

# Superconductivity fluctuation effects on the thermal conductivity of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$

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The superconductivity fluctuation contribution  $\kappa_{\text{fl}}$  to the thermal conductivity of a  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal is extracted from precise experimental data. The crossover from two-dimensional (2D) to 3D behavior theoretically predicted by Varlamov and Livanov is well marked. The crossover temperature  $T_{\text{VL}}$  and the amplitude of the fluctuation contribution lead to realistic values for the interlayer coupling energy  $J_c$  and the transport relaxation time  $\tau$  near  $T_c$ . [S0163-1829(96)51134-3]

## I. INTRODUCTION

There is much work already on superconductivity fluctuation contributions to high- $T_c$  superconductors (HTS's) transport properties,<sup>1</sup> mainly on the electrical resistivity  $\rho$  and the thermopower  $S$ . There was some report on the fluctuation contribution to the thermal conductivity  $\kappa_{\text{fl}}$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in the early days<sup>2</sup> though the data scattering was rather wide.

The HTS thermal conductivity  $\kappa$  remains an interesting and controversial transport property. Most of the works on thermal conductivity are devoted to the understanding of the peak structure observed below  $T_c$  in various high- $T_c$  materials.<sup>3</sup> While some authors believe that this peak is essentially due to the phonon contribution  $\kappa_{\text{ph}}$  (Refs. 3–7), others try to explain this feature by considering an alternative interpretation based on an electronic model.<sup>8–12</sup> Some relevant discussion concerns the characteristic dimensionality of the system.

In this paper, we investigate precise data on the thermal conductivity of a  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal near the critical temperature  $T_c$  in order to extract the contribution of fluctuations to this transport coefficient. The experimental results can be analyzed by using the theoretical model of Varlamov and Livanov (VL).<sup>13</sup> These authors derived the fluctuation contribution  $\kappa_{\text{fl}}$  to the thermal conductivity of high- $T_c$  superconductors by considering a Lawrence-Doniach model<sup>14</sup> to account for the layered structure of these materials.

Our experimental data can be very well reproduced by the VL theory, and we can distinguish between two-dimensional (2D) and 3D regimes. Besides, from the temperature crossover from 3D to 2D behavior and the amplitude of the fluctuation contribution, we estimate realistic values for the interlayer coupling energy  $J_c$  and the transport relaxation time  $\tau$  in this anisotropic material.

The theoretical results of Varlamov and Livanov<sup>13</sup> are recalled in Sec. II. The sample preparation and experimental technique to measure the temperature dependence of the thermal conductivity are presented in Sec. III. In Sec. IV, the experimental data are analyzed and discussed within the VL theory. Conclusions are finally drawn in Sec. V.

## II. THEORETICAL MODEL

The expression of the fluctuation contribution  $\kappa_{\text{fl}}$  to the thermal conductivity of layered HTS materials was calculated by Varlamov and Livanov.<sup>13</sup> This contribution is given by

$$\frac{\kappa_{\text{fl}}}{\kappa_n} = \frac{9\pi^5}{128[7\zeta(3)]^2} \frac{\hbar}{\varepsilon_F \tau} \left[ \frac{T-T_c}{T_c} \left( \frac{T-T_c}{T_c} + \delta^2 \right) \right]^{-1/2}, \quad (1)$$

where  $\kappa_n$  is the normal-state contribution to the thermal conductivity extrapolated from high temperature,  $\zeta(3)=1.202$  the Riemann  $\zeta$  function,  $\varepsilon_F$  the Fermi energy,  $\tau$  the transport relaxation time, and  $\delta$  is related to the interlayer coupling energy  $J_c$  by the expression

$$\delta = \left( \frac{7\zeta(3)J_c^2}{8\pi^2(k_B T_c)^2} \right)^{1/2}. \quad (2)$$

The expression given by Eq. (1) was obtained by considering a Lawrence-Doniach<sup>14</sup> spectrum in order to account for the anisotropic and layered structure of the high- $T_c$  cuprates.

