Thermoelectric power of YBCO superconducting thin films

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ABSTRACT. Measurements of electrical resistance and thermoelectric power of sputtered YBa₂-Cu₃O_{7- δ} superconducting thin films have been carried out. The thermopower (S) is positive in the measured interval of temperature (77 K-300 K), showing a non-monotonic behavior with a minimum around 130 K. the thermoelectric data fit well to an expression of the form S = AT + B/Tin the range 130 K to 300 K. On the other hand, the relationship found for the excess thermopower and the excess conductivity due to superconducting fluctuations near T_c , suggests that they have a common origin which might be of electrical nature.

RESUMEN. Presentamos mediciones de potencia termoeléctrica (TEP) y resistividad eléctrica en películas delgadas de YBa₂Cu₃O_{7- δ}. La potencia termoeléctrica es positiva en todo el intervalo de medición (77 K-330 K), mostrando un comportamiento no monotónico con un mínimo alrededor de 130 K. Los datos de la TEP se ajustan bien a una relación de la forma S = AT + B/T en el intervalo de 130 K-300 K. Además, la relación lineal encontrada entre el exceso de TEP y el exceso de conductividad eléctrica generada por fluctuaciones superconductoras cerca de la T_c sugiere que ellas tienen un origen común, el cual puede ser de naturaleza eléctrica.

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1. INTRODUCTION

The thermoelectric power (S) is one of the most fundamental properties of condensed matter. S is a powerful probe to study the electronic properties of conductors, such as sign and concentration of charge carriers and carrier-phonon coupling; besides, the magnitude of S at room temperature is often a good guide to infer the hole concentration in the CuO₂ planes in superconductor cuprates [1,2]. Several works on the thermopower of Y-Ba-Cu-O compounds have been reported before [3,8], and it is generally accepted that for sintered polycrystallline YBa₂Cu₃O_{7- δ} (hereafter referred as 123), the sign and magnitude of the thermoelectric power are linked to particular doping levels; moreover, S displays a characteristic peak close to T_c and a subsequent drop to zero at T_c . Some interpretations of the temperature dependence of S have been also proposed: it has been suggested that this is a consequence of both phonon-drag and diffusion effects [3, 6, 7]; one interpretation argue that the diffusion term of S is enhanced by the effect of an anomalously large electron-phonon coupling which introduces an 1/T dependence [8]. Other authors, however, have attributed the precursor peak near Tc to a mechanism of electrical nature due to superconducting fluctuations [4].

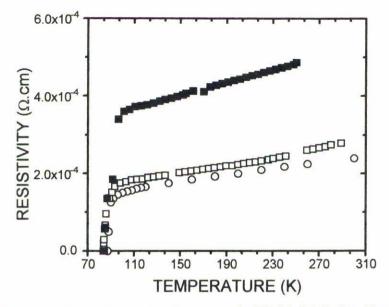


FIGURE 1. Resistivity ρ vs. temperature for YBCO/MgO thin films.

We present here measurements of resistivity and thermoelectric power carried out in superconducting $YBa_2Cu_3O_{7-\delta}$ thin films and intend to have some insight into the carrier scattering processes and the possible nature of the precursor peak.

2. EXPERIMENTAL

Nearly stoichiometric 123 superconducting thin films of ~ 0.8 μ m thick, were deposited on MgO(100) single crystals kept at ~ 500°C, by D.C. sputtering at 0.4 mtorr argon pressure. Thereafter the films were annealed at around 800°C during two hours in oxygen flow. The sputtered target of 50 mm diameter has the 123 composition and was made by solid state reaction using power of high purity. The electrical resistivity was measured with the standard four probe method and the S data were obtained using the differential technique with a temperature gradient < 1 K/cm across the sample [9].

3. Results and discussions

Typical x-ray diffraction patterns of the sperconducting thin films showed predominantly the peaks corresponding to the YBa₂Cu₃O_{7- δ} orthorhombic perovskite structure with a light preferred orientation along the c-axis, and also other phases such as YBa₂Cu₃O₆, although in lower concentrations, between 10% and 15%. Results of resistivity and thermopower as a function of the temperature for the three best samples are presented in Figs. 1 and 2.

The extrapolated residual resistivity lies between 0.11 m Ω cm and 0.35 m Ω cm. Typical transition widths were around 4 K and all samples showed predominance of the YBa₂Cu₃O_{7- δ} orthorrombic perovskite structure (80%).

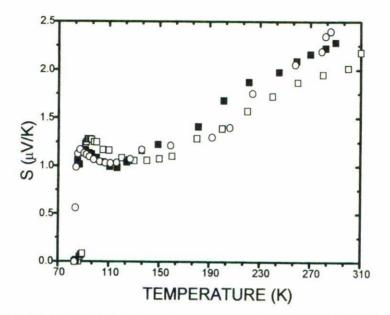


FIGURE 2. Thermoelectric power S vs. temperature for YBCO/MgO thin films.

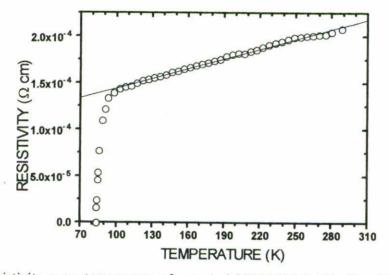


FIGURE 3. Resistivity ρ vs. temperature of a typical YBCO/MgO thin film. The straight-line linearly extrapolates to zero temperature the resistivity of the sample in the normal state.

