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Superconducting fluctuations in the thermoelectric power and resistivity of Bi-based polycrystalline cuprates

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Abstract. We report a study of fluctuation effects in the resistance, R , and the thermoelectric power, S , of melt-processed BiSrCaCuO (2212) and sintered BiPbSrCaCuO (2223) polycrystalline materials. Applying several theoretical models to the analysis of experimental results we extract a number of physical parameters for these systems: the Fermi energy, interlayer coupling strength and atomic Debye temperatures for Bi and Pb. The results are in good agreement with data obtained in other studies of similar materials.

1. Introduction and relevant background

Thermoelectricity in high- T_c materials has attracted significant research interest in the few years since their discovery in 1987. In particular, the Seebeck effect has been widely studied in polycrystalline samples [1-5], textured samples [6, 7] and single crystal specimens [8-12] of various cuprates. Several groups have been involved in comparative analysis of the influence of thermodynamic fluctuations on thermoelectric power, S , and conductivity, $\sigma = 1/\rho$, in these materials [13-15]. The effect of fluctuations on the latter transport property, however, has been investigated [16-20] far more extensively than has been done in thermopower studies. Frequently the analysis of the fluctuations in S and σ , measured in the zero-magnetic-field regime, does not extend beyond the determination of the interlayer coupling strength and the dimensionality crossover for the superconducting fluctuations.

In this paper we attempt to extend additionally the analysis of fluctuations in S and σ in order to estimate other important physical quantities: Fermi energy and Debye temperature, Ω_D .

We first outline the widely used procedure [13-21] for analysis of superconducting fluctuations above the fluctuation-corrected critical transition temperature, T_{c0} . In high- T_c materials this usually differs strongly from the temperature of the onset of zero resistance, T_c , (see Ausloos *et al* [16] for a brief discussion of different approaches to the analysis of fluctuation regime). The excess conductivity or paraconductivity, $\Delta\sigma$, is defined as

$$\Delta\sigma = \sigma - \sigma_N \quad (1)$$

where $\sigma = 1/\rho$ is the measured sample conductivity and $\sigma_N = 1/\rho_N$ is the normal state conductivity background which has to be determined by extrapolation from

temperatures above $\simeq 2T_c$. In the single layer model developed by Aslamazov and Larkin [22] (hereafter, AL), $\Delta\sigma$ is given by

$$\Delta\sigma = F\epsilon^{-\lambda} \quad (2)$$

where $\epsilon = (T - T_c)/T_c$, $F = e^2/(16\hbar d)$ and $\lambda = 1$ for the two-dimensional case; for three-dimensional fluctuations, $F = e^2/(32\hbar\xi_0)$ and $\lambda = 1/2$. Here d and ξ_0 stand for the effective layer thickness and coherence length at $T = 0$ K, respectively. Plotting, therefore, $(1/\Delta\sigma)^{1/\lambda}$ versus T one can estimate the value of T_c from the intercept of the extrapolated straight line with the temperature axis and also d or ξ_0 , depending on the dimensionality of the system [23], from the slope of this line.

The multi-layer superconducting structure considered by Lawrence and Doniach [24] (hereafter, LD) results in

$$\Delta\sigma = (G/\epsilon)(1 + \nu/\epsilon)^{-1/2} \quad (3)$$

where $G = e^2/(16\hbar d)$ and the interlayer coupling strength, ν , is given by $\nu = (2\xi_{0\perp}/d)^2$. $\xi_{0\perp}$ is the coherence length in the stacking direction of the multi-layer structure at $T = 0$ K. Equation (3) reduces to the three- and two-dimensional AL results for $\nu \gg 1$ and $\nu \ll 1$, respectively.

