

## Superconducting fluctuations in $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ thin films: Paraconductivity, excess Hall effect, and magnetoconductivity

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A detailed study of normal-state magnetotransport properties in  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  thin films with a zero-resistance critical temperature  $T_{c0} = 105$  K prepared by dc-magnetron sputtering on MgO substrates is reported. Measurements of the electrical resistivity, the magnetoresistance, and the Hall effect are analyzed with regard to contributions of the superconducting order-parameter thermodynamic fluctuations, using theories for two-dimensional, layered superconductors. We have obtained a consistent set of parameters, i.e., the in-plane coherence length  $\xi_{ab}(0) = 1.6$  nm, the out-of-plane coherence length  $\xi_c(0) = 0.14$  nm, and the electron-hole asymmetry parameter  $\beta = -0.38$ . At temperatures below 118 K, we observe a remarkable enhancement (above theoretical predictions) of both the excess Hall effect and magnetoconductivity, whereas no such effect is detected for the zero-field paraconductivity. The above anomalies are attributed to a nonuniform critical temperature distribution inside our samples and can be well explained assuming a Gaussian distribution of  $T_c$ 's with a standard deviation  $\delta T_c = 2.3$  K. The excess Hall effect caused by superconducting fluctuations is negative in the entire accessible temperature range, which indicates, together with the paraconductivity and magnetoconductivity results that the indirect (Maki-Thompson) fluctuation process for  $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  is vanishingly small at temperatures from  $T_c$  to 130 K.

### I. INTRODUCTION

Although the phenomenon of thermodynamic fluctuations of the superconducting order parameter is well known in conventional superconductors and a considerable amount of experimental and theoretical studies have been collected so far,<sup>1</sup> it has tremendously regained interest with the discovery of the high-temperature superconductors (HTS's).<sup>2,3</sup> Several features of the cuprate superconductors like the high-energy value of the critical temperature  $T_c$ , the short coherence length along the copper-oxide planes, and the very small coherence length perpendicular to the planes, give rise to a large magnitude and a considerable extended temperature region of fluctuation effects, in this new class of materials. In HTS's, the fluctuation corrections to the higher-order components of the normal-state magnetoconductivity tensor are within experimental limits, and investigations of the thermodynamic fluctuations in transport properties are not limited to the influence on the material's electrical conductivity (usually denoted as *paraconductivity*), as they were in conventional superconductors, but may be extended to the Hall effect and to the magnetoresistivity.

A large amount of investigations of the paraconductivity in several HTS materials, including  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO),  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (YBCO), the 2:2:1:2 and 2:2:2:3 phases of Bi-Sr-Ca-Cu-O (BSCCO) and Tl-Ba-Ca-Cu-O

(TBCCO), have been performed. It turned out that the early results obtained on bulk ceramic samples could be well reproduced in highly oriented thin films and single crystals. There is, however, a major drawback in the analysis of superconducting fluctuations from the paraconductivity data alone in the cuprate superconductors. Since it is technically impossible to suppress the superconducting fluctuations by a high magnetic field, one has to make assumptions on the temperature dependence of the normal-state resistivity, in order to separate between those two contributions to the conductivity above  $T_c$ . Usually, a linear temperature dependence of the resistivity in the normal state is postulated, but such assumption is lacking an undisputed theoretical background. This ambiguity is one of the primary reasons for some controversy in the analysis of superconducting fluctuations in HTS's.

Following the initial derivations of the paraconductivity by Aslamazov and Larkin<sup>4</sup> (AL) and the extensions to the model of a layered superconductor by Lawrence and Doniach<sup>5</sup> (LD), the dimensionality of the superconducting fluctuations and, in the latter model, the crossover from two-dimensional (2D) to three-dimensional (3D) behavior in the vicinity of  $T_c$  can be estimated. There has been a considerable discussion whether the results in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  indicate a 2D behavior,<sup>6,7</sup> with a possible crossover to 3D,<sup>8</sup> or isotropic superconductivity.<sup>9</sup> This question is of substantial interest with regard to the role of coupling along the  $c$  axis in the superconducting state.

