Bull. Mater. Sci., Vol. 17, No. 6, November 1994, pp. 585–594. © Printed in India.

Critical exponent of the electrical conductivity in the paracoherence region of a thin film of $YBa_2Cu_3O_{7-x}$

MANMEET K MARHAS, K BALAKRISHNAN, V GANESAN, R SRINIVASAN, D KANJILAL*, G K MEHTA*, G K MURALIDHAR**, G MOHAN RAO**, SENTHIL NATHAN** and S MOHAN**
Inter University Consortium for DAE Facilities, University Campus, Khandwa Road, Indore 452 001, India
*Nuclear Science Centre, Post Box 10502, Aruna Asaf Ali Marg, New Delhi 110 067, India
**Instrumentation and Services Unit, Indian Institute of Science, Bangalore 560 012, India

Abstract. Critical exponent of the electrical conductivity in the paracoherence region (γ) of the high temperature superconductor $YBa_2Cu_3O_{7-x}$ (YBCO) has been estimated for high quality thin film on ZrO_2 substrate prepared by high pressure oxygen sputtering. High energy ion irradiation was carried out using 100 MeV $^{16}O^{7+}$ ions at liquid nitrogen temperature to see the effects of disorder on the value of the exponent. The critical exponent (γ) changes from a value of about 2 to 1·62 upon irradiation. Studies were also carried out on this film to see the effect of ageing and annealing.

Keywords. High temperature superconductors; resistivity; critical exponents; paracoherence; heavy ion irradiation.

1. Introduction

Excess conductivity studies in the paracoherence region of high temperature superconductors (HTSC) is a subject of interest for the past few years. Superconducting transition is a second order phase transition characterized by an order parameter which is zero above the transition temperature T_{co} and grows to a finite value below T_{co}. Large number of studies were performed to study the effect of fluctuations of this order parameter as a tool to understand the nature of the phase transition near T_{co}. Fluctuation induced excess conductivity above the mean field temperature (T_m) in the so called paraconductivity region has been well studied by many authors (Frietas *et al* 1987; Ausloos and Laurent 1988; Hagen *et al* 1988; Veira *et al* 1988; Srinivasan *et al* 1989). However there are only a few studies in the paracoherence region of the high temperature superconductors.

In granular high temperature superconductors in a region of a few degrees above the temperature of zero resistivity it has been noted that the conductivity varies as $(T-T_{co})^{-\gamma}$ (Rosenblatt *et al* 1990; Krishnan *et al* 1992; Pureur *et al* 1990; Soret and Ammor 1991). Rosenblatt *et al* (1988) explained this behaviour as a phase transition arising from the onset of phase coherence between the different grains, in accordance with the theory of Deutscher *et al* (1974). However Rosenblatt *et al* (1990) found a value of 2·7 for γ in sintered pellet while the other investigators have found a value close to 1·3. Rosenblatt and his coworkers (Lebeau *et al* 1988) have shown that the tunneling Hamiltonian for these granular superconductors can be put in one to one correspondence with the Hamiltonian for a 3D XY ferromagnet. The critical exponent in the conductivity will then match with the critical exponent for the susceptibility of the 3D XY ferromagnet. This susceptibility exponent has been calculated to be 1·316 from renormalization group theory calculation (Le Guillou and Zinn–Justin 1977). It was also shown by several workers that the disorder in the strength of the exchange interaction does not change the critical exponent. If the γ in the conductivity of the granular superconductor is to be identified with the corresponding exponent in the susceptibility of the 3D XY ferromagnet, one would expect that the conductivity exponent also to be independent of disorder.

We performed experiments on a superconducting thin film of $YBa_2Cu_3O_{7-x}$ of about 3000 Å with heavy ion irradiation to see if the conductivity exponent remains unchanged or it changes with irradiation and subsequent annealing. The results are reported.

