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LETTER TO THE EDITOR

The thermoelectric power of pure and oxygenated $Bi_2Sr_2CaCu_2O_{8+y}$ systems: the role of BiO layers

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Abstract. The thermoelectric power of optimized ($T_c = 90$ K) and oxygenated ($T_c = 70$ K) samples of the $Bi_2Sr_2CaCu_2O_{8+y}$ system has been measured, and analysed using a two-band model. The analysis shows that intercalation of oxygen in Bi–O layers plays an important role in determining the thermopower of the system. Scanning tunnelling spectroscopy of optimized and oxygenated samples provides qualitative support to the model.

In the superconducting system $Bi_2Sr_2CaCu_2O_{8+y}$ (Bi-2212) there are strong indications from band-structure calculations [1], scanning tunnelling spectroscopy [2] (STS), and from some studies on angle-resolved photoemission spectroscopy [3] that BiO layers play an important role in controlling the superconducting critical temperature of this high-T_c system. These are redox layers which play a role analogous to that of Cu–O chains in 1–2–3 systems [4]. Because of this, there is a need for further clarification of the relevance of the BiO layers in the superconductivity of the Bi-2212 system. A study of the behaviour of various other properties of the Bi-2212 system seems promising. In this work we have studied the thermoelectric power of the Bi-2212 system. We have considered two samples of this system: (i) an optimally doped or pure sample (sample a); and (ii) an oxygenated sample in which the Bi–O layers are intercalated with oxygen (sample b).

In the literature, a variety of approaches have been followed to explain the thermoelectric power [4]. In the present system where the BiO layers are believed to be important, it is advantageous to consider a two-band model. Here the BiO band is considered in conjunction with the CuO_2 band resulting from hybridized Cu $3d_{x^2-y^2}$ and O $2p_{x,y}$ states [5]. In particular, we have used the two-band model proposed recently by Xin *et al* [6] for a Tl-based superconductor. In this model, when applied to the Bi-2212 system, the BiO layers are effectively considered in the form of semiconducting n-type layers. This approach has been found to be quite successful for describing the thermoelectric power of various Tl- and Bi-based systems [6, 7].

Samples were prepared by solid-state reaction of appropriate mixtures of Bi_2O_3, $CaCu_3$, $SrCo_3$ and CuO, each of 4N purity. The powders were mixed, pulverized and calcined at 820 °C, 830 °C and 840 °C successively for 12 h each, and this was followed by furnace cooling. In the final step, the samples were partially melted at 930 °C and then subjected to sustained heating at 860 °C for 12 h before being finally quenched to room temperature. The sample of this batch is called sample a. Some of the samples of type a were oxygenated

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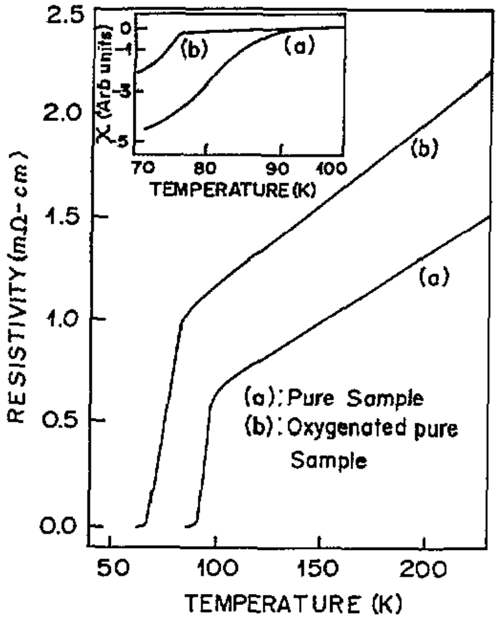

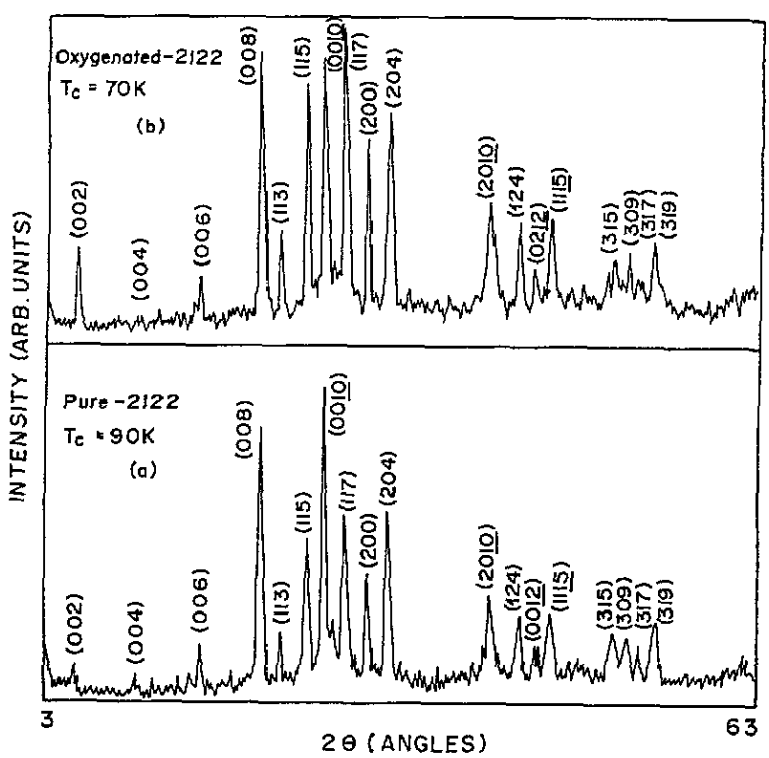

Figure 1. Top panel: the resistivity of samples a and b of the Bi-2212 system; inset: the AC susceptibility of samples a and b. Bottom panel: XRD patterns for sample a and b (sample b above), taken at room temperature.

at 860 °C for 12 h and then furnace cooled to room temperature over a span of 6 h. The sample of this batch is called sample b.

