ON THE HIGH T_c-SUPERCONDUCTIVITY IN $(Y_{1-x}Ba_x)_2CuO_{4-\delta}$ AND $La_{1.8}Sr_{0.2}CuO_4$

A. M. UMARJI, I. K. GOPALAKRISHNAN*, J. V. YAKHMI*, L. C. GUPTA, R. VIJAYARAGHAVAN and R. M. IYER*

Tata Institute of Fundamental Research, Homi Bhabha Road, Bombay 400 005, India.

**Bhabha Atomic Research Centre, Trombay, Bombay 400 085, India.*

ABSTRACT

We have discussed briefly the results of our experimental investigations on extra high T_c-superconducting multiphase material $(Y_{1-x}Ba_x)_2CuO_4$. In order to understand the microscopic mechanism of superconductivity in the single phase system $La_{1.8}Sr_{0.2}CuO_4$, we have undertaken detailed investigations such as NMR of copper nuclei and the response of superconducting behaviour to the presence of paramagnetic impurities. Initial results of these efforts have been presented.

PAST couple of months have witnessed an unprecedented excitement over the discovery of high T_c-superconductivity in copper-based oxide materials. It was Bednorz and Muller[1] who first suggested the possibility of high T_c-superconductivity in a mixture of crystalline phases in the Ba-La-Cu-Θ system. Subsequently, Chu *et al*[2] confirmed the existence of high T_c-superconductivity ($T_{co} \sim 36$ K) in this system prepared by coprecipitation method. Their sample was also multiphase. Cava *et al*[3], however, published almost simultaneously the discovery of superconductivity ($T_c \sim 36$ K) in a single phase material $La_{1.8}Sr_{0.2}CuO_4$. The performance of their sample was very much improved over that of Chu *et al* in that the width of transition T_c was ~ 1.4 K and the fraction of the sample which underwent superconducting transition was ~ 60–70%. We have confirmed[4] their findings and extended their investigations to other chemically similar materials, namely, $La_{1.8}Ba_{0.2}CuO_4$ and $La_{1.8}Ca_{0.2}CuO_4$. Details of these studies will soon appear in a series of publications. Similar results have also been obtained by other groups as well.

The situation took a dramatic turn when Wu *et al*[5] reported superconductivity occurring at an extra high temperature ($T_c \sim 93$ K) in a multiphase system Y-Ba-Cu-O. The superconductivity properties of this system, however, are extremely sensitive to the sample preparation technique, heat treatment conditions and the chemical composition. For instance, four independently prepared materials of the same nominal composition $(Y_{0.6}Ba_{0.4})_2CuO_{4-\delta}$ have been found[6-9] to exhibit superconductivity with the superconductivity onset temperature between 110 K and 87 K and the midpoint transition temperature T_c lying between 90 K and 73 K.

We describe here briefly the results of our measurements on three samples of the system $(Y_{1-x}Ba_x)_2CuO_{4-\delta}$ ($x = 0.3$, 0.4 and 0.5). The samples were prepared using the procedure described by Wu *et al*[5]. The sample with $x = 0.3$ is parrot-green, hard and an insulator. A piece of this sample with dimensions 2 mm × 2 mm × 10 mm has a room temperature (25° C) resistance of more than 5 MΩ which becomes ~ 40 MΩ at $T \sim 100$ K. Sample with $x = 0.5$ also exhibits a similar insulating behaviour. Sample with $x = 0.4$ which is greenish in colour with black dots dispersed in it, however, has drastically different electrical properties. A piece of this material with similar dimensions (2 mm × 2 mm × 10 mm) has a room temperature resistance of about 7Ω.

Temperature dependence of the resistance, $R(T)$, of the sample with $x = 0.4$ is shown in figure 1. The temperature coefficient of the resistance, $dR(T)/dT$, is negative initially, just as in a semiconductor, until at $T \sim 90$ K; $R(T)$ starts decreasing rapidly with further decrease in temperature. At and below about 57 K, the resistance becomes zero within the measuring accuracy of our instrument. The width of the transition ΔT_c is 25 K with the midpoint superconducting transition temperature $T_c \sim 73$ K. An additional heating at 1200 K for about 12 hr improves the performance. T_{co} becomes ~ 93 K and $T_{CF} \sim 65$ K. This behaviour must be constrasted with that observed by Chu *et al* in their sample. While ΔT_c in their sample was 13 K, T_c was as high as 93 K. Also their sample exhibits a metallic behaviour viz $dR(T)/dT$ positive, at $T > T_{co}$.

