International Journal of Modern Physics B Vol. 3, No. 1 (1989) 87–92 © World Scientific Publishing Company

# EFFECTS OF LOCALISATION IN ATOMIC-DISORDERED HIGH-T, SUPERCONDUCTORS

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Received 27 June 1988

The influence of disordering on the properties of high- $T_{\rm c}$  superconductors is investigated. The results show that pairing takes place in the systems of localised electrons even in slightly disordered samples. Superconductivity exists till the radius of localisation exceeds the typical size of the Cooper pairing in highly disordered system.

#### 1. Introduction

Fast neutron irradiation is one of the purest methods to investigate the effects of disordering on physical properties of high-temperature superconductors (HTSC). It is known that disorder which is not connected with magnetic impurities according to Anderson theory should not appreciably affect superconducting transition temperature  $T_{\rm c}$  of the conventional (wide-band) superconductors. However, the properties of such superconducting compounds such as A-15, C-15, Chevrel phases turn out to be highly sensitive to disorder. Thus, the response of the system to introduction of defects helps to better understand the peculiarities of its electronic properties in the ordered state characterised in our case by high values of  $T_{\rm c}$ .

Previously<sup>1-3</sup> we have shown that disordering of YBaCuO and LaSrCuO leads to a rapid decrease in  $T_c$ . The derivative of the upper critical field  $H'_{c2}$  practically does not change. At large fluences instead of linear dependence the Mott-type temperature dependence of  $\rho(T)$  is observed in the temperature interval 5 < T < 300 K:

$$\rho(T) = A \exp(Q/T^{1/4}). \tag{1}$$

While measuring  $\rho$  of both systems at 80 K directly during fast neutron irradiation we found that  $\rho_{80}$  grows exponentially with fluence  $\phi$ , i.e.  $\rho_{80} \sim \exp(a\phi)$  beginning with the smallest  $\rho$ . According to experimental data localisation effects are essential in these systems even at the smallest extent of disorder.

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The present study is a further investigation of localisation effects in high- $T_{\rm c}$  superconductors including BiSrCaCuO (zero resistivity at  $T=70~{\rm K}$ ,  $\rho_{300}\approx 2~{\rm m}\Omega\cdot{\rm cm}$  and linear temperature dependence of  $\rho(T)$ ), and single-crystal YBaCuO sample ( $T_{\rm c}\approx 80~{\rm K}, \rho_{300}\approx 2~{\rm m}\Omega\cdot{\rm cm}$ ). Here we investigated also the influence of isochronous low-temperature annealing ( $T_{\rm ann}\leq 300~{\rm K}$ ) on electrical resistivity of high- $T_{\rm c}$  materials disordered under fast neutron irradiation ( $T_{\rm irr}=80~{\rm K}$ ). Besides, we present here the data on the NQR spectrum and spin-lattice relaxation time  $T_{\rm l}$  for the sample of YBa<sub>2</sub>CU<sub>3</sub>O<sub>6.95</sub> irradiated by a fluence of  $5\times 10^{18}~{\rm cm}^{-2}$ .

# 2. Experimental Results

Radiation disordering, deviation from oxygen stoichiometry, introduction of impurities into Cu sublattice suppress superconductivity in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. Structural changes are different in all cases. Orthorhombic parameter (*b*-*a*) gradually decreases with decreasing oxygen content. In this case oxygen is removed from sites O4 (in chains). As for sites O5, they are filled slightly with increasing O<sub>2</sub> just before the tetragonal phase transition.<sup>4</sup> Radiation disordering results in oxygen distribution over sites O4 and O5 and growth of the mean-quadratic atomic displacements. The structure, however, remains orthorhombic. Appearance of oxygen vacancies in sites O4 is clearly seen in the NQR spectra for the irradiated sample of YBaCuO (Fig. 1). The structure of the NQR line on the <sup>63</sup>Cu atoms in Cu2 site is associated with vacancy configurations in O4 sites as well as in oxygen-deficient samples.<sup>5</sup> Spin-lattice relaxation time  $T_1$  on <sup>63</sup>Cu nuclei in Cu2 sites in normal state ( $T = T_c$ ) increases from 0.5 to 150 msec on changing the oxygen content from  $\delta \approx 0$  to  $\delta \approx 0.2$ . Radiation disordering ( $T_c \approx 70$ 