2. Experimental

High quality thin film of YBCO was prepared using high pressure oxygen sputtering facilities available at the Indian Institute of Science, Bangalore. C axis oriented film of YBCO was grown on (100) plane of the ZrO_2 as the substrate. Orientation was confirmed from powder X-ray diffraction pattern carried out using Rigaku D/MAX X-ray diffractometer. Figure 1 shows the X-ray diffractogram of the film before irradiation. Precise four-probe electrical resistance measurements were carried out using a fully automated electrical resistance measurements system comprising of a closed cycle refrigerator, a Schlumberger nanovoltmeter, an Advantest constant current source and a DRC-93C Lakeshore temperature controller. Temperature was controlled to an accuracy of 10 mK using a calibrated platinum resistance thermometer.

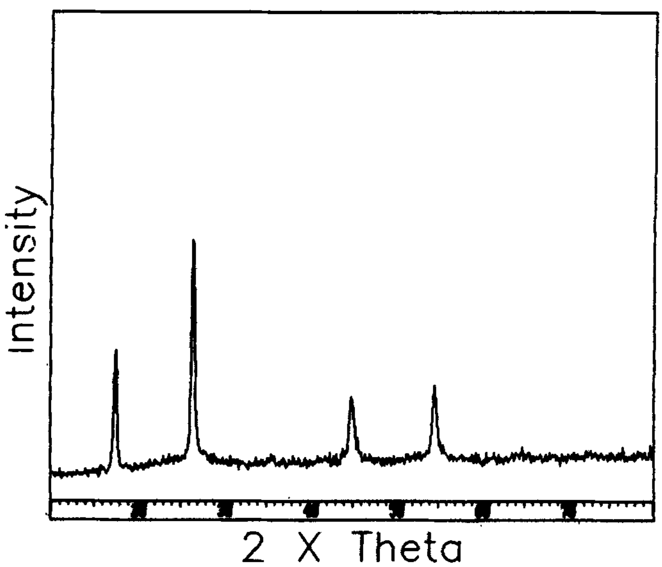

Figure 1. X-ray diffractogram for unirradiated film of YBCO.

Conducting silver paint was used to make ohmic contacts and extreme care was taken for the temperature stability and to correct the effects of thermal emfs.

The sample was irradiated by 100 MeV oxygen ion beam [$^{16}O^{+7}$] from the 16 MV tandem Pelletron Accelerator (Kanjilal *et al* 1993) at the Nuclear Science Centre, New Delhi. It was mounted on a liquid nitrogen cooled cold finger of the scattering chamber in the materials science beamline. The chamber was evacuated to a pressure of $1{\cdot}6 \times 10^{-7}$ m bar. The temperature of the top surface of $YBa_2Cu_3O_{7-x}$ film was about 95 K. The irradiation to a fluence level of 10^{13} particles/cm^2 was carried out using a beam having a flux of about 10^{11} particles/cm^2/sec and keeping the sample in a secondary electron suppressor geometry. The total number of particles falling on the sample was estimated by a combination of current integrator and pulse counter. After irradiation the cold finger got warmed up naturally to room temperature. After 10 h the chamber was vented to atmospheric pressure using dry nitrogen gas.

Resistance measurements were carried out before and after irradiation as well as after 60 days of irradiation to see the effects of ageing. The film was then annealed