Four-probe DC resistivity data for rectangular slab samples were obtained in the temperature range 50 K to 230 K in a liquid helium bath cryostat using a carbon glass resistor temperature sensor. The entire measurement system, consisting of a nanovoltmeter (Keithley 181), a constant current source (Keithley 224), a temperature controller and an indicator (Lakeshore Model 120), was hooked to a HP 216 system controller for automatic data acquisition and control. Magnetic susceptibility measurements were carried out with a fully computer-controlled AC susceptometer (Lakeshore 7000). The samples were characterized for their phase identification, homogeneity and the lattice parameters through x-ray diffraction using a Siemens D-500 diffractometer with Cu Kα radiation. Thermoelectric power measurements were carried out using a closed-cycle refrigerator [7].

The resistivity measurements for these samples show transition temperatures $T_c(R = 0)$ of 90 K for sample a and 70 K for sample b (figure 1 (upper panel)). For the pure Bi-2212 system Cho and co-workers also observed a T_c of about 90 K. This demonstrates the consistency of our results with that of other workers. From XRD patterns a single phase for the two samples is confirmed (figure 1 (lower panel)), with lattice parameters of $a = 5.43$ Å, $b = 5.41$ Å and $c = 30.79$ Å for sample a and $a = 5.46$ Å, $b = 5.4$ Å and $c = 30.61$ Å for sample b, which are in agreement with other reports [2]. AC susceptibility *measurements show a diamagnetic onset a little above 90 K for sample a and at 76 K for* sample b (inset of figure 1 (upper panel), in accordance with the resistive transitions. The results of thermoelectric power (S) measurements for the two samples are depicted in the form of S versus T curves in figure 2. The behaviour of S at the superconducting transition corroborates the $T_c(R = 0)$ values mentioned above.

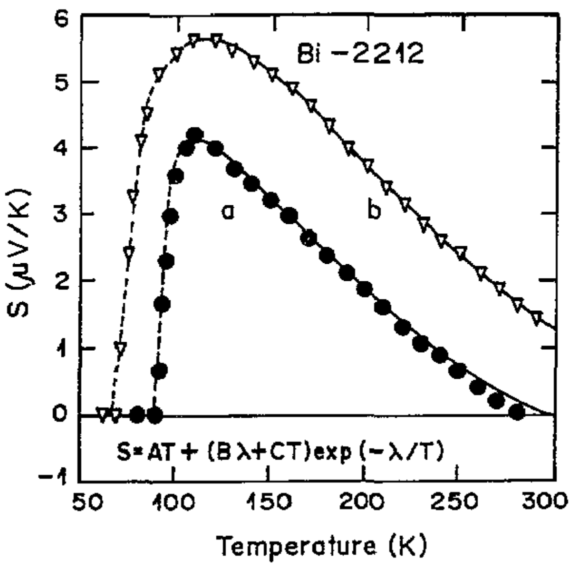

Figure 2. The thermoelectric power of samples a and b of the Bi-2212 system.

According to Xin *et al* [6] and Awana *et al* [7], for Tl-2223 and Bi-2223 systems, respectively, in the two-band model, the cumulative thermoelectric power is given by

$$S = AT + [B\lambda + CT]\exp(-\lambda/T)]. \qquad (1)$$

Here A, B, C and λ are the material's parameters. In the above equation, the first part represents the role of p-type conduction (holes) in Cu–O planes, while the second term represents the role of n-type conduction (electrons) in the semiconducting Bi–O layers.

We have applied the above two-band picture (equation (1)) to the thermoelectric power data for samples a and b. The solid lines in figure 2 are computational fits of (1) to the experimental data. The fitting parameters A, B, C and λ are found to be $A = 0.0708$ μV K^{-2}, $B = 0.1479$ μV K^{-2}, $C = -0.04231$ μV K^{-2} and $\lambda = 280.31$ K for sample a, and $A = 0.0758$ μV K^{-2}, $B = -0.1579$ μV K^{-2}, $C = -0.04331$ μV K^{-2} and $\lambda = 340.37$ K for sample b.

We use the above two values of λ to estimate the energy gap in the semiconductor-like band structure of the Bi–O layers. The semiconductor energy gap E_g in the intrinsic semiconductor is usually given by $E_g = 2E_c$. According to the relation $\lambda = E_c/k_B$ (where k_B is the Boltzmann constant), for sample a, with $\lambda = 280.31$ K, E_g is found to be 0.046 eV, and for sample b, with $\lambda = 340.37$ K, E_g is found to be 0.056 eV. Thus the semiconductor gap E_g is larger in the case of the oxygenated sample, sample b, than for the pure sample, sample a, which is in qualitative accord with the STS observations on the Bi–O layers of the two samples published earlier [2], confirming an increase in the semiconducting energy gap of Bi–O layers after oxygenation.

In conclusion, we have observed the thermoelectric power of pure and oxygenated Bi-2212. The data have been analysed using a two-band model to throw light on the role of the BiO layers in the Bi-2212 system. The thermoelectric power, STS studies [2] and resistivity data all seem to indicate that BiO layers in Bi-2212 are semimetallic and play an important role. The semimetallic nature of BiO layers, which is enhanced by oxygenation, plays an important role in the superconducting behaviour of pure and oxygenated samples [2].

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