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Ion-Beam Induced Metal Insulator Transition in YBCO Films.

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Abstract

We have performed light ion irradiation experiments on high quality YBCO thin films, and studied the metal insulator transition induced by disorder. At small damage level the observed decrease of Tc is compatible with a depairing process. For disorder of about 0.04 dpa (displacement per atom), a localization transition is observed, leading to a 3D Variable Range Hopping process for the conductivity. We will discuss the connection between these two phenomena, and the specific role of intrinsic disorder vs extrinsic one.

1. Introduction

Since the discovery of the cuprate superconductors, many studies have been devoted to the interplay between their structure and their unique electronic properties. In particular, ion irradiation experiments have been carried out to induce disorder in these materials in a controllable manner [1], inducing a rapid transition to an insulator state [2]. The type of insulator created is still under debate. For Meyer at al [3], a band insulator is found, whereas Valles et al [4] argue in favour of a Coulomb gap. Here, we present a set of recent experiments on He irradiation on high quality YBCO thin films performed at low temperature (10K) to freeze the disorder. The main results of this paper are the following : (i) the initial decrease of Tc *is not* only due to the reduction of the carrier density, but also of the lifetime of the Cooper pairs: (ii) the metal insulator transition is driven here by the disorder, leading to a conventional 3D Anderson insulator. Emphasis will be made on how to distinguish between intrinsic and extrinsic properties revealed by these experiments.

2. Experiments

YBCO thin films are grown by co-deposition of the metals in a partial pressure of ionized oxygen onto a hot single-crystal substrate.[5] Using (100) $SrTiO_3$ substrates, we routinely obtain films from 10nm to 100nm thick, stoechiometric, with Tc in the 88K-90K range, and resistivity at room temperature between 250 and 300 μΩcm. In our irradiation experiments, we used 80nm thick films, with evaporated silver contacts to perform standard four-probe measurements. Special care was taken to insure that the whole film area was irradiated. The films were mounted in a home-built cryostat, on-line with a 2 MV Van de Graaf accelerator. 1 MeV He+ ions were sent onto the films held below 10K. The projected range of such particules is 2 μm : therefore, the ions do not stop in the film *and* the defect profile is very flat. This leads to an homogeneous concentration of defects and insures a correct determination of the electronic transport properties. Resistivity as a function of temperature was measured from 1.5K to 120K between each dose. This latter temperature was chosen to avoid annealing of defects, and consecutive recovery of superconducting properties [6].

3. Results

We irradiated the samples with fluences up to $3.7 10^{16}$ He^+/cm^2. It has been shown [7] that nuclear losses determine the behavior of these materials under light ion irraditions in the MeV range. We will not consider here heavy ion irradiation experiments where the deposited energy is high, the cascades very dense and the defects correlated. Using TRIM code [8] and a displacement energy of 20 eV, we thus calculate a rate of 0.0012 dpa (displacement per atom) for 10^{15} He^+/cm^2, and use the dpa parameter to renormalize experiments made with different ions and energies.

1.1. *Reduction of Tc*

The stricking features of the observed ρ vs T curves as irradition proceeds are the following :

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the resistivity increases, the Tc (critical temperature) decreases, and δTc (width of the transition) increases first moderately, and then dramaticaly as Tc goes to zero. Let us first focus on Tc. To deal with intrinsic properties, we chose to study the so-called TcOnset (Tc_o), where the resistance drops sharply. Studying the superconductive fluctuations in the Lawrence and Doniach (LD) model [9], we can show that Tc_o corresponds approximatively to the mean field Tcmf (extracted from the best fit -inset figure 1-). At the very beginning of the experiments ($dpa<8.10^{-3}$), the rounding of the transition corresponds to a rapid increase of the normal state resistivity ρ_n, and thus an amplification of the thermodynamic fluctuations.