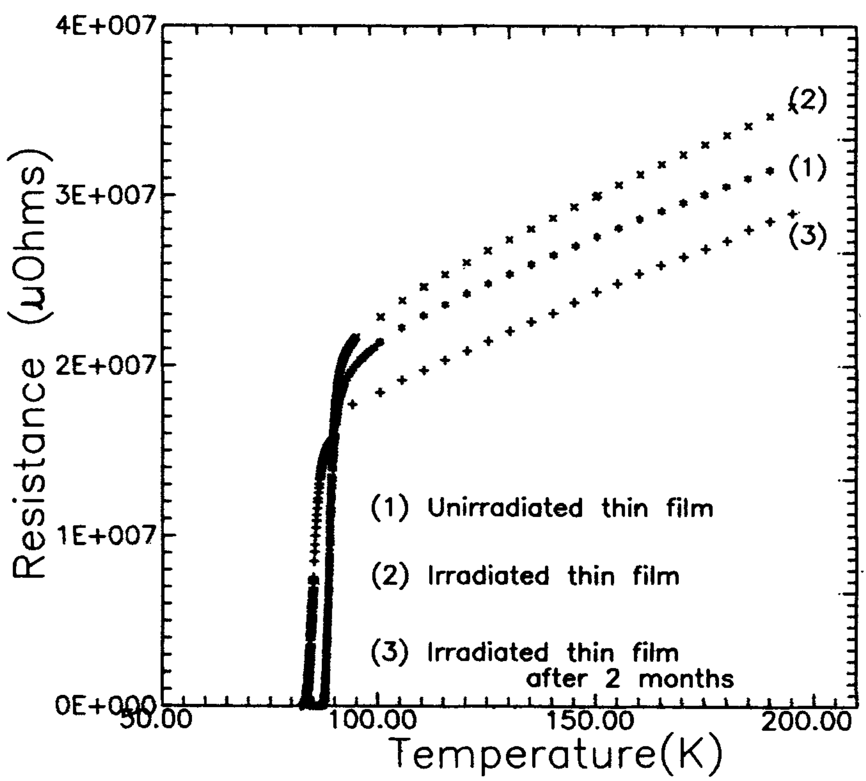

Figure 2. Variation of resistance as a function of temperature for unirradiated sample (1), irradiated sample (2) and irradiated sample after 2 months (3) in the temperature range 80 K to 200 K.

in flowing oxygen at 500°C and once again the measurements were done to see the effect of annealing.

3. Results

Thickness of the film used was approximately 3000 Å. TRIM calculations for the electronic energy loss as well as the range of oxygen ions in both $Y_1Ba_2Cu_3O_{7-x}$ and ZrO_2 substrate were made. For a thick sample of $Y_1Ba_2Cu_3O_{7-x}$ the range is 48 μm. Since the range is more than the thickness of the film used, all the oxygen ions passed through the film and there was no implantation of oxygen ions. One can expect a uniform damage to the sample. The electronic stopping power for 100 MeV oxygen ion beam in $YBa_2Cu_3O_{7-x}$ is 145·7 eV/A and nuclear stopping power is 0·0828 eV/A. These are nearly constant over the depth of the film. However the oxygen ions would have been stopped in ZrO_2 substrate in which the range was 300 μm.

The results of the electrical resistance measurements as a function of temperature is shown in figure 2. It is apparent that electrical resistance of the sample at room

