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1989 J. Phys.: Condens. Matter 1 1785

(http://iopscience.iop.org/0953-8984/1/9/021)

J. Phys.: Condens. Matter **1** (1989) 1785–1787. Printed in the UK

LETTER TO THE EDITOR

The thermoelectric power of the system $SmBa_{2-x}Sr_xCu_3O_{7-\delta}$

L D Firth
Physics Department, Paisley College of Technology, Paisley PA1 2BE, UK

Received 12 December 1988

Abstract. Thermoelectric power in the range 77–310 K has been measured for compounds $SmBa_{2-x}Sr_xCu_3O_{7-\delta}$ with $0 \leq x \leq 1$. Above the superconducting transition temperature, positive thermopowers were observed in all cases. Over most of the temperature range, the thermopower can be described by $S = A + B/T$ where A is (5.1 ± 0.3) $\mu V K^{-1}$, and B depends on the Sr content. A sudden increase in B at $x \simeq 0.6$ is associated with a structural transition from orthorhombic to tetragonal. For $x \leq 0.2$ a departure from the formula at lower temperatures may indicate a positive phonon drag thermopower.

The compounds $MBa_{2-x}Sr_xCu_3O_{7-\delta}$ (M = Y, Gd, Sm) have been shown (Currie 1988, Currie and Forrest 1988) to be orthorhombic at room temperature up to $x \simeq 0.6$. While the substitution of Sr for Ba depresses the temperature T_c of the onset of superconductivity, the rate of depression $-dT_c/dx$ is quite low ($\simeq 10$ K), so the essential features of the superconducting mechanism are not greatly altered. However, the phonon spectrum will be distorted, with an increase of characteristic temperature, and the scattering of both electrons and phonons will be increased. There will also be changes in the electronic band structure associated with small changes in the lattice parameters. Thermoelectric power (TEP) is rather sensitive to these changes. Interpretation is far from simple, especially in a polycrystalline sample. However measurements on single crystals of $YBa_{2-x}Sr_xCu_3O_{7-\delta}$ suggest (Crommie *et al* 1988) that both the resistivity and the TEP of the polycrystalline material are controlled mainly by the properties in the *a–b* plane.

The method of sample preparation has already been described (Currie and Forrest 1988). The powdered material was pelletised, producing 8 mm diameter cylinders. The sample lengths were between 9 and 13 mm. The pellets were fired in flowing oxygen at 920 °C for 18 h. Copper blocks were fixed to the flat faces using silver conducting paint. One block was maintained at liquid nitrogen temperature (or at the ice point for measurements above this temperature) while the other was heated. The temperature difference across the specimen was measured using thermocouples embedded in the blocks, and the Seebeck EMF using copper wires. Calibration of the blocks and wires against lead enabled absolute TEP to be determined. Readings were taken every degree but a temperature difference of 3 K was used for the purpose of calculating the TEP. All the measurements were completed during a ten-day period, with the specimens taken in random order so as to minimise any systematic aging effects such as progressive oxygen loss.

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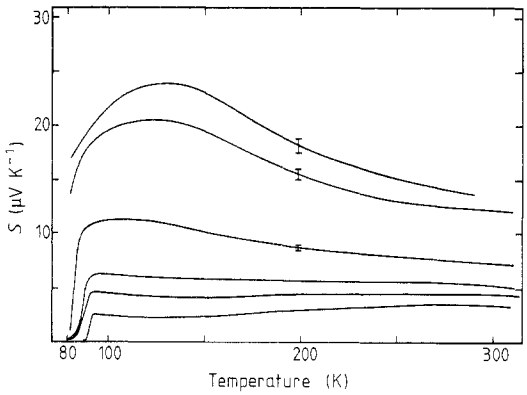

Figure 1. The thermoelectric power of $SmBa_{2-x}Sr_xCu_3O_{7-\delta}$ with $x = 0, 0.2, 0.4, 0.6, 0.8$ and 1.0. The TEP increases monotonically with increasing x. The full curves are fits drawn by eye to some 250 computer-plotted points. The error bars show typical measurement uncertainty, where significant.

