

Excess conductivity in 2:2:1:2-phase Bi-Sr-Ca-Cu-O epitaxial thin films

G. Balestrino, M. Marinelli, and E. Milani

Dipartimento di Ingegneria Meccanica, Università di Roma "Tor Vergata," Via O. Raimondo 8, I-00173 Roma, Italy

Lucia Reggiani and R. Vaglio

Dipartimento di Fisica, Università di Salerno, I-84081 Baronissi, Salerno, Italy

A. A. Varlamov

Moscow Institute of Steel and Alloys, Leninskij pr. 4, Moscow, U.S.S.R.

(Received 2 March 1992)

Measurements of the excess conductivity above the transition temperature on selected, high-quality, superconducting epitaxial films of the 2:2:1:2-phase Bi-Sr-Ca-Cu-O (BSCCO), obtained by liquid-phase epitaxy, are reported. The data confirm the two-dimensional nature of the thermodynamical fluctuations in BSCCO compounds in a wide range of temperature above T_c and very good agreement is found with an extended version of the Aslamazov-Larkin theory [L. Reggiani, R. Vaglio, and A. A. Varlamov, *Phys. Rev. B* **44**, 9541 (1991)].

INTRODUCTION

Because of the high- T_c and low-coherence-length values, a large excess conductivity $\Delta\sigma$ due to thermodynamical fluctuations is generally observed in high- T_c superconductors.¹ The widespread interest in this effect is closely related to the question of dimensionality in high- T_c superconductors. In fact, since the cuprate superconductors present a layered structure, both two-dimensional (2D) and three-dimensional (3D) behavior can be observed in them, depending on the relative values of the temperature-dependent perpendicular coherence length $\xi_c(T)$ and on the layer spacing.²⁻⁵

It is well known that in the vicinity of the critical temperature ($T - T_c \ll T_c$) longitudinal paraconductivity of a layered superconductor can be presented in the general form⁶

$$\Delta\sigma(T) = \frac{e^2}{16\hbar d} \varepsilon^{-1} \left[1 + 2 \left(\frac{\xi_c(T)}{d} \right)^2 \right]^{-1/2}, \quad (1)$$

where $\varepsilon = \ln(T/T_c)$ [$\varepsilon \simeq (T - T_c)/T_c$ for $T \rightarrow T_c$] and d is the characteristic thickness of the layer.

In the immediate vicinity of the critical temperature, $\xi_c(T) \gg d$ and 3D behavior of the fluctuation conductivity takes place ($\Delta\sigma \sim \varepsilon^{-1/2}$). Further from T_c when $\xi_c(T) \leq d$ (but still $T - T_c \ll T_c$), this result is reduced to the well-known 2D Aslamazov-Larkin (AL) paraconductivity⁷

$$(\Delta\sigma)_{2D} = \frac{e^2}{16\hbar d} \varepsilon^{-1}. \quad (2)$$

Crossover between these two regimes takes place at temperature $T_{cr} - T_c \sim w^2/T_c$ (w is a value of the electron hopping integral between layers), when $\xi_c(T) \sim d$. Depending on the value of w , the crossover from 3D to 2D behavior can occur near or farther from the critical

temperature. So in Y-Ba-Cu-O (YBCO) compounds, 3D behavior was found in a sufficiently wide range of temperatures above T_c .^{3,4} In Bi-Sr-Ca-Cu-O (BSCCO) compounds, 3D behavior was observed in the nearest vicinity of the critical temperature only,⁶ but in a wide range of temperatures, clear 2D behavior of the paraconductivity was found. On these samples the fluctuation contribution is still observable even when the main assumption of the AL theory, that is $T - T_c \ll T_c$, breaks down. It is why this theory has been recently expanded over a wider range of temperatures by Reggiani, Vaglio, and Varlamov.⁸ It was shown that at any temperature $T \geq T_{cr}$ the 2D excess conductivity can be written as

$$(\Delta\sigma)_{2D} = \frac{e^2}{16\hbar d} f(\varepsilon), \quad (3)$$

where the explicit expression for $f(\varepsilon)$ is reported in Ref. 8. The function $f(\varepsilon) = \varepsilon^{-1}$ for $T - T_c \ll T_c$ and $f(\varepsilon) \sim \varepsilon^{-3}$ when $T \gg T_c$ (see Ref. 8).

The aim of the present Brief Report is to present careful measurements on 2:2:1:2-phase BSCCO superconducting thin films obtained by liquid-phase epitaxy and to show that very good agreement is found with Eq. (3), with very reasonable values for the fitting parameters.