The results for the Bi (Refs. 10–20) and Tl cuprates<sup>21–23</sup> seem to indicate a 2D behavior, but, as we will point out, the 3D region close to  $T_c$  may be masked by an inhomogeneous  $T_c$  distribution in those samples. In granular ceramic superconductors, a fractal behavior, different from 2D and 3D, has been found and interpreted in terms of a superconducting percolation network.<sup>24</sup>

Another point of discussion is the possible presence or absence of the indirect fluctuation contribution, which was initially proposed by Maki<sup>25</sup> and Thompson<sup>26</sup> (MT). This question is of significant importance since it is possible to derive the AL contribution in the framework of the phenomenological time-dependent Ginzburg-Landau theory;<sup>27</sup> thus, the AL mechanism is insensitive to the nature of superconductivity. On the other hand, the MT process was calculated on a BCS-based microscopic ansatz and, consequently, should be dependent on the pairing mechanism. Strong inelastic scattering as it was proposed for the cuprate superconductors<sup>28</sup> is known to suppress the MT process;<sup>29</sup> also, symmetry considerations, like *s*-wave, extended *s*-wave, and *d*-wave superconductivity in the cuprates could possibly affect the MT contribution.<sup>30</sup> Whereas the investigations generally indicate a considerable weight of the MT contribution in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  (Refs. 31–36), possibly with a modified temperature dependence,<sup>37,38</sup> the indirect contribution seems to be absent in the paraconductivity of the more anisotropic BSCCO compound.<sup>10–20,39</sup>

To overcome the above-mentioned problems with the unknown transport properties of the normal-state in HTS, an analysis of the fluctuation *magnetoconductivity* above  $T_c$  has been proposed.<sup>29,40–43</sup> Due to the method of the magnetoconductivity measurement, the normal-state resistivity cancels, and, consequently, an assumption of its temperature dependence is no longer needed. The magnetoconductivity involves a nonzero magnetic field and, hence, represents a more subtle probing of the transport properties in the fluctuation region than the paraconductivity. Several studies of the fluctuation magnetoconductivity have been performed on YBCO ceramics, thin films, and single crystals,<sup>3</sup> but the results from superconducting conductivity fluctuations under the influence of a magnetic field in BSCCO are rather scarce, and were not analyzed using the available theoretical background.<sup>12,20,44,45</sup>

The influence of superconducting fluctuations on the off-diagonal components of the magnetoconductivity tensor (usually denoted the *excess Hall effect*) was observed in YBCO,<sup>38</sup> but, to our best knowledge, no such analysis has been performed on either BSCCO or TBCCO. Interestingly, the excess Hall effect provides some additional insights into details of the band structure<sup>46</sup> and also is a sensitive method for detection of the MT process, since it may have a different sign than the AL contribution.<sup>38</sup>

Although the quality of HTS's has been improved considerably since the first studies on ceramic samples, minor inhomogeneities of  $T_c$  cannot be completely ruled out. Such effect of a  $T_c$  distribution on the paraconductivity<sup>47,48</sup> and on the fluctuation magnetoconductivity<sup>48</sup> has been investigated previously and will be extended to the excess Hall effect in this paper.

It seems evident that complementary measurements of different transport phenomena, like fluctuation magnetoconductivity and excess Hall effect, in addition to the paraconductivity data, are needed to obtain a full physical picture of superconducting fluctuations in HTS's and a corresponding set of reliable parameters. In this paper, we present a comprehensive study of superconducting fluctuations in thin films of the 2:2:2:3 phase of BSCCO and an analysis using a consistent set of parameters. The paper is organized as follows: In Sec. II, the theories for paraconductivity, excess Hall effect, and fluctuation magnetoconductivity are reviewed, and modifications for samples with nonuniform  $T_c$ 's are proposed. In Sec. III we describe the sample preparation and the measurement techniques, and present our experimental results. In Sec. IV, the analysis of the data with regard to superconducting fluctuations is discussed. Finally, in Sec. V, we comment on some implications coming from our results.