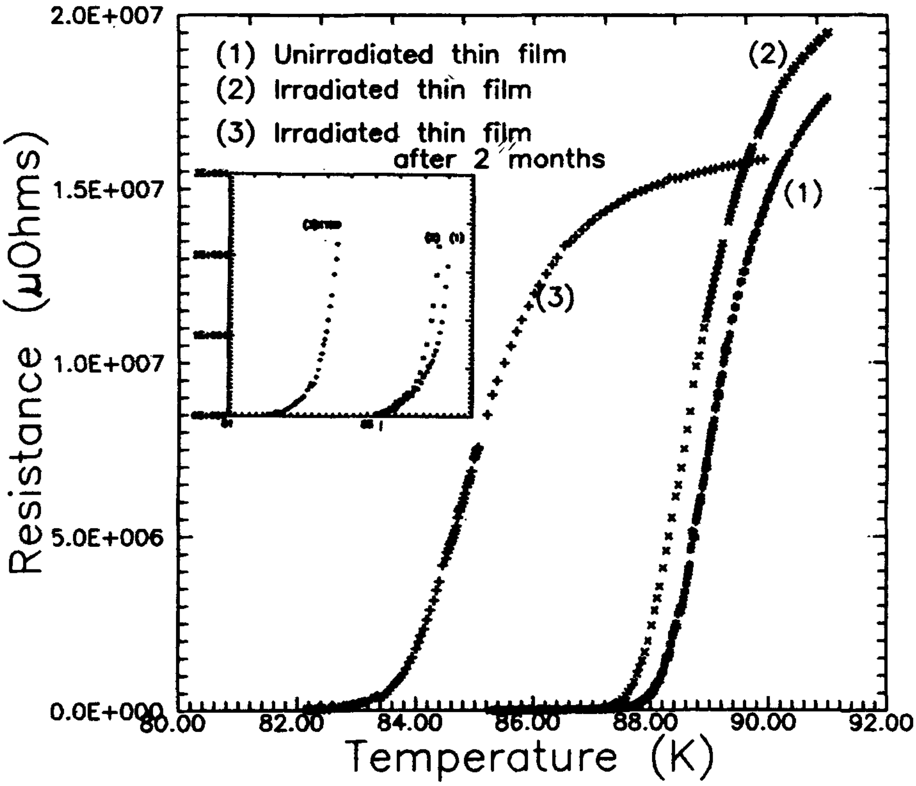

Figure 3. Variation of resistance as a function of temperature near the superconducting transition temperature for unirradiated sample (1), irradiated sample (2) and irradiated sample after 2 months (3).

temperature increased after irradiation. Also the slope (dR/dT) was more for the irradiated sample than for the unirradiated sample. Such a behaviour has been observed in samples in which oxygen content is decreased or other defects are induced. Figure 3 shows the resistance vs temperature in a very narrow region near the transition temperature. It is clear from this figure that the zero resistance transition temperature of the irradiated sample is slightly less than that of the unirradiated sample.

Data points were taken at every 50 mK interval. Then a plot of $\log(R)$ was made against $\log(T-T_{co})$ using an approximate value of T_{co} by judging at which temperature resistance went to zero in figure 3, the exact values of γ and T_{co} were determined by the following procedure. The plot of $\log(R)$ vs $\log(T-T_{co})$ shows a linear region in the temperature range of ~1°K above T_{co}. From this plot the slope γ of the linear region is estimated. This is a linear plot whose intercept should give T_{co}. This T_{co} was slightly different from the T_{co} which was used in the plot of $\log(R)$ vs $\log(T-T_{co})$. This value of T_{co} was again used to get a fresh value of γ. This procedure was repeated twice and we found that the value of T_{co} and γ converged very rapidly. Figures 4–6 show the plots of $\log(R)$ vs $\log(T-T_{co})$. The value of

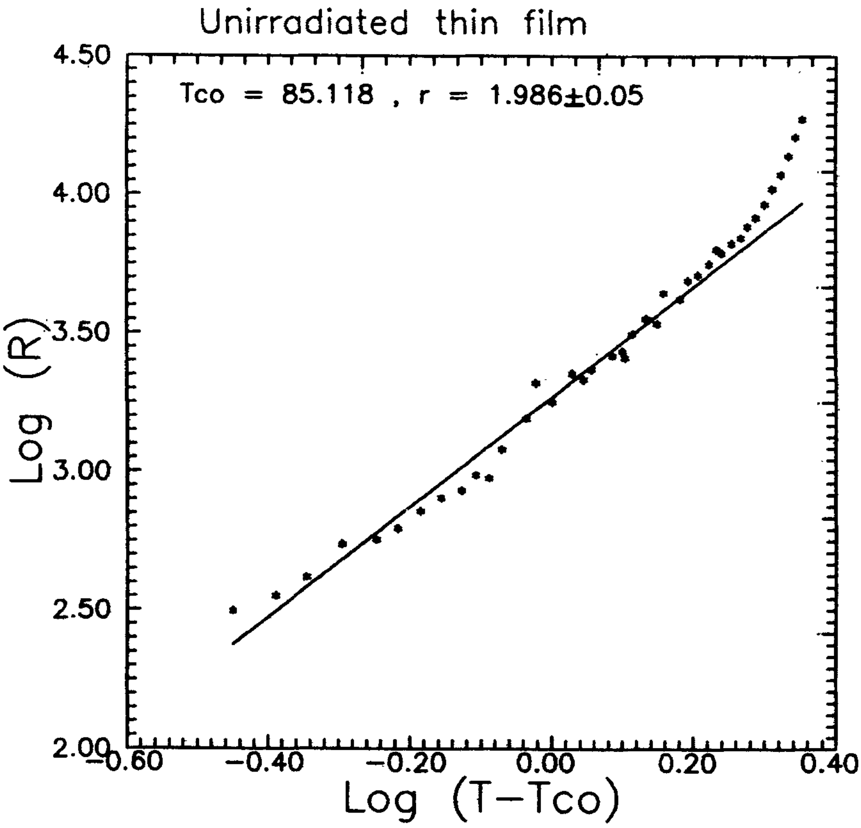

Figure 4. Plot of $\log(R)$ vs $\log(T-T_{co})$ for unirradiated sample.