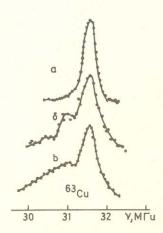


Fig. 1. Evolution of the NQR spectrum on the nuclei  $^{63}$ Cu in Cu2 site for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> a)  $\delta = 0.05$ ;  $\delta$ ) the sample irradiated by a fluence of 5  $\times$  10<sup>18</sup> cm<sup>-2</sup> ( $T_c \approx 70$  K); b)  $\delta = 0.2$  ( $T_c \approx 70$  K). Frequency is given in MHz.

 $K, \phi = 5 \times 10^{18} \, \mathrm{cm}^{-2}$ ) leads also to an increase in  $T_1$  but to a lesser extent. Assuming that the whole  $T_1$  increase is connected with the change of electron states density on the Fermi level  $N(E_{\rm F})$  it will decrease more than by order in the first case and by 25% in the second. Distribution of relaxation times over Cu1 sites of the irradiated sample points to appearance of disorder in the electron environment of the Cu atoms in these sites. Measuring  $T_1$  in a superconducting state ( $T_{\rm c} \approx 70 \, \mathrm{K}$ ) of the irradiated sample yields the value  $2\Delta/kT_{\rm c} \sim 12$  (as in the initial sample). This shows a very strong coupling regime (bearing in mind the maximum value of a gap). In an oxygen-deficient sample with approximately the same  $T_{\rm c}(\delta \approx 0.2)$  the value of a reduced gap decreases to  $2\Delta/kT_{\rm c} \sim 4$ –6 for both sites of the Cu atoms. Temperature independent contribution to magnetic susceptibility  $\chi_0$  also behaves differently in both cases under consideration, i.e. it drops with increasing  $\delta$  and grows—under—irradiation. Growth—of— $\chi_0$ —under—irradiation—and simultaneous increase of spin-lattice relaxation time shows that an attempt to explain their change only by  $N(E_{\rm F})$  variations leads to a contradiction.

Variation of the oxygen content directly alters the electron structure of the YBaCuO system since the concentration of charge carriers changes. Radiation disordering affects the electron structure only through disorder. Differences in behavior of YBaCuO on introducting defects of different types stress, however, the general property: degradation of  $T_c$  always goes with transition to dielectric behavior of resistivity. The samples of BiSrCaCuO, as well as YBaCuO, with high enough  $T_c$  (relatively small disordering) are characterised by linear growth of electrical resistivity with temperature (Figs. 2,3).

Increasing extent of disorder leads to exponential temperature dependence of electrical resistivity described by Eq. (1). After annealing for 20 min at T = 300 K  $T_c$  appears and linear dependence of  $\rho(T)$  re-establishes practically within the

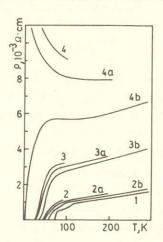


Fig. 2. Temperature dependence of electrical resistivity for the disordered BiSrCaCuO sample: 1) the initial sample; 2)  $\phi = 2 \times 10^{18}$ ; 3)  $\phi = 5 \times 10^{18}$ ; 4)  $\phi = 7 \times 10^{18}$  cm<sup>-2</sup>. Indexes a and b correspond to annealing at T = 200 K and T = 300 K, respectively.

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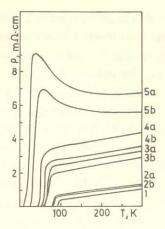


Fig. 3. Temperature dependences of electrical resistivity for disordered YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.95</sub> samples: 1) the initial sample; 2)  $\phi = 2 \times 10^{18}$ ; 3)  $5 \times 10^{18}$ ; 4)  $7 \times 10^{18}$ ; 5)  $10 \times 10^{18}$  cm<sup>-2</sup>. Indexes a and b correspond to annealing at T = 300 K for 20 min and two weeks, respectively.

