sensitive than a rich oxygenated one, that is to say that if we consider a sequence of oxygen depleted samples, the T_c reduction must be faster for higher values of T_{c0} . We have observed exactly the opposite when we have irradiated sintered samples with different oxygen contents by electrons of 2.4 MeV [34]. The same results have been obtained by Vichery *et al.* [6] on single crystals irradiated by 2.5 MeV electrons. All this experimental evidence weakens the predictions of the Abrikosov-Gorkov model and further clearly establishes a difference between oxygen depleted and irradiated samples. Indeed, if the irradiation effects on T_c were the same as those due to oxygen removal, then the irradiation induced T_c decrease rate should progressively increase as T_c reduces (as observed in oxygen depleted samples) : this is in total disagreement with our results. On the other hand, irradiation results are consistent with results obtained from Zn substituted samples for which the T_c reduction rate induced by Zn doping decreases with the oxygen content [32].

5. Conclusion.

We have brought the first experimental evidence of the influence of displacing copper atoms by electron irradiation on the T_c reduction of YBCO single crystals. Furthermore we have determined threshold energies for oxygen and copper atoms to be 10 and 15 eV respectively. Our results clearly show that irradiation effects have nothing to do with oxygen removal effects, while they show that the former are similar to Zn doping effects. Finally, we have shown that the conventional Abrikosov-Gorkov depairing model induced by localized magnetic moments is not appropriate to explain the T_c depression either on irradiated samples or in Zn doped ones.

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