From Eq. (1), one can see that in the limiting case of two- and three-dimensional behavior the fluctuation contributions to the thermal conductivity read

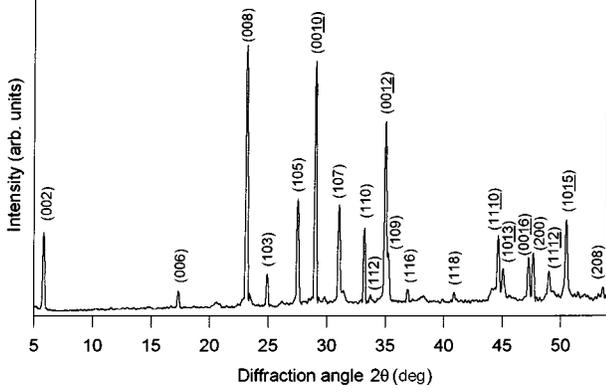


FIG. 1. X-ray diffraction pattern of a  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal.

$$\frac{\kappa_{\text{fl}}}{\kappa_n} = A \begin{cases} (1/\delta)\varepsilon^{-1/2} & \text{if } \delta^2 \gg \varepsilon \quad (3\text{D}) \\ \varepsilon^{-1} & \text{if } \delta^2 \ll \varepsilon \quad (2\text{D}), \end{cases} \quad (3)$$

where  $A = (9\pi^5\hbar)/(128[7\zeta(3)]^2\varepsilon_F\tau)$  and  $\varepsilon = (T - T_c)/T_c$ . Notice that the critical exponents related to the fluctuation contributions to the thermal conductivity are similar to the Azlamazov-Larkin prediction for the paraconductivity.<sup>1</sup> Varlamov and Livanov thus predict a crossover from 3D to 2D behavior at the  $T_{\text{VL}}$  temperature given by

$$T_{\text{VL}} = T_c + T_c\delta^2. \quad (4)$$

Consequently, the experimental derivation of  $T_{\text{VL}}$  should allow not only to estimate the range of the critical region but also to estimate the interlayer coupling energy  $J_c$  in the material, see Eq. (2). Let us remind that the expression of  $J_c$  in the Lawrence-Doniach model is given by

$$J_c = \frac{2\hbar}{\gamma d} \sqrt{\frac{\varepsilon_F}{m_{ab}^*}} \quad (5)$$

with  $d$  the interlayer spacing and  $\gamma = \sqrt{m_c^*/m_{ab}^*}$ .

### III. SAMPLE PREPARATION AND MEASUREMENT TECHNIQUE

The sample was specifically prepared to be as pure as possible following the method of Maeda *et al.*<sup>15</sup> We started from a  $\text{Bi}_2\text{O}_3$ ,  $\text{SrCO}_3$ ,  $\text{CaCO}_3$ ,  $\text{CuO}$  mixture for an intended  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  stoichiometry. The calcined powders were pressed into pellets, heated up to  $830^\circ\text{C}$  at a  $150^\circ\text{C/h}$  rate and sintered during three days in air. The samples were later quenched to room temperature to take x-ray data, then reheated to the  $830^\circ\text{C}$  sintering temperature during two more days, quenched again to take x-ray data, then reheated to  $850^\circ\text{C}$  and sintered during two more days. After quenching at room temperature x-ray data were again taken (Fig. 1). There is no doubt that the characteristic (00l) peaks of the 2212 phase have the highest intensity, which was seen to be increasingly higher on each x-ray successive pattern. Moreover, the samples can be said to be quasi single phase and well oriented from an x-ray point of view.

The thermal conductivity measurements face complications not found in the paraconductivity effect. The signals

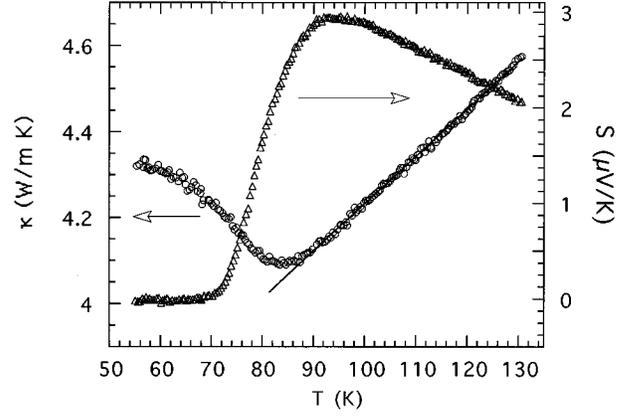


FIG. 2. Thermal conductivity  $\kappa$  and thermoelectric power  $S$  of a  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal as a function of temperature  $T$ . The solid line is a fit to the data by a first order polynomial for  $T > 100$  K (see text).