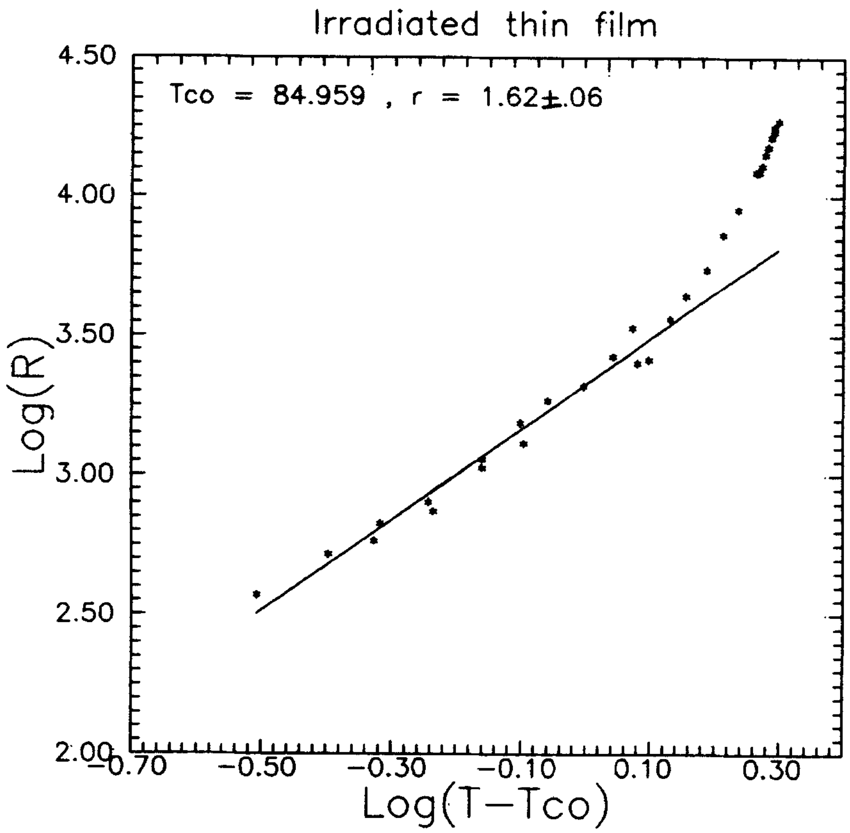

Figure 5. Plot of log(R) vs log(T–T_{co}) for irradiated sample.

conductivity exponent (γ) changes from its original value of $1{\cdot}99 \pm 0{\cdot}05$ to a value of $1{\cdot}62 \pm 0{\cdot}06$ upon irradiation. A value of ~2 or more for critical exponent has been found by other workers for good quality thin films showing a sharp transition (Nedellec *et al* 1989; Krishnan 1993).

The film was kept stored in a desiccator for about another two months and once again the whole experiment was repeated to find whether any change is there in the value of γ due to ageing. Curve 3 in figure 3 shows a variation of resistance as a function of temperature, it is obvious that T_{co} for this sample has come down to a value of ~82 K. An accurate analysis of resistance data gives a value of 81·88 K. The exponent γ changes from $1{\cdot}99 \pm 0{\cdot}05$ before irradiation to $1{\cdot}62 \pm 0{\cdot}06$ after irradiation and to $2{\cdot}39 \pm 0{\cdot}089$ after ageing. A look at the resistance curve shows that the resistance of the aged film at room temperature is lower than the resistance of the unirradiated film and the temperature coefficient dR/dT of the aged film is almost the same as the dR/dT for the unirradiated film. This indicates that the defects created by the heavy ion irradiation have atleast been partially annealed by ageing.