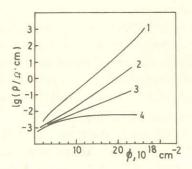


Fig. 4. Dependences of  $\rho_{80}$  on fluence of fast neutrons for ceramic and single-crystal YBaCuO samples (curves 1 and 2, respectively), BiSrCaCuO (3) and SnMo<sub>6</sub>S<sub>8</sub> (4).

whole temperature range beginning from  $T_c$  to 300 K. For  $\phi \ge 10^{19}\,\mathrm{cm}^{-2}$  annealing at  $T \le 300\,\mathrm{K}$  leads only to a decrease in coefficients A and Q in Eq. (1) while  $T_c$  does not appear at all. Figure 4 shows the dependence of  $\rho_{80}$  on fluence of fast neutrons for materials under study. The value of  $\rho_{80}$  grows exponentially with fluence. According to measurements made in the basal plane of a single crystal, YBaCuO sample exponential growth of resistivity of  $\rho_{80}$  is the property of the material itself and not a consequence of the preparation technique of ceramic samples. The data from Fig. 4 as well as the behavior of  $dH'_{c2}/dT^6$  and  $\rho(T)$  in the disordered HTSC sample show that localisation effects are likely to appear well before the disappearance of  $T_c$ . But superconducting transition does not allow observation of this experimentally on the temperature dependence of  $\rho(T)$ .



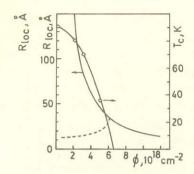


Fig. 5. Dependence of  $R_{loc}$  (solid line) and  $T_c$  (open circles) on fluence of fast neutrons for YBaCuO. The dashed line shows the values of  $R_{loc}$  calculated by Eq. (2).

#### 3. Discussion

It is well known that sufficient disorder may result in the metal-insulator transition due to electron localisation (Anderson transition). Anderson showed that in localised electron system Cooper pairing may take place only between the electrons with centres of localisation lying within the sphere of the order of localisation length  $R_{loc}$ . The states of such electrons are split by energy to a value of the order  $[N(E_F)R_{loc}^3]^{-1}$ . Apparently the condition should be held that the value of a superconducting gap  $\Delta$  (at T=0) is essentially higher than the value of this splitting:

$$\Delta \sim T_{\rm c} >> [N(E_{\rm F})R_{\rm loc}^3]^{-1}$$
 (2)

Equation (2) is equivalent to the requirement that the localisation radius should be essentially higher than the typical size of the Cooper pair in highly disordered systems. Using experimental data on electrical resistivity of radiation-disordered samples of YBaCuO one may calculate variation of  $R_{loc}$  depending on fluence and with Eq. (2) estimate the limiting values of  $R_{loc}$  for the experimental values of  $T_c$ . Suppose that the left side of Eq. (2) equals  $5T_c$  and  $N(E_F) \approx 5 \times 10^{33} \, (\text{erg} \cdot \text{cm}^3)^{-11}$  for all fluences one may obtain the values of  $R_{loc}$  shown by dots in Fig. 5. From this figure it is seen that at  $6 \times 10^{18} \, \text{cm}^{-2}$  the qualitative criterion of Eq. (2) is not valid. These estimates are in surprisingly good agreement with the experimental data.

#### 4. Conclusion

Quasi-two-dimensional systems to which the superconductors under study belong are expected to show strengthening of effects of localisation that occurs at values of conductivity appreciably exceeding typical values of three-dimensional

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systems. The properties of electronic system of HTSC's make them close to Anderson metal-insulator transition even in the ordered state. This feature differentiates new HTSC's from previously known superconductors of other systems. In the first case pairing is likely to occur in systems of localised electrons even at the smallest disorder.  $T_c$  sharply decreases when at low temperatures the dependence  $\rho(T)$  of type (1) is observed. Superconductivity is fully suppressed when energetic splitting between the localised states becomes comparable with the value of a superconducting gap.

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