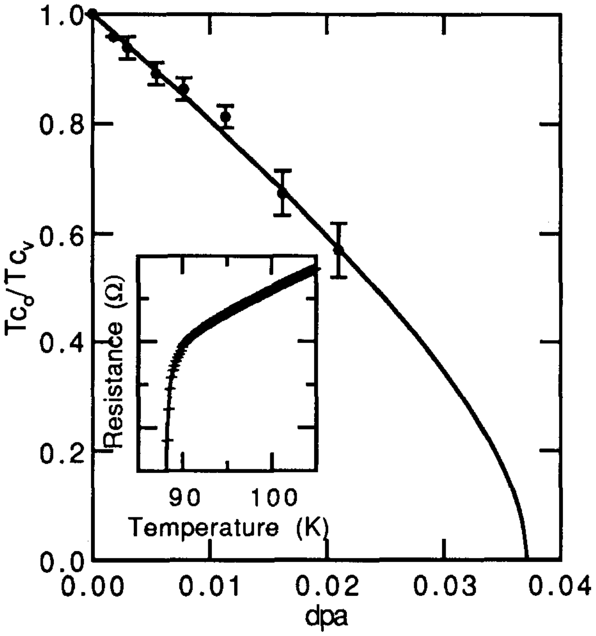

Fig 1. Normalized critical temperature as a function of dpa. The inset shows a fit (solid line) of the fluctuations using the LD model.

In figure 1, we plot the Tc_o reduction (normalized by Tc_v, the critical temperature of the virgin film) as a function of dpa. Within 25%, these data fall on the same curve than the one reported in ref 10, a compilation of different light ion irradiation experiments. This confirms the specific and reproductible sensitivity of Tc in the cuprate superconductors in presence of small disorder. In previous papers [11], authors proposed that the main contribution to the Tc_o depression was the disorder in the CuO chains of YBCO. Recent electron irradiations experiments on YBCO single crystals [12] gave a solid basis to this hypothesis ; the displacement energies of the Cu and O atoms of the chains have been found to be respectively 10 eV and 15 eV, much lower than the other atoms in the structure. Theoretical investigations by Gupta et al [13] lead to the conclusion that fully disordered CuO chains cut down the charge transfer to the CuO2 planes, and thus induce a superconductor-insulator transition. But in the experiments, Tc_o goes to zero for a critical disorder on the order of 0.03 to 0.04 dpa. This is far from reaching the point where *all* the CuO chains are disordered. Therefore, an additional and efficient mechanism has to be incorporated to explain the drastic Tc_o reduction.

In a previous paper [10] we proposed a more complete form for the Tc_o reduction. We supposed that a defect in YBCO introduces a finite lifetime for the Cooper pairs, and that the Tc_o/Tc_v obeys a depairing-like law :

$$\ln\left(\frac{Tc_o}{Tc_v}\right)=\psi\left(\frac{1}{2}\right)-\psi\left(\frac{1}{2}+0.14\frac{Tc_v\,.\,dpa}{Tc_o\,.\,dpa_c}\right) \quad (1)$$

where Ψ is the di-gamma function and dpa_c a critical value of disorder where Tc_o becomes null. Applying this to the new experiments (solid line in figure 1) leads to $dpa_c=0.037$, very close to the previous determination. Because of a lack of very precise data close to dpa_c, it is difficult to unambiguiously determine the origine of the depairing (magnetic impurities, proximity effects, approach of an Anderson transition ...). We will return to this point in the next section.

1.2. Metal Insulator Transition (MIT)

Figure 2 displays on a logarithmic scale the ρ vs T laws for increasing disorder between 0.02 to 0.045 dpa. We clearly observe an insulator state well before the orthorhombic to tetragonal transition is known to occur (>0.08 dpa [14]). This essentially means that the MIT is not simply driven by the structural transition as in the oxygen-depleted samples, but by the disorder itself. Let us use the Mott criteria [15] to get a rough estimate of the expected transition region.

$$\sigma_m = 0.026\,.\,\frac{e^2}{hd} \quad (2)$$

where σ_m is the minimum metallic conductivity and d the interatomic distance (here d=4Å). This