It is, therefore, abundantly clear that the superconductivity as well as the normal state properties of this system are highly unstable with respect to any

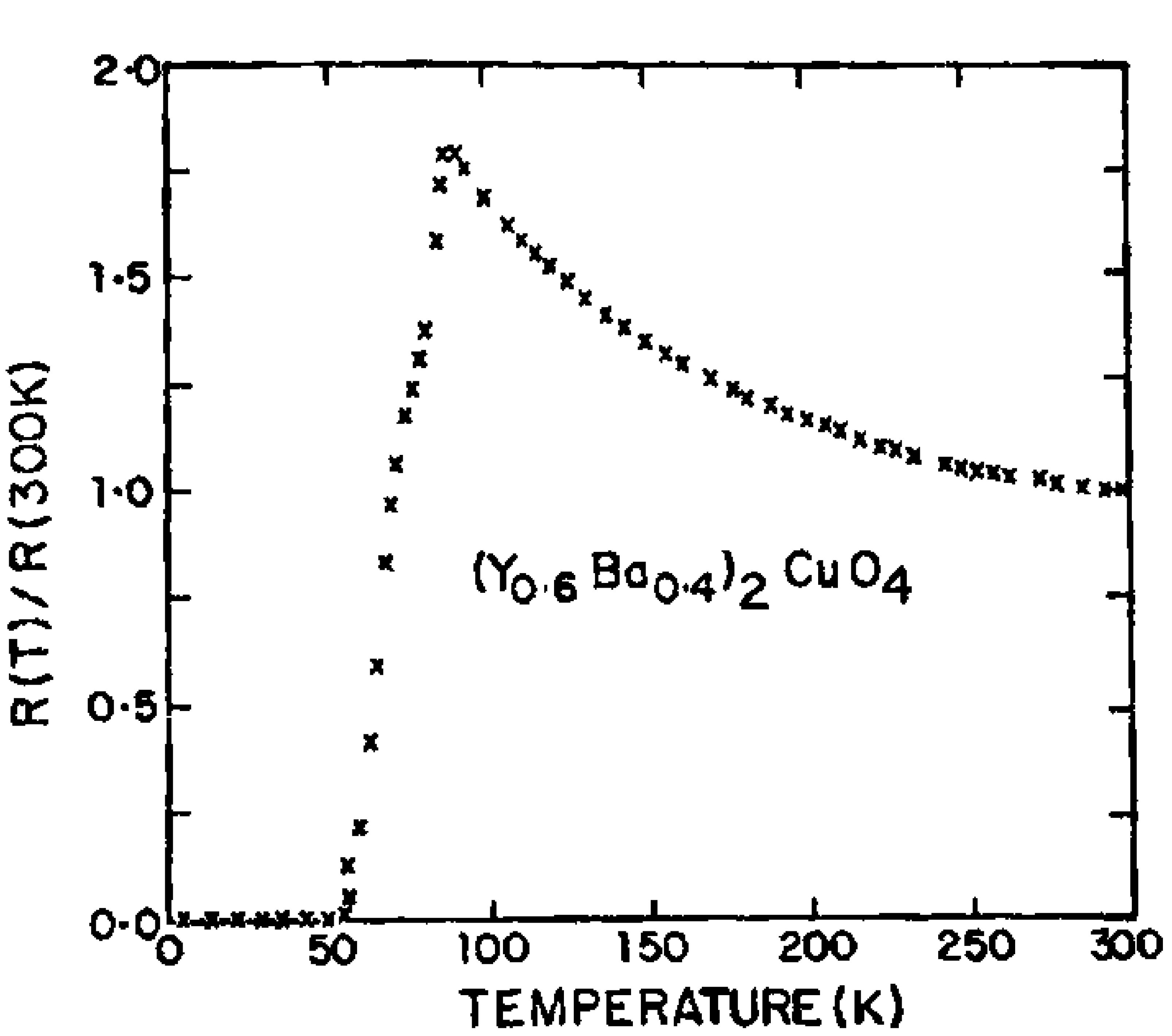

Figure 1. Resistance $R(T)$ of a sample of the system $(Y_{1-x}Ba_x)_2CuO_4$ with nominal composition $(Y_{0.6}Ba_{0.4})_2CuO_4$ as a function of temperature. The resistance has been normalized with respect to R(300 K), the value at room temperature.