SAMPLE PREPARATION AND CHARACTERIZATION

When performing experiments on fluctuation conductivity, extreme care should be taken in the choice of the sample to be studied. In fact, fluctuation conductivity measurements are extremely sensitive to the shape of the resistance-vs-temperature curve in the interval $T_c < T < 2T_c$. Any sample-dependent deviation of the $R(T)$ curve from the optimal "intrinsic" behavior must therefore be carefully excluded. In particular, the follow-

ing requirements must be fulfilled.

- (i) The samples should be homogeneous single crystals.
- (ii) The behavior of the resistance in the normal-state region must be metallic with a low extrapolated resistance at 0 K.

- (iii) The transition must be as narrow as possible, with no broadening other than the fluctuation-induced one.

Recently, the liquid-phase-epitaxy (LPE) method has been used to grow high-quality, truly epitaxial $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{7+\delta}$ films (2:2:1:2-phase BSCCO).⁹ By this method we have grown a set of films, which have been carefully characterized in order to check their compatibility with the above-defined requirements.

The growth procedure is described in detail in Ref. 10. The 1- μm -thick, *c*-axis-oriented films used in this experiment were grown on NdGaO_3 substrates, since a previous study¹¹ showed that this substrate allows one to obtain films having very good homogeneity and structural and transport properties. Twenty films were prepared and fully characterized by x-ray diffractometry (XRD), scanning electron microscopy (SEM), and energy-dispersive microprobe analysis (EDS). Resistivity measurements were performed with the standard four-probe technique.

The films were checked to be single phase using a Bragg-Brentano diffractometer with Cu to $K\alpha$ radiation. No contamination from 2:2:0:1 or 2:2:2:3 phases was detected. The mosaic spread of the films determined by a θ scan of the intense (0010) reflection was lower or very close to 0.1 (full width at half maximum). The films were checked to be truly epitaxial with respect to the substrate using asymmetric Bragg reflections as described elsewhere.⁹

For each film EDS measurements were taken in at least three different spots (1 μm^2 size) to evaluate their compositional homogeneity. The composition was normalized to seven cations per formula unit, and the films were discarded if for two different spots there was a difference in the content of any cation larger than 0.05 atoms per formula unit (afu), corresponding to our experimental uncertainty.

Finally, the resistance-vs-temperature curves (Fig. 1) were analyzed. A high-resolution nanovoltmeter and a very-high-stability current generator were used to mea-

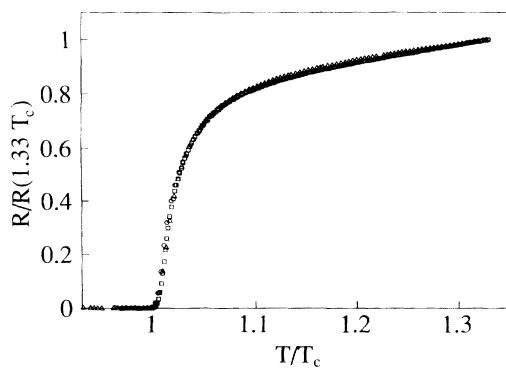


FIG. 1. Resistance-vs-reduced-temperature T/T_c curves for the three samples utilized for fluctuation measurements. The resistances are normalized for their values at $T = 1.33T_c$.

sure the $R(T)$ curves to obtain high-quality data. The temperature curve was monitored using a calibrated GaAs diode temperature sensor, and the cooling rate was kept constant to 12 K/h during the whole measurement.

RESULTS AND DISCUSSION

Only samples fulfilling the requirements for fluctuation studies discussed in the previous section have been considered. In particular, we will report, in the following, results relative to three films which showed optimal characteristics. It is interesting to note that the transitions chosen were those of films post-annealed in oxygen atmosphere at about 500°C. A possible explanation of this is that the increase in oxygen content leads to a more homogeneous distribution of the oxygen itself and therefore sharpens the transition. The decrease in T_c due to oxygen annealing has no effect on fluctuation conductivity studies, provided, of course, that the transition width is not increased, as in our case. In order to check that this selection was effective to avoid sample-dependent effects on the excess conductivity induced by fluctuations, a plot of the resistive transition was drawn for the chosen films.

To make a direct comparison among the curves, the temperatures were normalized to the critical temperatures T_c , defined as $R(T_c) = R_{\text{lin}}(T_c)/2$, where $R(T)$ are the measured resistances and $R_{\text{lin}}(T)$ represent the linear best fit to the data in the region $120 \leq T \leq 160$ K, where a fully metallic behavior is expected and observed ($T_c = 80.8, 77.0, \text{ and } 75.0$ K for the three films, respectively). The resistances were normalized to their values at $T = 1.33T_c$. Though the critical-temperature values are quite different, the curves for the three selected films superimpose very well, and we believe that they represent indeed the “intrinsic” form of the resistive transition for the 2:2:1:2-phase BSCCO films.