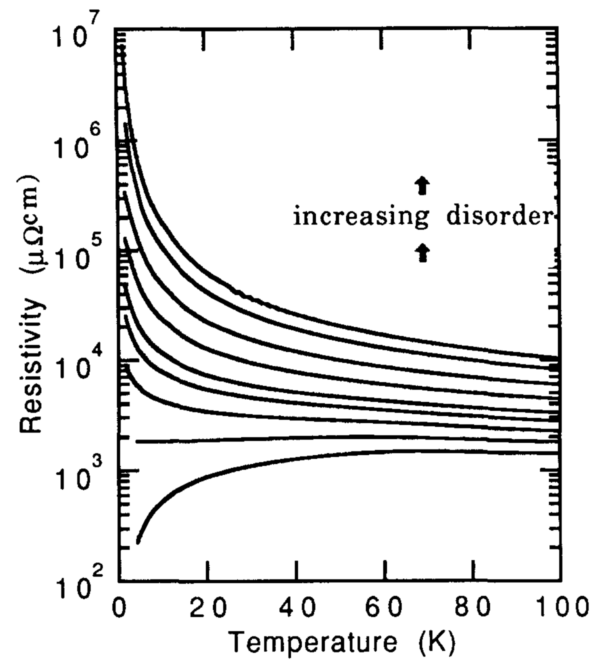

Fig 2. Resistivity as a function of temperature for fluences (in 10^{15} He/cm^2) : 17.5 ; 20 ; 22 ; 24 ; 26 ; 29 ; 32 ; 35 ; 37.

corresponds to a resistivity of about 6000 μΩcm.. The arrow on figure 2 indicates the region where the transition occurs, i.e for a residual resistivity of about 5000 to 6000 μΩcm, in fair agreement with the Mott criteria. To confirm that we do observe a localization transition, we analyse the exponential decay of the resistivity at low temperature :

$$\rho=\rho_o \,.\, \exp\left(\left(\frac{T_o}{T}\right)^n\right) \qquad (3)$$

where ρ_o is a constant, n=1 for an activated process above a gap, n=0.25 for a Mott Variable Range Hopping (VRH) between localized states in 3D, and n=0.5 for VRH with strong electron-electron interactions [16] .It is clearly seen from the plot in figure 3 that n=0.25 is the best candidate to fit our data.

In fact the VRH process is observed in the whole insulating region, with an increasing To as the disorder increases. The calculated To displayed in table 1 range from 600 to 16000 K, as usualy observed in disordered systems [16] . From them we can compute the localization length ξ by

$$\xi=(n(0)\ k_B\ T_o)^{-1/3} \qquad (4)$$

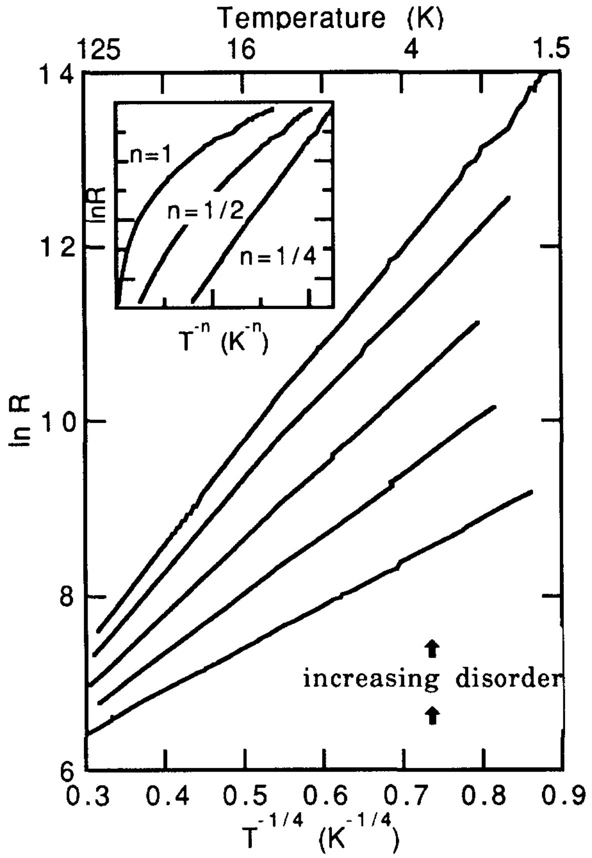

Fig 3. VRH laws for fluences (in 10^{15} He/cm^2) : 26 ; 29 ; 32 ; 35 ; 37. The inset shows different exponential laws (see text) : VRH is the best one.

where n(0) is the Fermi Density of States. The reported values of ξ in table 1 have been calculated with n(0)=5 10^{21} states/cm^3/eV. It is interesting to note that they correspond to few atomic spacings ; in particular, they are rapidly shorter than the c-axis parameter of YBCO (i.e 12 Å). Therefore the original anisotropy of these materials is progressively distroyed, and a 3D VRH is indeed observed.