We tried to anneal the sample at 500°C in flowing oxygen to see whether the superconducting properties are recovered. But the sample deteriorated and lost the

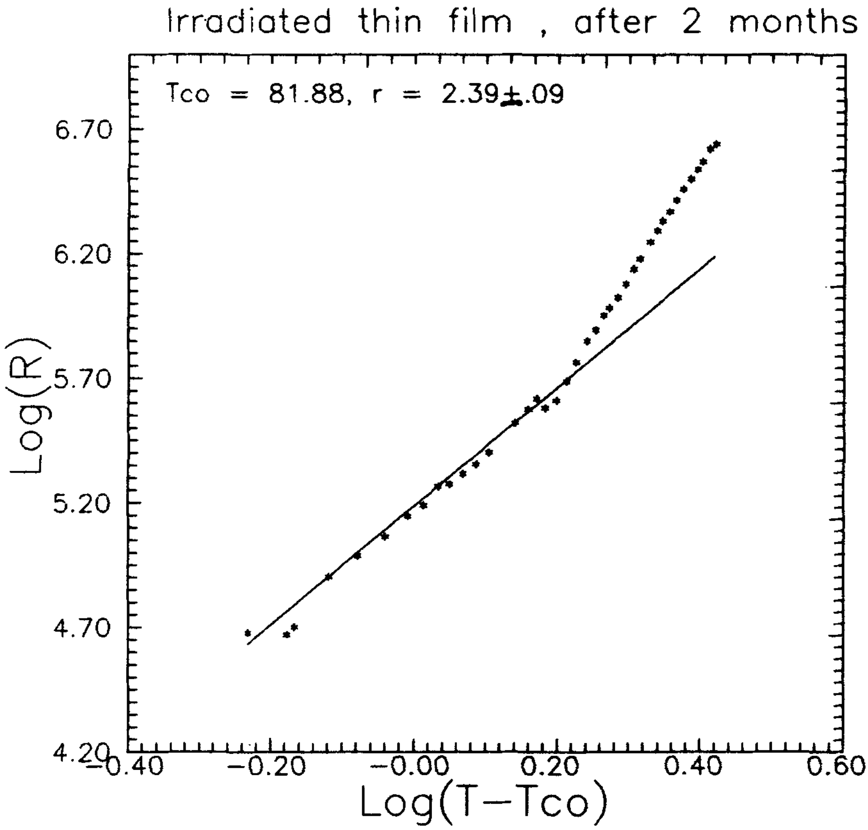

Figure 6. Plot of log(R) vs log(T–T_{co}) for irradiated sample after two months.

superconducting properties. The resistance at room temperature had increased by a factor of 8382 at room temperature and also the temperature variation was semiconductor-like and such a plot is shown in figure 7. In fact a plot of log(R) vs $T^{-1/4}$ (figure 8) in the temperature range between 300 K to 80 K showed a linear behaviour with a break at about 150 K. Such a linear behaviour is attributed to variable range hopping. However we are unable to explain as to why there is a break at 150 K.

4. Discussion

When the energy of the incident projectile is beyond a certain value it has been observed that the electronic loss can also produce defects and atomic displacements which cause a change in the electrical properties of the material. We used oxygen ions in the hope that these ions will produce such displacements preferentially at the oxygen sites in the superconductor especially at the grain boundaries. The increase in electrical resistance and slope dR/dT certainly arises due to such defects.

However it was surprising that T_{co} value did not come down substantially on irradiation. The observed effect of ageing on the resistance and dR/dT is to be expected. However the substantial reduction in T_{co} indicates that the strength of the weak links has been reduced probably because the oxygen displaced at the grain boundary has slowly diffused out. However the annealing procedure we followed resulted in further increase in deterioration of weak links and also possibly of the grains.