The experimentally observed excess conductivity $\Delta\sigma$, normalized to the room-temperature conductivity σ_0 , is determined, for the three films, by the relation

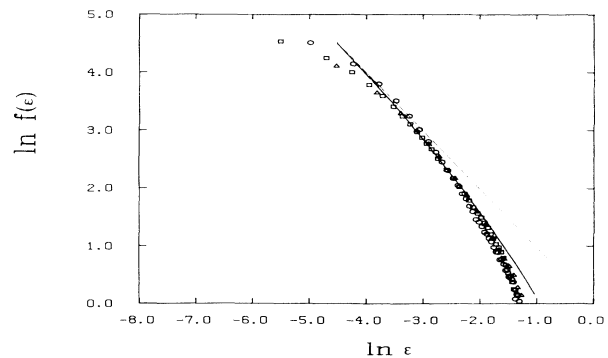


FIG. 2. Normalized excess conductivity $f(\epsilon) = (16\hbar d/e^2)\Delta\sigma$ vs $\epsilon = \ln(T/T_c)$ in a ln-ln scale. The solid line represents the extended theory by Reggiani, Vaglio, and Varlamov, and the dashed line represents Aslamazov-Larkin theory.

$$\frac{\Delta\sigma}{\sigma_0} = \rho_0 \left[\frac{1}{\rho(T)} - \frac{1}{\rho_{\text{lin}}(T)} \right]$$

$$= R_0 \left[\frac{1}{R(T)} - \frac{1}{R_{\text{lin}}(T)} \right], \quad (4)$$

where $\rho_0 = \sigma_0^{-1}$. In Fig. 2, $\Delta\sigma/\sigma_0$, normalized to $e^2\rho_0/16\hbar d$, is compared with the theoretical predictions [Eqs. (2) and (3)], assuming, for all films, $\rho_0/d = 15 \times 10^2 \Omega$.

For all the selected films, the agreement with Eq. (3) (continuous line in Fig. 2) is indeed excellent in the range $-4 \leq \ln \epsilon \leq -2$ ($1.5 < T - T_c < 11$ K), and it is clear how the extended theory by Reggiani, Vaglio, and Varlamov provides a better fit of the data with respect to

Aslamazov-Larkin theory [Eq. (2), dashed line in Fig. 2]. In this temperature range, the results are not seriously affected by the T_c definition or by the selected temperature range for the determination of $R_{\text{lin}}(T)$. The observed deviation of the data from the theoretical prediction for $\ln \epsilon < -4$ can be ascribed to the crossover to a 3D behavior approaching T_c , since the coherence length becomes comparable to the layer spacing, whereas the high-temperature deviation for $\ln \epsilon > -2$ can be ascribed to the adopted range for the definition of $R_{\text{lin}}(T)$ that imposes the condition $\Delta\sigma(T \geq 120 \text{ K}) = 0$.

Finally, it is worth emphasizing that the value of the fitting parameter $\rho_0/d = 15 \times 10^2 \Omega$ is consistent with the measured values of ρ_0 ($\approx 150 \mu\Omega \text{ cm}$) and with the typically assumed values of d ($\approx 10 \text{ \AA}$) for 2:2:1:2-phase BSCCO.

- ¹P. P. Freitas, C. C. Tsuei, and T. S. Plaskett, Phys. Rev. B **36**, 833 (1987).
²B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, Phys. Rev. B **37**, 7861 (1988).
³Felix Vidal, J. A. Veira, J. Maza, J. J. Ponte, F. Garcia-Alvarado, E. Moran, and M. A. Alario, Physica C **153-155**, 1371 (1988).
⁴Felix Vidal, J. A. Veira, J. Maza, F. Miguelez, E. Moran, and M. A. Alario, Solid State Commun. **66**, 421 (1988).
⁵G. Balestrino, A. Nigro, R. Vaglio, and M. Marinelli, Phys. Rev. B **39**, 12 264 (1989).
⁶W. E. Lawrence and S. Doniach, in *Proceedings of the Twelfth*

International Conference on Low Temperature Physics, edited by Eizo Kanda (Keigaku, Tokyo, 1971), p. 361.

- ⁷L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela (Leningrad) **10**, 1104 (1968) [Sov. Phys. Solid State **10**, 875].
⁸L. Reggiani, R. Vaglio, and A. A. Varlamov, Phys. Rev. B **44**, 9541 (1991).
⁹G. Balestrino, M. Marinelli, E. Milani, A. Paoletti, and P. Paroli, J. Appl. Phys. **68**, 361 (1990).
¹⁰G. Balestrino, M. Marinelli, E. Milani, A. Paoletti, and P. Paroli, J. Appl. Phys. **70**, 6939 (1991).
¹¹G. Balestrino, V. Foglietti, M. Marinelli, E. Milani, A. Paoletti, and P. Paroli, Solid State Commun. **79**, 839 (1991).