variation in its physical conditions. While the phenomenon of high T_c-superconductivity witnessed in this system is truly exciting, the system *per se* may not be a suitable candidate from the physicist's point of view to characterize the nature of high T_c-superconductivity. On the other hand, unless the material is prepared under highly controlled physical conditions, it may not be very much suited for industrial applications.

The oxide system $La_{1.8}Sr_{0.2}CuO_2$ does not seem to be so much sensitive to the variation in physical conditions. This provides an ideal testground to examine the nature of high T_c-superconductivity. A detailed investigation of this system, therefore, is essential.

One of the most fundamental question that arises with respect to the phenomenon of high T_c-superconductivity in $La_{1.8}Sr_{0.2}CuO_4$ is whether there is a localized magnetic moment on copper atoms, or is it that the d-orbitals of copper atoms form a narrow d-band having d-holes (with large effective mass) that are responsible, possibly along with low-lying electronic excitation in a rather flat band (low effective mass), for this enhanced superconducting behaviour[10]? The possibility of an impending crystallographic phase transition (softening of a phonon mode) just before the transition in this material must be investigated. This will need material in the form of a single crystal for which efforts are being made. Some of these questions regarding the underlying microscopic mechanism of superconductivity in well characterized samples of the system $La_{1.8}Sr_{0.2}CuO_4$ are already being looked into in our studies. According to our preliminary results, Knight Shift of copper nucleus is − 1.0% and largely temperature independent. With more detailed NMR investigations, we hope to obtain information about the magnetic behaviour of this system. Another important issue concerning the microscopic mechanism of superconductivity is to examine as to how sensitive T_c is with respect to the presence of paramagnetic impurities in the system.

In a sample with composition $La_{1.75}Gd_{0.05}Sr_{0.2}CuO_4$, we find that the width of superconducting transition becomes at least twice that in a sample without any Gd impurity. There are well defined predictions[11] with respect to pair breaking in a superconducting matrix doped with paramagnetic rare earth impurities. We are examining effects of other impurities, besides Gd, on the superconducting behaviour of this system.

To summarize, we find that extra high T_c-superconducting behaviour in the multiphase system $(Y_{1-x}Ba_x)_2CuO_{4-\delta}$ is very sensitive to preparation technique, heat treatment and chemical composition. Our initial efforts to obtain information on the microscopic mechanism of superconductivity in a single phase system $La_{1.8}Sr_{0.2}CuO_4$, such as Cu-NMR and response of the system to paramagnetic impurities, have been briefly described.

9 March 1987

1. Bednorz, J. G. and Müller, K. A., *Z. Phys.*, 1986, **B64**, 189.
2. Chu, C. W., Hor, P. H., Meng, R. L., Gao, L., Huang, Z. J. and Wang, Y. Q., *Phys. Rev. Lett.*, 1987, **58**, 405.
3. Cava, R. J., Van Dover, R. B., Batlogg, B. and Rietman, E. A., *Phys. Rev. Lett.*, 1987, **58**, 408.
4. Gopalakrishnan, I. K., Umarji, A. M., Yakhmi, J. V., Gupta, L. C., Iyer, R. M. and Vijayaraghavan, R., *Mat. Lett.*, 1987, (communicated).
5. Wu, M. K. *et al.*, *Phys. Rev. Lett.*, 1987, **58**, 908.
6. Rao, C. N. R., private communication.
7. Grover, A. K. and Sampathkumaran, E. V., private communication.
8. Multani, M. S., private communication.
9. Malik, S. K., private communication.
10. Jha, S. S., (personal discussion regarding this aspect).
11. Abrikosov, Gor'kov, L. P., *Sov. Phys. JETP*, 1961, **12**, 1243.