In the AL theory, the fluctuation dimensionality, D , is related to the critical exponent λ by

$$D = 2(2 - \lambda). \quad (4)$$

Here it should be noted that according to the LD model, even in the case of 2D thermodynamic fluctuations, the behaviour is expected to be 3D in nature for temperatures close enough to T_c . The LD estimate for the characteristic temperature interval, ΔT_{23} , that separates these two regimes is

$$\Delta T_{23} = T_c\nu. \quad (5)$$

One can, therefore, evaluate the interlayer coupling strength and, given the value of d , obtain an estimate for $\xi_{0\perp}$ (see equation (3) above).

Plotting $\ln(\Delta\sigma)$ as a function of $\ln(\epsilon)$ one can obtain λ and D from the experimental data. When λ is reasonably close to 1/2 or 1, T_c can be re-evaluated by plotting $(1/\Delta\sigma)^{1/\lambda}$ as a function of T in the appropriate temperature interval [17, 19]. Here the interested reader is referred to the extensive literature on paraconductivity for conventional and high- T_c materials (see Ausloos *et al* [16], Veira and Vidal [21] and Skocpol and Tinkham [25]).

Other approaches for the analysis of experimental data exist, including the Maki [26] model for the fluctuation regime in S , which has been applied by Howson *et al* [27]. For the purpose of this study, however, we rely on the AL and LD models as well as on the recent theoretical study by Varlamov and Livanov (VL) [28], who provided a satisfactory explanation for the unusual data of Howson *et al* [9, 27].

In the 2D case, the VL model [28] produces the following relation for the fluctuation-mediated correction to the conductivity, $\Delta\sigma$:

$$\Delta\sigma/\sigma_N = (1/E_F)(\hbar/\tau)B\epsilon^{-1} \quad (6)$$

where constant B should be $\simeq 0.20$ in order to produce results that are consistent with the single layer AL model [29]. $E_F = \pi \hbar^2 n d / m$, m is the effective electronic mass in the plane of the superconducting sheet of the thickness, d , and n is the carrier density of states at the Fermi level; the other quantities are as specified above.

Writing ΔS and ΔL for the fluctuation-induced parts of the Seebeck (S) and thermoelectric ($L = S\sigma$) coefficients, respectively, one obtains expressions similar to equation (1) above:

$$S = S_N + \Delta S \quad (7a)$$

$$L = L_N + \Delta L. \quad (7b)$$

Here again, the subscript N stands for the observed behaviour of these transport coefficients in the normal state. Using the VL model's expression for $\Delta L/L_N$

$$\Delta L/L_N = (1/E_F)(\hbar/\tau)A \ln(\Omega_D/T_{c0})\epsilon^{-1} \quad (8)$$

and the definitions given by equations (1), (7a) and (7b) with $\Delta L/L_N \equiv \Delta S/S_N + \Delta\sigma/\sigma_N + \Delta S\Delta\sigma/S_N\sigma_N$ one obtains

$$\Delta S/S_N = [(A/B) \ln(\Omega_D/T_{c0}) - 1]/[1 + \sigma_N/\Delta\sigma]. \quad (9)$$

The constant $A \simeq 5.28$. For high- T_c materials the scattering rate, $1/\tau$, is usually

$$1/\tau \simeq CTk_B/\hbar = C(1 + \epsilon)T_{c0}k_B/\hbar \quad (10)$$

where C is of the order of 1.4–2 (see [30] for a review of the common features of high- T_c materials). Combining equation (10) with equations (6) and (9) we get for the 2D case the following relations:

$$\sigma_N/\Delta\sigma = (E_F/k_B T_{c0})(1/BC)[\epsilon/(1 + \epsilon)] \quad (11)$$

$$S_N/\Delta S = [1 + \sigma_N/\Delta\sigma]/[(A/B) \ln(\Omega_D/T_{c0}) - 1]. \quad (12)$$

One observes that for $\epsilon \ll 1$ the asymptotic behaviour of $\sigma_N/\Delta\sigma$ and $S_N/\Delta S$ follows ϵ as it should for the 2D case of the AL, LD and VL models. Hence manipulation of the expressions provided by these models should, in principle, allow one to establish quantitative interrelations between E_F , d , n , τ , Ω_D and coupling strengths from the constructions suggested by equations (1)–(12).