Figure 1 summarises the measurements. Below 100 K the rapid drop towards zero TEP is prominent, although for $x = 0.8$ and $x = 1.0$ zero TEP was not observed in the range of measurement. There is good agreement between these results and those of resistivity measurements (Currie 1988) as to the dependence of T_c on Sr content.

The tendency of the TEPs of the different samples to converge at higher temperatures suggests plots against inverse temperature (figure 2). These show linearity from about 270 K down to about 130 K, and for $x = 0.4$ down almost to the superconducting transition. There appears to be a common intercept for these lines at $(5.1 \pm 0.3)\,\mu V\,K^{-1}$. For $x = 0$, 0.2 and 0.4 the intercept is $(5.06 \pm 0.06)\,\mu V\,K^{-1}$. The gradients depend on Sr concentration, and change rapidly with x in the region of $x = 0.6$. It is around this concentration that the room-temperature structure becomes orthorhombic. It is clear that the transition from the tetragonal structure will affect the electronic bands for states in the a–b plane, and so it is reasonable to suppose that the gradients are directly affected by the band structure.

In the free-hole model of a non-degenerate p-type semiconductor, the diffusion TEP is given (Smith 1978) by

$$S = (k/e)[\tfrac{5}{2} + (E/\tau)\,\mathrm{d}\tau/\mathrm{d}E + (E_F - E_V)/kT].$$

The gradient $\mathrm{d}S/\mathrm{d}(1/T)$ is simply the height of the Fermi energy above the top of the valence band, expressed in volts. Band-structure calculations (Krakauer *et al* 1988) for perovskite superconducting compounds show sharp and narrow peaks in the density of states near E_F and thus the position of E_F is rather sensitive to small changes in the band structure. In terms of the model, the experimental results suggest a rise in E_F with substitution of Sr for Ba.

In terms of the same model, the constant intercept signifies that the energy dependence of the hole relaxation time τ is unchanged by the substitution. This is surprising since the balance of phonon and impurity scattering is certainly altering as x increases, and these two forms of scattering normally have very different energy dependences. However if the TEP is dominated by holes in the a–b plane, and these are strongly localised in the copper and oxygen atoms, then scattering by impurity atoms outside this plane of atoms could be relatively weak.

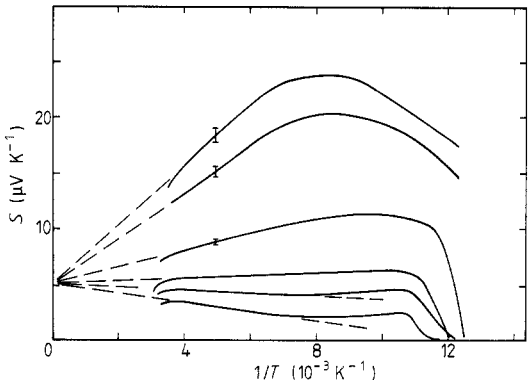

Figure 2. The data of figure 1, plotted against inverse temperature.

Above 270 K there are small departures from linearity; these could be caused by an upward shift in the Fermi energy and the consequent introduction of a small n-type or electron contribution to the TEP. However, it should be emphasised that the single-band model used in this discussion is quite inadequate for explaining the magnitudes of the observed TEPs; it would require $E_F - E_V < kT$ which conflicts with the requirement of no degeneracy. A two-band model that allows for a substantial TEP contribution from an electron-like band might explain the observed magnitudes and also the negative gradient for $x = 0$ and $x = 0.2$.

The transition to the superconducting state causes a departure from linearity at low temperatures; for $x > 0.6$ this process is broadened out, eroding the linear region. For $x = 0$ and $x = 0.2$ a different type of departure is apparent, consistent with the high-temperature tail of a phonon drag contribution to the TEP. This is normally observed at around $\theta_D/5$ for a pure conductor but is suppressed by the presence of substitutional impurity atoms of very different mass, such as Sr for Ba in this case. Such substitution sites scatter phonons strongly and so reduce the probability of momentum transfer from phonons to electrons.

The author is grateful to D B Currie and L Kirkup for assistance with specimen preparation.

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