What was surprising is the large change in the value of critical exponent γ from $1{\cdot}99 \pm 0{\cdot}05$ to $1{\cdot}62 \pm 0{\cdot}06$ upon irradiation to $2{\cdot}39 \pm 0{\cdot}09$ on ageing. If the analogy of the tunneling Hamiltonian with 3D XY Ferromagnet Hamiltonian is fully valid then such a large variation in the value of critical exponent would not be expected in the light of some renormalization group calculations and the experimental work of Kaul (1988). Heisenberg model (Lubensky 1975; Grinstein and Luther 1976; Khmelnitzki 1976; Jug 1983; Weinrib and Halperin 1983) including both bond and site disorder showed that the critical exponents remain unchanged by short range disorder as long as the specific heat critical exponent α is negative. On the other hand, unconventional renormalization treatment predicts that the value of the critical exponent will depend on frozen disorder even when the specific heat exponent is less than zero. However this matter requires further detailed study.

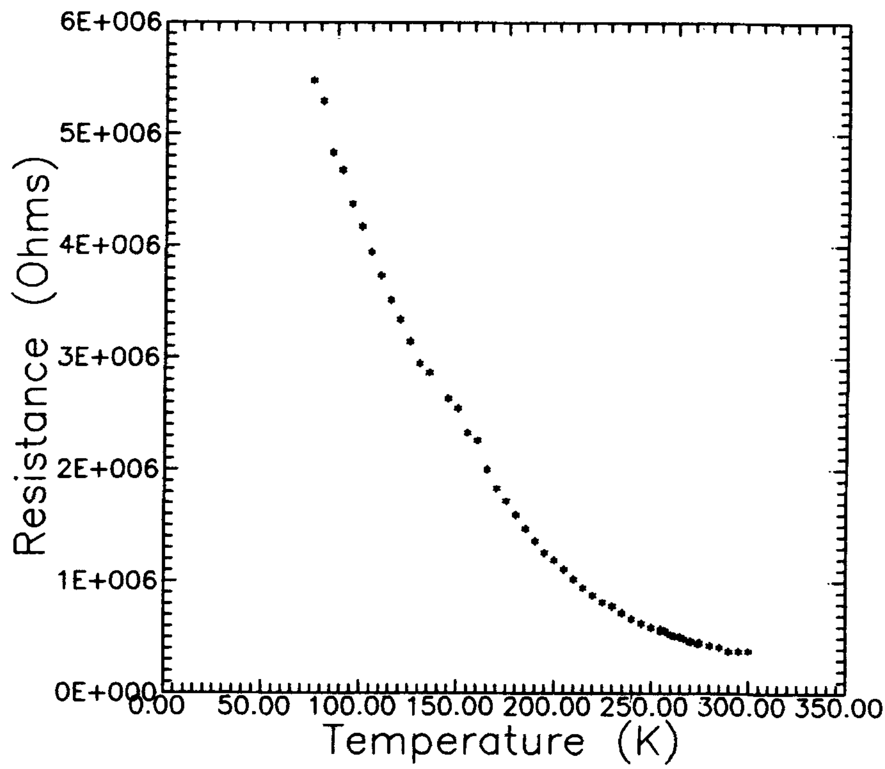

Figure 7. Plot of R vs T for the sample after annealing in flowing oxygen at 500°C for 30 min, after 70 days.