We now turn to discussion of our experimental results.

2. Experimental results and discussion

We have investigated fluctuation effects in the thermoelectric power and resistivity of polycrystalline sintered [31] $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$ (2223, $T_c \simeq 109$ K) and melt-processed [32] $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_x$ (2212, $T_c \simeq 90$ K) materials. The specimens were cut into bar shapes with typical dimensions of $L_x \times L_y \times L_z = 12.0 \times 1.2 \times 0.15$ mm^3 for the 2223 sample and of $9.3 \times 2.0 \times 0.7$ mm^3 for the 2212 sample. $R(T)$ was measured by a standard four-probe technique. Electrical contacts to the sample were made with silver paste. In figure 1 we present the temperature dependence of $R(T)$

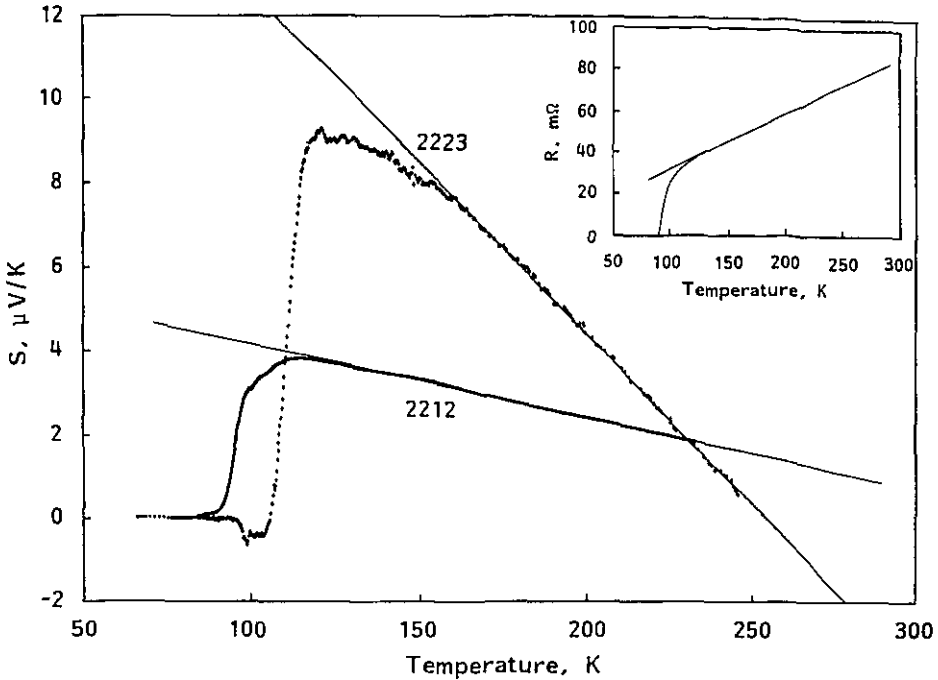


Figure 1. Temperature dependence of the thermoelectric power, $S = S(T)$; inset: the resistance of the 2212 samples. The straight lines are the extrapolation of the normal-state behaviour for these transport coefficients. In this and following figures the labels 2223 and 2212 stand for the data obtained from the respective phase of the Bi-based cuprates under study.