are less precise resulting from thermocouple differences. In order to appreciate the difficulty consider that a stable small and as constant as possible thermal gradient must be imposed but with a sweeping average temperature for the sample holder—the sweeping rate being slow, steplike, with the difference in the new average temperature with respect to the previous one less than the extremity temperature previous difference. Also a long time must evolve in order to await for a quasi steady state. A greatly enhanced noise background due to the intrinsically out of equilibrium conditions and electronic feedback controls further hinder resolution of the signal. Much care was taken and the sample holding assembly and data taken were very much monitored. We used the setup developed in Ref. 16 which allows us to measure the thermal conductivity and the thermoelectric power precisely and simultaneously.

In order to have fast thermal response times, thin films and single crystals are advantageous. However, the film substrates usually have a too high thermal conductivity. Moreover, it is almost impossible to have a single crystal on which the number of probes and oven can reasonably be soldered without interference effects. Thus we used the best sample available and much patience. Notice that unlike the thermopower measurements which depend only on the total integrated difference between voltage contacts, the thermal conductivity sensitively probes the local temperature differences.

### IV. ANALYSIS OF EXPERIMENTAL DATA AND DISCUSSION

The temperature dependence of the thermal conductivity  $\kappa$  and the thermoelectric power  $S$  of one  $(12 \times 2 \times 2)$  mm<sup>3</sup> size  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal is shown in Fig. 2. One can see from the large number of data points that  $\kappa$  is quasilinear at high temperatures as it is most usually observed in  $\text{BiSrCaCuO}$ -2212 compounds.<sup>10</sup> This behavior results from complicated interplay of various scattering mechanisms.<sup>4</sup> The solid line in Fig. 2 is a fit by a first-order polynomial of the data for  $T \in [100, 130]$  K to obtain the normal contribution  $\kappa_n$  to the thermal conductivity

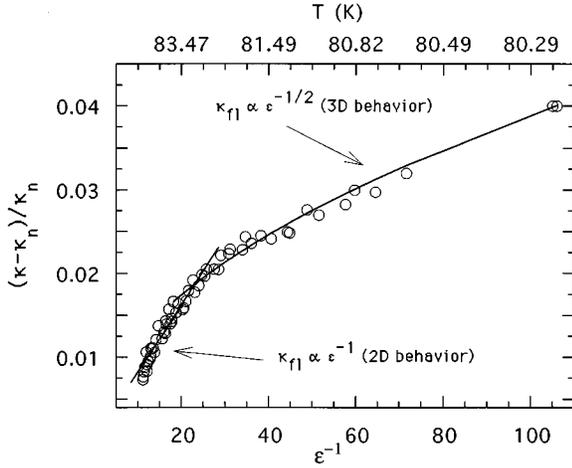


FIG. 3. Normalized fluctuation contribution  $(\kappa - \kappa_n)/\kappa_n$  to the thermal conductivity of a  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal as a function of  $\varepsilon^{-1}$  where  $\varepsilon = (T - T_c)/T_c$ .

$$\kappa_n(T) = 3.128 \pm 1.104 \times 10^{-2} T \quad (R = 0.99).$$

The fluctuation contribution  $\kappa_{fl}$  is obtained as usual<sup>1,2</sup> by subtracting this normal (background) contribution from the total thermal conductivity  $\kappa_{fl} = \kappa - \kappa_n$ . The normalized fluctuation contribution  $\kappa_{fl}/\kappa_n$  to the thermal conductivity of the  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  sample is shown in Fig. 3 as a function of  $\varepsilon^{-1}$  where  $\varepsilon = (T - T_c)/T_c$ . The critical temperature  $T_c = 79.5$  K was estimated from the inflexion point of the thermoelectric power  $S$ . From the formula of Presland *et al.*,<sup>17</sup> this corresponds to a hole carrier excess density of  $y = 0.20$  for overdoped Bi-2212-based samples. One can see from Fig. 3 that  $\kappa_{fl}$  follows a 2D behavior  $\kappa_{fl}/\kappa_n = 8.13 \times 10^{-4} \varepsilon^{-1}$  for  $T - T_c \in [3.3 \text{ K}, 7.9 \text{ K}]$  and that a crossover to a 3D behavior  $\kappa_{fl}/\kappa_n = 3.89 \times 10^{-3} \varepsilon^{-1/2}$  occurs at the temperature  $T_{VL} = 82.9$  K, in very good agreement with the theoretical prediction of Varlamov and Livanov.