Figure 3 shows the temperature dependence of the resistivity for a typical film. Transition temperatures $T_c \sim 90$ K and widths $\Delta T \sim 4$ K, were determined from the maximum of the derivative of the resistance vs. temperature curve. The extrapolated residual resistivity was $\rho_{\rm res} \sim 0.11 \, {\rm m\Omega \, cm}$, a value close to zero indicates the high quality of samples. The corresponding temperature dependence of S is displayed in Fig. 4. This behavior of S is in agreement with that found by other authors in bulk samples [3,10] and is qualitatively different from that of Bi and Tl based compounds, which show negative slopes [11,12]. In

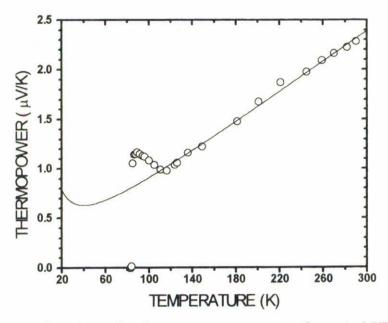


FIGURE 4. Corresponding thermoelectric power vs. temperature of a typical YBCO/MgO thin film. The solid line is a plot of S = AT + B/T.

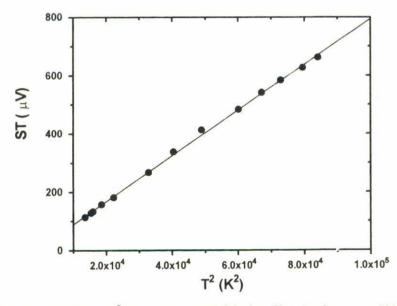


FIGURE 5. Plot of ST vs. T^2 for a typical YBCO thin film, in the range 130 K-300 K.

the present case S is positive in the interval 77 K-300 K with a magnitude of ~ 3 μ V/K at room temperature and with a positive slope ~ 0.0075 μ V/K². Below 130 K S rises up exhibiting an enhancement, in the characteristic rounding off interval of the resistivity, associated with thermodynamic fluctuations before falling to zero at T_c [13–15]. For all samples investigated, having $\rho_{\rm res}$ ranging from 01. m Ω cm to 0.3 m Ω cm, S is found to be positive and its magnitude at room temperature remains small, ranging from 2.5 μ V/K

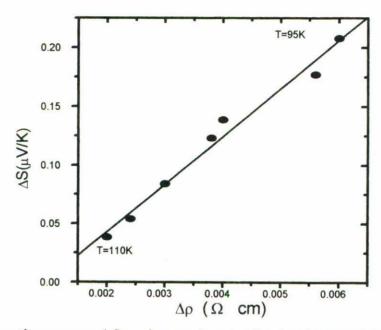


FIGURE 6. Excess thermopower ΔS vs. decrease in resistivity $\Delta \rho$ of a typical YBCO/MgO thin film.

to 4 μ V/K respectively. As ρ_{res} increases both the magnitude and the positive slope of S increase lightly, but no change in the sign of S is observed. Changes in the sign of S in bulk and single crystals samples have been associated with negative and positive contributions from the CuO chains and the CuO₂ planes respectively [2, 16].

Additionally, the temperature dependence of S between ~ 130 K and 300 K fits well to an expression of the form S = AT + B/T as displayed in Fig. 5. The linear relationship indicates the presence of two contributions: one proportional to T and due to the metallic diffusion part, in a temperature region much above T_c , and the other one proportional to T^{-1} , which has been attributed to different mechanisms [4,8,17].

In order to explore the possible origin of the excess thermopower, we have plotted the excess thermopower ΔS as a function of the decrease in resistivity $\Delta \rho$ due to superconducting fluctuations, as displayed in Fig. 6. The excess thermopower ΔS is determined as the difference of the measured S(T) with the extrapolated linear behavior from high temperatures $T \gg T_c$ (see Fig. 4). The decrement in resistivity is calculated as $\Delta \rho = \rho_n(T) - \rho(T)$, where $\rho(T)$ is the measured resistivity and $\rho_n(T)$ is the extrapolated one from the straight-line trend of the normal region (see Fig. 3).

Using the Mott's expression valid for conductivity in a metallic band, ΔS can be approximated to

$$\Delta S = aT\Delta\rho \frac{\partial}{\partial E} \left[\frac{1}{\rho_0}\right]\Big|_{E_F},$$

where it was assumed that the fluctuation part of the resistivity $\Delta \rho \ll \rho_0$.

Thus, the excess thermopower at a given temperature varies linearly with the decrease in resistivity $\Delta \rho$ due to superconducting fluctuations. In consequence, this linear relation suggests that the S component proportional to T^{-1} , which contributes mainly at the low temperature range, may arise from a mechanism of electrical nature due to thermodynamic fluctuations. The same behavior was found for all samples examined, where $\rho_{\rm res}$ ranged from 0.1 m Ω cm to 0.3 m Ω cm.

4. CONCLUSION

In summary, the results of the temperature dependence of S in the interval 130 K-300 K show the existence of two terms: the first one proportional to T corresponding to the normal carrier diffusion for $T \gg T_c$, and the second proportional to 1/T. The linear relationship found for ΔS and $\Delta \rho$ due to superconducting fluctuations, in the region close to $T \ge T_c$, indicates that they have a common origin, which suggests that the term proportional to T^{-1} might be of electrical nature. These results obtained for bulk and good quality YBCO thin films are similar to that found by us in the BSCCO system [11] and by other authors in YBCO bulk samples [4], which could be taken as an indication of a more general behavior.

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