for the 2212 material [31] and thermoelectric power, $S = S(T)$, for both materials. The experimental procedure for Seebeck measurements is similar to the one used by Gridin *et al* [4, 5]. The straight lines drawn on this figure represent the normal-state behaviour of R and S found by extrapolation to $T = 0$ K of the data above 200 K. This gives us $R_N(T)$ and $S_N(T)$ for both materials. The deviation of the sample's R and S from $R_N(T)$ and $S_N(T)$ when the temperature approaches T_c from above are due to superconducting fluctuations in these transport coefficients. Using the effective mass approximation ($\rho_N \approx m/ne^2\tau$, where τ is substituted according to equation (10) above) for the normal resistance of a conductor of thickness d , and in-plane mass m , we write for the dimensionless slope η of the normalized R_N/R^* :

$$\eta = m_e C^* k_B / (R^* p n e^2 \hbar). \tag{13}$$

Here p is the effective value of the cross-sectional area to length ratio for our samples which for polycrystalline materials can differ significantly from its geometrical definition: $L_y L_z / L_x$, using the notation $R = \rho/p$. For both the 2223 and the 2212 samples the values of η thus defined were 3.85×10^{-3} . R^* is the resistance of the samples measured at 260 K; for our samples of 2223 BSCCO $R^* \simeq 130$ m Ω and for 2212 $R^* \simeq 70$ m Ω . The numerical factor C^* incorporates both the material and sample dependent quantities: C , which results from the substitution for \hbar/τ in terms of equation (10); the deviation of the in-plane mass, m , from the free-electron mass, m_e ; oversimplification due to the presumption of complete absence of

interlayer coupling (a purely 2D treatment); and the intrinsically 'average' behaviour of polycrystalline materials in terms of electronic transport. We use the empirically obtained values of C^* for both materials and apply in what follows ρ (studied material) = ρ (free-electron) $\times C^*$ and E_F (studied material) = E_F (free-electron) / C^* .

In order to find an estimate for the corresponding T_{ω} 's we first plot $\sigma^*/\Delta\sigma = 1/(R^*/R - R^*/R_N)$ as a function of temperature. This is presented in figure 2. From equation (2), the slopes,

$$\beta = 16\hbar d / (pR^*e^2T_{\omega}) \tag{14}$$

of the extrapolation of $\sigma^*/\Delta\sigma$ to zero are $\beta(2223) \simeq 0.72 \text{ K}^{-1}$ and $\beta(2212) \simeq 0.29 \text{ K}^{-1}$ for the 2223 and 2212 BSCCO samples, respectively. The corresponding values for T_{ω} thus found are $T_{\omega}(2223) = 108.7 \text{ K}$ and $T_{\omega}(2212) = 93.5 \text{ K}$. Taking the effective thickness, $d = \beta R^* p e^2 T_{\omega} / 16\hbar$, of the superconducting electronic layer to be half of the c -axis distance for the respective case, we find $d(2223) \simeq 19 \text{ \AA}$ and $d(2212) \simeq 15 \text{ \AA}$, since $c(2223) = 37.1 \text{ \AA}$ and $c(2212) = 30.9 \text{ \AA}$. The value of the ratio, p (cross-sectional area to length), used in equations (13) and (14) can be estimated now to yield 0.0012 cm for 2223 and 0.0051 cm for 2212 samples. Inserting $n(2223) = 1.54 \times 10^{21} \text{ cm}^{-3}$ and $n(2212) = 4.2 \times 10^{21} \text{ cm}^{-3}$ [33] together with the estimated values of p we obtain from equation (13) that $C^*(2223) \simeq 2.1$ and $C^*(2212) \simeq 12.5$.