In Fig. 4, a blow up of the crossover region is shown. The crossover from 2D to 3D behavior is well marked on such a log-log plot. From Eq. (2) and Eq. (4), the interlayer coupling energy  $J_c$  in the sample is experimentally estimated to be  $4.2 \times 10^{-3}$  eV. This parameter can also be estimated by using Eq. (5). By fixing the following reasonable values of the physical parameters,  $\varepsilon_F = 0.08$  eV,  $m_{ab}^* = 8m_0$ ,  $d = 3$  Å,<sup>18</sup> and  $\gamma = 50$ ,<sup>19</sup> we obtain the theoretical value  $J_c = 3.68 \times 10^{-3}$  eV, in quite good agreement with the value obtained above.

Next, we can derive the order of magnitude of the transport relaxation time  $\tau$  from the amplitude of the fluctuation contribution, cf. Eq. (1). Fixing the Fermi energy to be  $\varepsilon_F = 0.08$  eV,<sup>18</sup> we obtain  $\tau = 3.07 \times 10^{-12}$  s. This relaxation time value lies in the same range of magnitude than those obtained from the analysis of low frequency surface resistance<sup>20</sup> and thermal conductivity data in high- $T_c$  materials<sup>7,21</sup> and is indicative of the clean limit case for HTS's.<sup>22</sup>

It should be recalled that in the report by Cohn *et al.*<sup>2</sup> the

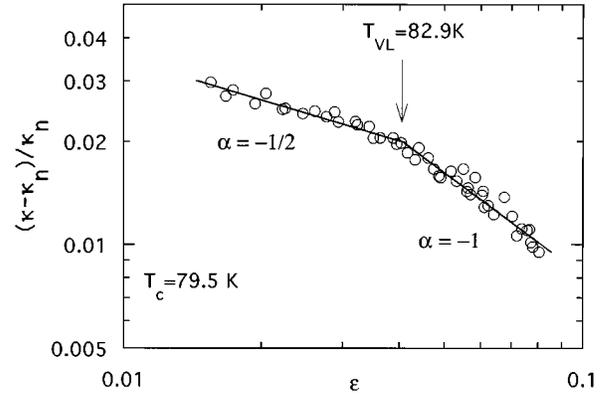


FIG. 4. Normalized fluctuation contribution  $(\kappa - \kappa_n)/\kappa_n$  to the thermal conductivity of a  $\text{Bi}_2\text{Sr}_{1.8}\text{Ca}_{1.2}\text{Cu}_2\text{O}_{8+y}$  polycrystal as a function of  $\varepsilon = (T - T_c)/T_c$ .

fluctuation contribution to the thermal conductivity of a  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystal was also observed, leading to an estimated interlayer coupling energy  $J_c \approx 5 \times 10^{-3}$  eV. However, from Eq. (5) and realistic values of the physical parameters and more specifically  $\gamma \approx 5$ ,<sup>23</sup> higher theoretical value, i.e., of the order of  $40 \times 10^{-3}$  eV should be obtained for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . This interlayer-coupling energy leads to a crossover temperature  $T_{VL} \approx 270$  K for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . Consequently either one should conclude that one could not observe this crossover in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  or that this temperature corresponds indeed to the spin-gap opening, and the onset of fluctuations as usually observed in such a temperature range in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .<sup>24</sup>

## V. CONCLUSION

In summary, we have successfully extracted the fluctuation contribution to the thermal conductivity of a  $\text{BiSrCaCuO}$ -2212 compound. Great care was taken to have numerous and reliable data. The fluctuation range is much more narrow than in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in agreement with the effective dimensionality of such different materials—an effective dimensionality which has been extracted. The experimental data can be well explained by the theory of Varlamov and Livanov.<sup>13</sup> From the temperature crossover from 3D to 2D behavior, we have obtained a very realistic value of the interlayer coupling energy  $J_c = 4.2 \times 10^{-3}$  eV in this highly anisotropic material. On the other hand, the value of the transport relaxation time  $\tau$  obtained from the amplitude of the fluctuation contribution seems quite reasonable.

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