## II. THEORETICAL BACKGROUND

### A. Paraconductivity

At temperatures above  $T_c$ , the magnetoconductivity tensor in the normal state of a superconductor is modified by corrections originating from thermodynamic fluctuations of the superconducting order parameter. In zero magnetic field, those corrections to the diagonal components are additive, resulting in an enhancement of the conductivity by the paraconductivity  $\Delta\sigma_{xx}(0)$ . Thus, the conductivity equals

$$\sigma_{xx}(0) = \sigma_{xx}^N(0) + \Delta\sigma_{xx}(0), \quad (1)$$

where  $\sigma_{xx}^N(0)$  is the normal-state value.

The calculation of the paraconductivity was reported by AL, who considered the acceleration of short-lived Cooper pairs, which are created in thermal nonequilibrium above  $T_c$ , in an electric field.<sup>4</sup> The main results for the so-called “direct” or “regular” contribution in two and three dimensions are, respectively,

$$\Delta\sigma_{xx}^{\text{AL(2D)}}(0) = \frac{e^2}{16\hbar t} \varepsilon^{-1}, \quad (2)$$

$$\Delta\sigma_{xx}^{\text{AL(3D)}}(0) = \frac{e^2}{32\hbar\xi(0)} \varepsilon^{-1/2}, \quad (3)$$

where  $e$  is the electron charge,  $\hbar$  is the reduced Planck constant,  $t$  is the thickness of the film,  $\xi(0)$  is the zero-temperature Ginzburg-Landau coherence length and  $\varepsilon = \ln(T/T_c) \approx (T - T_c)/T_c$  is a reduced temperature.

The initial AL expressions were derived in the BCS model, but the same results can be obtained in the phenomenological Ginzburg-Landau approach.<sup>27</sup> This indicates that the direct contribution is insensitive to the microscopic mechanism responsible for the carrier pairing and therefore should be also valid in HTS's. LD extended the calculations to a superconductor that consists of a stack of two-dimensional superconducting sheets, linked by Josephson coupling between adjacent layers.<sup>5</sup> Obviously this scenario resembles closely the situation in most HTS's and, consequently, will be used in our further con-

siderations. The paraconductivity along the layer is

$$\Delta\sigma_{xx}^{\text{LD}}(0) = \frac{1}{16} \frac{e^2}{\hbar d} (1+2\alpha)^{-1/2} \varepsilon^{-1}, \quad (4)$$

where  $d$  is the layer spacing,

$$\alpha = \frac{2\xi_c^2(0)}{d^2} \varepsilon^{-1} \quad (5)$$

is a dimensionless coupling parameter, and  $\xi_c(0)$  the zero-temperature coherence length along the stacking direction. The LD formula reduces to the corresponding AL results: Eq. (2) in the 2D limit ( $\alpha=0$ ) and Eq. (3) in the 3D limit ( $\alpha \rightarrow \infty$ ), respectively.

An additional indirect contribution to the paraconductivity, arising from the decay of superconducting pairs into quasiparticles and vice versa, was first calculated by MT and, recently, extended to the LD model by Hikami and Larkin<sup>29</sup> and Maki and Thompson,<sup>41</sup>

$$\Delta\sigma_{xx}^{\text{MT}}(0) = \frac{e^2}{8\hbar d} \frac{1}{\varepsilon - \delta} \ln \left\{ \frac{\varepsilon}{\delta} \frac{1 + \alpha + (1 + 2\alpha)^{1/2}}{1 + \alpha\varepsilon/\delta + (1 + 2\alpha\varepsilon/\delta)^{1/2}} \right\}. \quad (6)$$

Here  $\delta$  is a pair-breaking parameter, which was introduced semiphenomenologically by Thompson<sup>26</sup> and is related to the shift of  $T_c$  by magnetic impurities<sup>49</sup> or inelastic scattering processes that limit the phase-relaxation time  $\tau_\phi$  of the quasiparticles involved in the MT process. In the limit of dirty superconductors

$$\delta = \pi\hbar / (8k_B T \tau_\phi), \quad (7)$$

while for clean superconductors  $\delta$  has to be divided by 1.203 [ $l/\xi_{ab}(0)$ ];  $l$  is the quasiparticle mean free path and  $\xi_{ab}(0)$  the zero-temperature coherence length along the superconducting layers.<sup>42</sup>