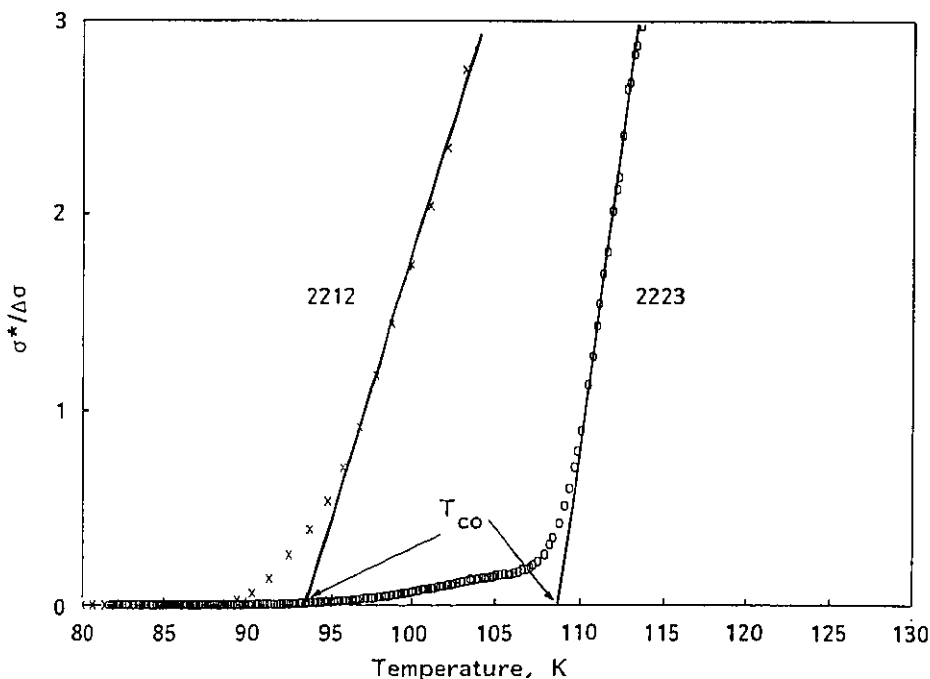


Figure 2. Temperature dependence of $\sigma^*/\Delta\sigma$, see text and equation (14). The straight lines with slopes β are the extrapolation of a 2D behaviour. The values of T_{ω} are, respectively, $93.5 \pm 0.5 \text{ K}$ and $108.7 \pm 0.4 \text{ K}$ for the 2212 and 2223 samples.

Using the values of d we now estimate the coherence length in the CuO layer stacking direction, ξ_{0L} . This is achieved by plotting $\ln(\Delta\sigma)$ versus $\ln(\epsilon)$ (see

equations (3)–(5) and their discussion for the interrelation between the interlayer coupling strength and $\xi_{0\perp}$). This construction is presented in figure 3. The corresponding temperature intervals for deviation from 2D towards 3D behaviour are $\Delta T_{23}(2223) \simeq 2.0$ K and $\Delta T_{23}(2212) \simeq 3.0$ K. This produces $\xi_{0\perp}(2223) \simeq 1.3$ Å and $\xi_{0\perp}(2212) \simeq 1.4$ Å suggesting similar coherence lengths along the c -axis for both materials.