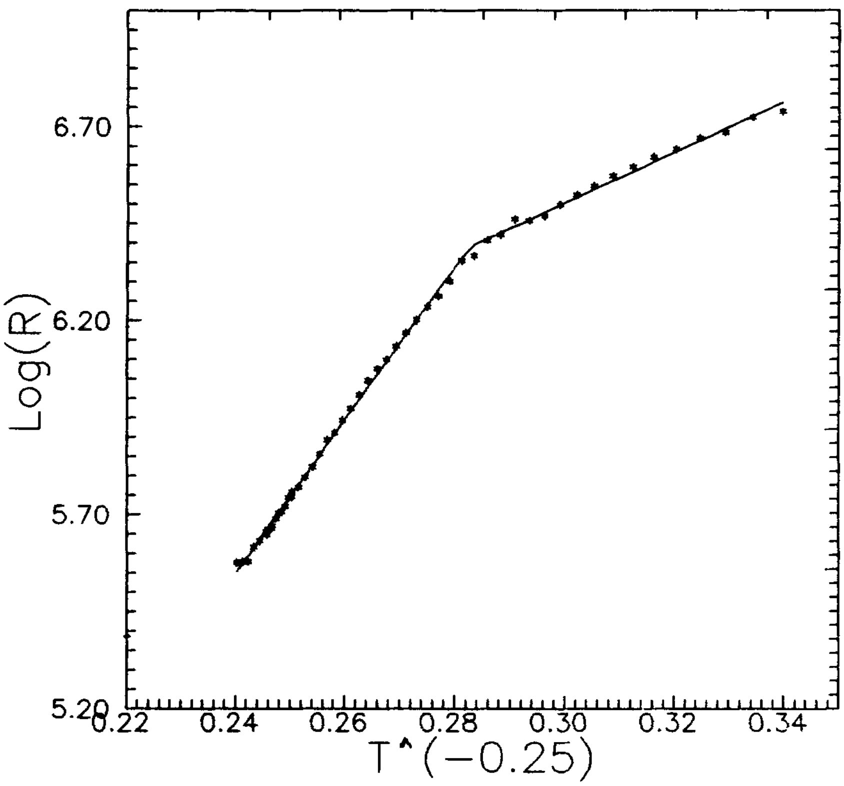

Figure 8. Plot of R vs T^(−1/4) for the semiconducting sample of YBCO film after annealing.

Acknowledgement

The authors of IUC-DAEF are thankful to Prof. S Lal for his constant encouragement right from the beginning of this work.

References

Ausloos M and Laurent Ch 1988 *Phys. Rev.* **B37** 611
Deutscher G, Imry Y and Gunther L 1974 *Phys. Rev.* **B10** 4598
Freitas P P, Tsuei C C and Plaskett T S 1987 *Phys. Rev.* **B36** 833
Grinstein G and Luther A 1976 *Phys. Rev.* **B13** 1329
Hagen S J, Wang Z Z and Ong N P 1988 *Phys. Rev.* **B38** 7137
Jug G 1983 *Phys. Rev.* **B27** 609
Kanjilal D, Chopra S, Narayanan M M, Iyer S I, Jha V, Joshi R and Datta S K 1993 *Nucl. Instr. Meth.* **A328** 97
Kaul S N 1988 *Phys. Rev.* **B38** 9178
Khmelnitzki D E 1976 *Sov. Phys. JETP* **41** 981
Krishnan H, Sankaranarayanan V and Srinivasan R 1992 *Physica* **C194** 163
Krishnan H, 1993 *Paraconductivity and paracoherence studies on some high temperature superconductors*, Ph. D. Thesis, Indian Institute of Technology, Madras
Lebeau C, Raboutou A, Peyral P and Rosenblatt J 1988 *Physica* **B152** 100

Le Guillou J C and Zinn-Justin J 1977 *Phys. Rev. Lett.* **39** 95

Lubensky T C 1975 *Phys. Rev.* **B11** 3573

Nedellec P, Lesueur J, Traverse A, Bernas H, Dumoulin L and Laibowitz R B 1989 *J. Less Common Metals* **151** 443

Pureur P, Schaf J, Gusmao M A and Kunzler J V 1990 *Progress in high temperature superconductivity* (ed.) R Nicholsky (Singapore: World Scientific) **25** 236

Rosenblatt J, Peyral P, Raboutou A and Lebeau C 1988 *Physica* **B152** 95

Rosenblatt J, Raboutou A, Peyral P and Lebeau C 1990 *Rev. Phys. Appl.* **25** 73

Soret J C and Ammor L 1991 *Physica* **C177** 45

Srinivasan R, Krishnan H, Sankaranarayanan V, Gopalakrishnan I K, Sastry P V P S S, Yakhmi J V and Iyer R M 1989 *Physica* **C159** 797

Veira J A, Maza J and Vidal Felix 1988 *Phys. Lett.* **A131** 310

Weinrib A and Halperin B I 1983 *Phys. Rev.* **B27** 413