From Eqs. (4) and (6) it follows that the indirect contribution is of importance relative to the AL process only if the inelastic-scattering length is large compared to the coherence length. Due to the different temperature dependence in 2D superconductors, however, the direct fluctuation contribution is in any case dominant close to  $T_c$  with a possible crossover to the MT contribution at higher temperatures. In an  $s$ -wave BCS superconductor, nonmagnetic impurities neither change  $T_c$  nor affect the AL and the MT terms. On the other hand in  $p$  or  $d$ -wave superconductors those impurities may act pair breaking<sup>50</sup> and are supposed to suppress the MT contribution.<sup>30</sup>

### B. Excess Hall effect

The fluctuation corrections also influence the off-diagonal components of the normal-state magnetoconductivity tensor, viz. the Hall conductivity. The total Hall conductivity may be expressed as

$$\sigma_{xy} = \sigma_{xy}^N + \Delta\sigma_{xy}, \quad (8)$$

where  $\sigma_{xy}^N$  is the normal-state part and  $\Delta\sigma_{xy}$  is the fluctuation correction originating from the excess Hall effect.

An initial calculation of the fluctuation Hall effect by

Abrahams, Prange, and Stephen,<sup>51</sup> was followed by a microscopic theory by Fukuyama, Ebisawa, and Tsuzuki<sup>46</sup> (FET). More recently, Ullah and Dorsey<sup>52</sup> (UD) have examined the problem on the grounds of the time-dependent Ginzburg-Landau theory and another microscopic calculation of the excess Hall effect was performed by Varlamov and Livanov.<sup>53</sup>

For further analysis we use the result of UD in the low-field limit  $h \ll \varepsilon$ , i.e., in small magnetic fields and for temperatures not too close to  $T_c$ ;  $h = \ln[T_c(0)/T_c(B)] = 2e\xi_{ab}^2(0)B/\hbar$  reflects the shift of the mean-field  $T_c$  in a magnetic field  $B$ . Although the magnetic field used in our experiments is moderate, we include the  $T_c$  shift into the analysis by renormalizing  $\varepsilon$  in finite magnetic field,  $\varepsilon_H = \ln[T/T_c(B)] = \varepsilon + h$ . The excess Hall effect induced by the AL process is

$$\Delta\sigma_{xy}^{\text{UD}} = \frac{e^2}{16\hbar d} \frac{\sigma_{xy}^N}{\sigma_{xx}^N} \beta \frac{\pi d}{72\xi_c(0)} \frac{1+1/\alpha}{(1+1/2\alpha)^{3/2}} \varepsilon_H^{-3/2}, \quad (9)$$

where  $\beta$  reflects an asymmetry between electrons and holes. This electron-hole asymmetry parameter appears both in the microscopic calculations of FET (denoted  $\alpha$ ) and in the phenomenological model of UD. FET found that  $\beta$  depends on the energy derivative of the electron density of states at the Fermi energy and UD incorporated the electron-hole asymmetry into the time-dependent Ginzburg-Landau theory by postulating a complex relaxation time of the order parameter. Interestingly, recent calculations<sup>54,55</sup> of the Hall effect resulting from vortex motion below  $T_c$  connect between the excess Hall effect and the Hall effect in the flux-flow region via the imaginary part of the relaxation time, which corresponds to our  $\beta$ . According to this picture, the sign of  $\beta$  governs both the sign of the AL excess Hall effect and the sign of the Hall effect below  $T_c$ .