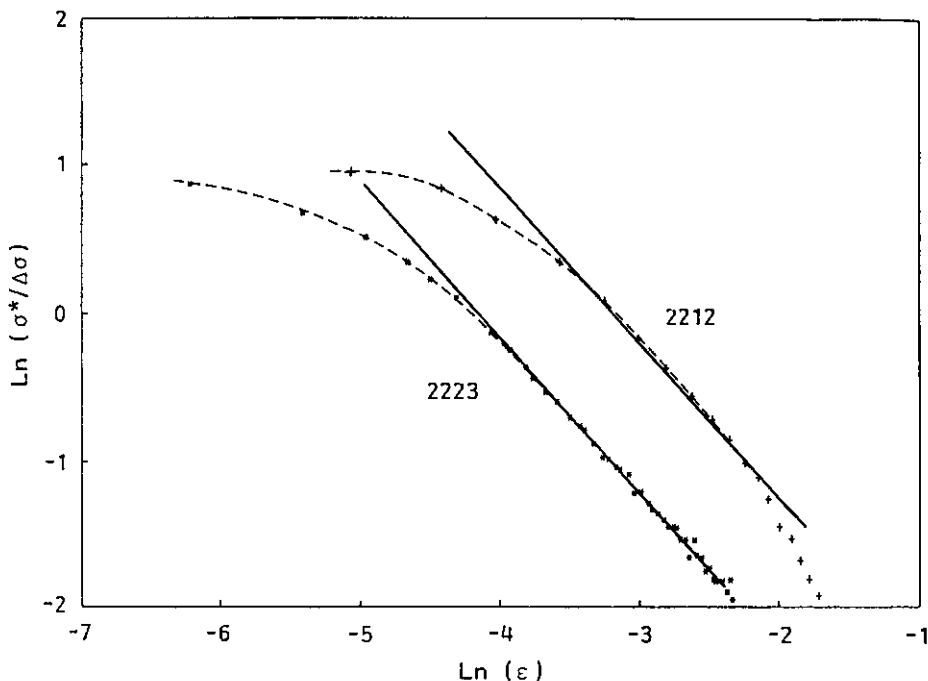


Figure 3. $\ln(\sigma^*/\Delta\sigma)$ as a function of $\ln(\epsilon)$, $\epsilon \equiv (T - T_{c0})/T_{c0}$ with the appropriate values of T_{c0} for the 2212 and 2223 cases. The straight lines have a slope of -1 , which is an indication of the quasi-2D behaviour. Proximity to T_{c0} is evident from the deviation of the data points (eye guided by dashed curves) towards smaller magnitude of the slope, which represents the dimensionality crossover from 2D to 3D.

In figure 4 we plot $\sigma_N/\Delta\sigma$ for both materials. According to equation (11) the slope, γ , of the extrapolated straight line (for the pure 2D case) down to T_{c0} is

$$\gamma = E_F / (k_B T_{c0}^2 B C^*). \quad (15)$$

We insert the 2D Fermi energy expression ($E_F = \pi \hbar^2 n d / m_e$) to obtain

$$\gamma = \pi \hbar^2 n d / (k_B T_{c0}^2 B m_e C^*). \quad (15a)$$

This yields $C^*(2223) \times B \simeq 0.5$ and $C^*(2212) \times B \simeq 3.0$ ($\gamma(2223) = 1.20$ K $^{-1}$, $\gamma(2212) = 0.61$ K $^{-1}$ and we use the previously introduced values of d and n). The theoretical value [29] for B is $B \simeq 0.2$. In fact, inspecting equations (13)–(15) it is apparent that independent of the value of C^* for each material, and irrespective of the sample parameter p , B should be given by