Table 1.

He/cm^2	26.10^{15}	29.10^{15}	32.10^{15}	35.10^{15}	37.10^{15}
To (K)	580	2100	5140	10050	15950
ξ (Å)	18.6	12.1	9	7.2	6.2

3. Discussion

In the latter section we showed that the observed MIT displays the usual features of an

Anderson transition. It is well known that localization effects depress Tc in strongly disordered superconductors [17]. It is thus interesting to see if this applies to irradiated YBCO.Maekawa et al [18] computed the corrections to Tc induced by weak localization effects :

$$\delta Tc/Tcv \approx \rho^2 \text{ in 3D} \quad \delta Tc/Tcv \approx R\square \text{ in 2D}$$

We thus plot the normalized Tco as a function of the normalized resistivity for different low temperature irradiation experiments on YBCO (figure 4). Surprisingly, the data lie on two separate curves. The upper one represents the low resistivity samples ($\rho(100K) \approx 100 \mu\Omega cm$), the other one a great variety of samples with resistivities up to 10 times higher. It is clear that extrinsic effects dominate in this latter case, and that no quantitative comparison with theories for homogeneous media can be made. It is worthwhile noticing that the Tco decrease as a function of dpa is roughly the same for all these samples.

The overall Tco variation is linear for the best samples, which could be interpreted as a 2D behavior. But looking closer to the data (inset figure 4), we observe an *upward* curvature at low resistivity, not compatible with the above equations. Furthemore, no evidence of weak localization effects are seen in the normal state resistivity curves in this disorder range. It thus appears that these effects cannot simply explain the initial Tc depresssion, unless YBCO is not a "regular disordered metal" as proposed by Coffey et al [19] few years ago.

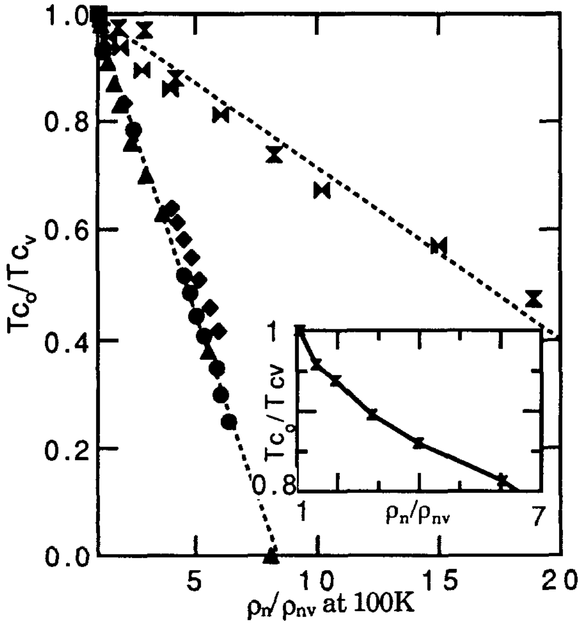

Fig 4. Normalized Tc_0 as a function of normalized resistivity showing two kinds of samples (lines are guides for the eyes). The inset shows the upward curvature at low resistivity.

4. Conclusion

The Tc depression of YBCO in ion irradiation experiments cannot be simply explained in terms of carrier density lowering, but involves a depairing-like process. Furthermore, an Anderson metal-insulator transition is induced , 3D in character and for a surprisingly low disorder (dpa≈4-5%). The initial lowering of Tc is not due to localization effects, and more work needs to be done to get a clearer picture on these puzzling effects.

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