We know of no independent calculation of the MT contribution to the excess Hall effect in the LD model, but based on the 2D and 3D limits in the FET theory, one may deduce a scaling relation<sup>56,38</sup> of the MT term of the fluctuation Hall conductivity with the corresponding paraconductivity expression and, applying it to Eq. (5), obtain

$$\Delta\sigma_{xy}^{\text{MT}} = \frac{e^2}{16\hbar d} \frac{\sigma_{xy}^N}{\sigma_{xx}^N} \frac{4}{\varepsilon_H - \delta} \times \ln \left\{ \frac{\varepsilon_H}{\delta} \frac{1 + \alpha + (1 + 2\alpha)^{1/2}}{1 + \alpha\varepsilon_H/\delta + (1 + 2\alpha\varepsilon_H/\delta)^{1/2}} \right\}. \quad (10)$$

Finally, combining Eqs. (9) and (10), one can get the excess Hall effect as  $\Delta\sigma_{xy} = \Delta\sigma_{xy}^{\text{UD}} + \Delta\sigma_{xy}^{\text{MT}}$ .

### C. Fluctuation magnetoconductivity

The effect of a magnetic field on the superconducting fluctuations has been considered by Abrahams, Prange, and Stephen<sup>51</sup> for 2D superconductors and, recently, extended to the LD model by Hikami and Larkin<sup>29</sup> and Maki and Thompson.<sup>41</sup> Their work has been based on the magnetic-field pair-breaking effect that leads to a suppression of  $T_c$  in mean-field theory and decreases the

fluctuation conductivity. Hence, the above process results in a positive magnetoresistance, which is usually expressed in terms of a negative magnetoconductivity,  $\sigma_{xx}(B) - \sigma_{xx}(0)$ . The conductivities in the presence of  $B$ ,  $\sigma_{xx}(B) = \sigma_{xx}^N(B) + \Delta\sigma_{xx}(B)$ , as well as zero-field values  $\sigma_{xx}(0) = \sigma_{xx}^N(0) + \Delta\sigma_{xx}(0)$  are, in each case, the sum of the normal-state contribution and the fluctuation correction. Due to a very small magnetoresistance of HTS's in the normal state,  $\sigma_{xx}^N(B) \approx \sigma_{xx}^N(0)$ , the normal-state conductivity cancels in the measurement of the magnetoconductivity. Thus, the total change of the conductivity fluctuation corrections in a magnetic field is  $\Delta\sigma_B \equiv \Delta\sigma_{xx}(B) - \Delta\sigma_{xx}(0) \approx \sigma_{xx}(B) - \sigma_{xx}(0)$ .

The fluctuation magnetoconductivity comprises of four different contributions, resulting from interactions of the magnetic field with the carrier orbits and spins, and taking into account both the AL and MT processes. The orbital effect on the AL contribution (ALO) in the layered superconductor model<sup>29</sup> is given by

$$\Delta\sigma_B^{\text{ALO}} = \frac{e^2}{8\hbar} h^{-2} \int_0^{2\pi/d} \epsilon_k \left[ \psi \left[ \frac{1}{2} + \frac{\epsilon_k}{2h} \right] - \psi \left[ 1 + \frac{\epsilon_k}{2h} \right] + \frac{h}{\epsilon_k} \right] \frac{dk}{2\pi} - \Delta\sigma_{xx}^{\text{LD}}(0), \quad (11)$$

where  $\epsilon_k = \varepsilon[1 + \alpha(1 - \cos kd)]$ ,  $k$  is the momentum parallel to  $B$ , and  $\psi$  is the di-gamma function. In the low-field limit  $h \ll \varepsilon$ , Eq. (11) may be expanded to

$$\Delta\sigma_B^{\text{ALO}} \approx -\frac{1}{64} \frac{e^2}{\hbar d} \frac{2 + 4\alpha + 3\alpha^2}{(1 + 2\alpha)^{5/2}} \varepsilon^{-3} h^2. \quad (12)$$

Thus, the ALO part of the magnetoconductivity in the 2D case,  $\Delta\sigma_B^{\text{ALO(2D)}} \propto \varepsilon^{-3}$ , is even more divergent than the excess Hall effect towards  $T_c$ .