$$B = (\beta/\eta\gamma)(\pi/16T_{c0}). \quad (16)$$

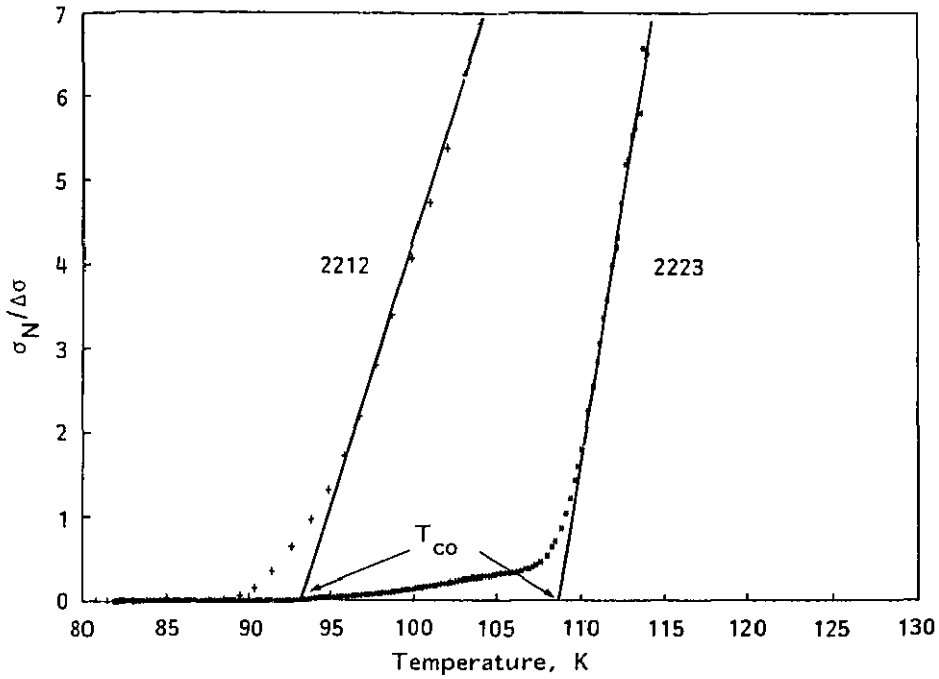


Figure 4. The temperature dependence of $\sigma_N/\Delta\sigma$. The slope of the straight lines, γ , is discussed in the text, see equations (15) and (15a).

Inserting the experimental values for the slopes β , η and γ together with the appropriate values of T_{co} we obtain $B(2223) \simeq 0.28$ and $B(2212) \simeq 0.26$ instead of the expected [29] value of 0.2 for this figure. We consider this discrepancy to be small and possibly due to the fact that the straightforward 2D Fermi energy expression ($E_F^0 = \pi \hbar^2 n d / m_e$) used to arrive at equation (16) together with the effective mass approximation approach are not readily applicable to the real material where coupling between the CuO layers is non-zero. For the quantitative analysis therefore we use the values of $B(2223)$ and $B(2212)$ above instead of 0.2. This produces $C^*(2223) \simeq 1.8$ and $C^*(2212) \simeq 11.7$ similar to the results obtained from the slopes, η , above. Alternatively $E_F^0(2223) \simeq C^*(2223) \times 0.343$ eV and $E_F^0(2212) \simeq C^*(2212) \times 0.120$ eV from the data of figure 4 for both materials. Since $\bar{E}_F = E_F(\text{studied material}) = E_F(\text{free-electron})/C^* \equiv E_F^0/C^*$, we determine the Fermi energies of our materials by this method which yields, respectively, $\bar{E}_F(2223) \simeq 0.343$ eV and $\bar{E}_F(2212) \simeq 0.120$ eV. The estimated values of \bar{E}_F are in reasonable agreement with the findings of other groups [30, 34] for the high- T_c cuprates.

Finally, equation (12) suggests the possibility of estimating Ω_D [35, 36] which enters the VL model. We note that the fluctuation effects in S , σ and L are closely related, since they all result from the influence of thermodynamic fluctuations of order parameter on non-equilibrium transport processes [37]. In figure 5 we present $S_N/|\Delta S|$ as a function of temperature for both materials. As has been noted in the text following equation (12), the slope ϕ of the asymptotic behaviour of $S_N/|\Delta S|$ should extrapolate to that of the product of $(\sigma_N/\Delta\sigma)$ and

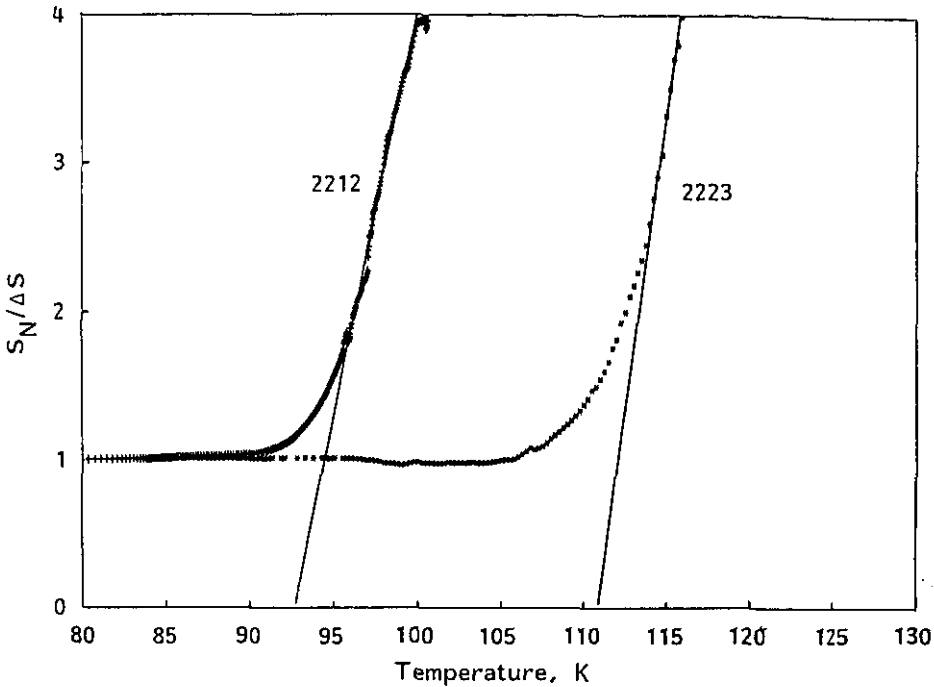


Figure 5. Temperature dependence of $S_N/\Delta S$, see equation (16) for information related to the slopes, ϕ , of the extrapolated straight lines.

$[1/|(A/B) \ln(\Omega_D/T_{\infty}) - 1|]$ for $\epsilon \rightarrow 0$, i.e.

$$\phi = \gamma/|(A/B) \ln(\Omega_D/T_{\infty}) - 1|. \quad (17)$$

The experimental values for ϕ were $\phi(2223) \simeq 0.72 \text{ K}^{-1}$ and $\phi(2212) \simeq 0.55 \text{ K}^{-1}$. Inserting the appropriate values for γ and B and solving for $\ln(\Omega_D/T_{\infty})$ we obtain $\Omega_D(2223) \simeq 105 \div 125 \text{ K}$ and $\Omega_D(2212) \simeq 93 \div 105 \text{ K}$ (the lower and upper values for the range of Ω_D are produced by the two possible solutions of equations (17)). Recently, Yu *et al* [38] have reported a heat capacity study of polycrystalline BiPbSrCaCuO material. In order to fit their data at the vicinity of the superconducting transition, i.e. at $T \simeq 109 \text{ K}$, they introduced a fitting routine that used atomic Debye temperatures weighted accordingly with the atomic fraction for each mass per unit formula. Their fit was successful to within 10% at 100 K and produced $\Omega_D(\text{Pb}) = 163 \text{ K}$ and $\Omega_D(\text{Bi}) = 162 \text{ K}$. These values are about 30–60% higher than the ones obtained by our method yet much closer to our results than the 470 K value for the single effective Debye temperature needed by Yu *et al* [38] to fit their data at 100 K. Note the proximity of our values to the atomic Debye temperatures of Pb (= 105 K) and Bi (= 119 K). Alternatively, since our values are less than half the average initial Debye temperatures (i.e. obtained at low temperatures from the heat capacity data) for 2212 ($\simeq 250 \pm 25 \text{ K}$) and 2223 ($\simeq 260 \pm 20 \text{ K}$) [39], we might suggest $\chi \simeq 2 \div 3$ for a simple modification of the VL model for the similar analysis of the fluctuation regime in high- T_c cuprates [35].

3. Summary

Special emphasis has been placed here on the application of different quasi-two-dimensional models to the analysis of fluctuation effects in the resistance and the thermoelectric power of melt-processed BiSrCaCuO (2212) and sintered BiPbSrCaCuO (2223) polycrystalline materials. Our approach allows an estimate of several important physical parameters for these systems: the Fermi energy, interlayer coupling strength (or $\xi_{0\perp}$) and atomic Debye temperatures for Bi and Pb. The results, especially for the former two parameters, are in a good agreement with the data obtained by other experimental investigations of similar materials.

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