The orbital effect on the MT contribution (MTO) in the dirty limit is<sup>29</sup>

$$\Delta\sigma_B^{\text{MTO}} = \frac{e^2}{8\hbar} \frac{1}{\varepsilon - \delta} \int_0^{2\pi/d} \left[ \psi \left[ \frac{1}{2} + \frac{\epsilon_k}{2h} \right] - \psi \left[ \frac{1}{2} + \frac{\epsilon_k}{2h} - \frac{\varepsilon - \delta}{2h} \right] \right] \frac{dk}{2\pi} - \Delta\sigma_{xx}^{\text{MT}}(0). \quad (13)$$

Aronov, Hikami, and Larkin<sup>40</sup> (AHL) extended the theory to include the interaction of the magnetic field with electron spins and obtained two additional Zeeman contributions to the fluctuation magnetoconductivity. The Zeeman effect on the AL fluctuation process (ALZ) is

$$\Delta\sigma_B^{\text{ALZ}} = \frac{e^2}{16\hbar d} [(1 + 2\alpha')^{-1/2} \varepsilon'^{-1}] - \Delta\sigma_{xx}^{\text{LD}}(0), \quad (14)$$

with

$$\varepsilon' = \varepsilon + \text{Re} \left[ \psi \left[ \frac{1}{2} + i \frac{g\mu_B B}{4\pi k_B T_c} \right] - \psi \left[ \frac{1}{2} \right] \right], \quad (15)$$

and  $\alpha' = 2\xi_c^2(0)/(d^2\varepsilon')$ . Here  $\mu_B$  denotes the Bohr magneton and  $g$  is the Landé factor that was assumed to be  $g = 2$ .

The Zeeman effect on the MT contribution (MTZ) is smaller than originally predicted in the AHL theory<sup>43,32</sup> and will be ignored in our investigations.

Theoretical considerations, as well as experimental analysis, show that the ALO contribution dominates near  $T_c$  in  $B$  oriented perpendicular to the superconducting layers. The situation is drastically changed, however, if  $B$  is parallel to the planes, where, in the two-dimensional limit, the orbital contributions are suppressed and the Zeeman terms become prevailing.

Finally, we want to mention that in the clean limit,<sup>42</sup> the magnetoconductivity is modified in the same manner as Eq. (7), while the consideration of the nonlocal effect<sup>43</sup> results only in minor changes to the MT terms.

#### D. Modifications for a nonuniform critical temperature

In a recent paper, we investigated the influence of a nonuniform  $T_c$  on the paraconductivity and on the fluctuation magnetoconductivity.<sup>48</sup> Using an effective-medium approach, we assumed that the fluctuation corrections are small additive contributions to the normal-state magnetoconductivity tensor. Furthermore we presupposed that various fluctuation processes are independent, thus, limited our considerations to temperatures not too close to  $T_c$ . Under the above approximations, the total fluctuation corrections can be obtained by summing over all individual fluctuation domains within a sample. For simplicity we base our analysis on a Gaussian distribution of  $T_c$ 's, with a mean value  $\bar{T}_c$  and a standard deviation  $\delta T_c$  and additionally assume that  $\delta T_c \ll \bar{T}_c$ . Using a first-order expansion of the effective-medium theory, we obtain the total paraconductivity in the sample with a different local critical temperature  $\vartheta_c$  as

$$\Delta\tilde{\sigma}_{xx}(0) = \int_0^\infty \frac{1}{\delta T_c \sqrt{2\pi}} \exp \left[ -\frac{(\vartheta_c - \bar{T}_c)^2}{2\delta T_c^2} \right] \times \{ \Delta\sigma_{xx}^{\text{LD}}(0) + \Delta\sigma_{xx}^{\text{MT}}(0) \} d\vartheta_c. \quad (16)$$

In our model, the length scale of the  $T_c$  inhomogeneities is temperature independent and considerably larger than the in-plane coherence length  $\xi_{ab}(T)$ . If  $\xi_{ab}(T) = \xi_{ab}(0)\varepsilon^{-1/2}$ , exceeds that length scale at temperatures close to the local  $\vartheta_c$ , we expect a significant reduction of the amplitude of the local superconducting fluctuations. To account for the above-mentioned limit we introduce a phenomenological, low-temperature cutoff which is incorporated in the LD and MT theories by replacing  $\varepsilon$  in Eqs. (4) and (6) by a modified function

$$\varepsilon^* = \begin{cases} \ln(T/\vartheta_c), & \ln(T/\vartheta_c) \geq \varepsilon_c \\ \varepsilon_c, & \ln(T/\vartheta_c) < \varepsilon_c, \end{cases} \quad (17)$$

where  $\varepsilon_c = \ln(T_{\text{cutoff}}/T_c)$  is the reduced cutoff temperature.

In an equivalent procedure, we modify the expressions

for the excess Hall effect and obtain

$$\Delta\bar{\sigma}_{xy} = \int_0^\infty \frac{1}{\delta T_c \sqrt{2\pi}} \exp\left[-\frac{(\vartheta_c - \bar{T}_c)^2}{2\delta T_c^2}\right] \times \{\Delta\sigma_{xy}^{\text{UD}} + \Delta\sigma_{xy}^{\text{MT}}\} d\vartheta_c, \quad (18)$$

where  $\varepsilon_H$  is now replaced by  $\varepsilon^* + h$ .

In Fig. 1, we compare (for a typical set of parameters) the results of Eq. (18) for materials with a  $T_c$  distribution with  $\Delta\bar{\sigma}_{xy}$  from Eqs. (9) and (10), which are based on the uniform  $T_c$ . The difference in the UD expressions is insignificant at higher temperatures,  $\varepsilon > 0.1$ , but in the temperature range  $0.01 < \varepsilon < 0.1$  our model predicts that the excess Hall effect is larger by a factor of up to 1.7. On the other hand, the indirect (MT) contribution remains practically unchanged in our modified theory. We observe, that the shape of the UD curve is altered in the respect that the curvature due to the 2D $\rightarrow$ 3D crossover in the LD model near  $T_c$  is masked by a counteracting change of slope due to the  $T_c$  distribution. A similar observation also has been made for the paraconductivity.<sup>48</sup>

Using the previous considerations, we obtain for the total magnetoconductivity in the  $T_c$  distribution model

$$\Delta\bar{\sigma}_B = \int_0^\infty \frac{1}{\delta T_c \sqrt{2\pi}} \exp\left[-\frac{(\vartheta_c - \bar{T}_c)^2}{2\delta T_c^2}\right] \times \sum_i \Delta\sigma_B^i(\varepsilon^*, B) d\vartheta_c, \quad (19)$$

where  $i = \{\text{ALO, MTO, ALZ, MTZ}\}$  represents the four different contributions.

In fact, the impact of an inhomogeneous  $T_c$  on  $\Delta\bar{\sigma}_B$  is

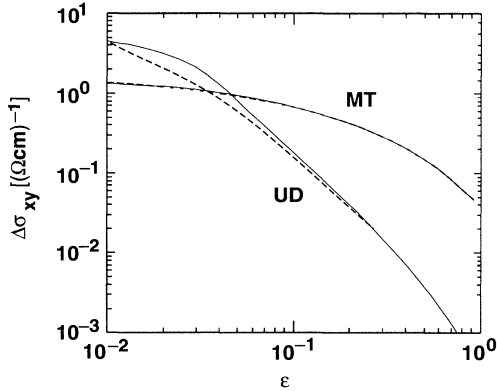


FIG. 1. Excess Hall effect  $\Delta\sigma_{xy}$  calculated for a uniform  $T_c$  (broken lines) and for the model with a Gaussian distribution of  $T_c$ 's in the sample (solid lines) as a function of the reduced temperature  $\varepsilon$ . The curves labeled UD and MT are the direct and the indirect fluctuation contribution, respectively. The parameters used are  $\xi_{ab}(0) = 1.5$  nm,  $\xi_c(0) = 0.15$  nm,  $d = 1.85$  nm,  $\delta = 0.3$ ,  $\beta = 0.4$ ,  $T_c = \bar{T}_c = 100$  K,  $\delta T_c = 2$  K, and  $\varepsilon_c = 5 \times 10^{-3}$ . The normal-state Hall effect is modeled by Anderson's formula (Ref. 64)  $\cot\theta_H = \sigma_{xx}^N / \sigma_{xy}^N = AT^2 + C$ , assuming  $A = 0.2$  and  $C = 0$ . The anomalous temperature dependence of the Hall angle in HTS is responsible for the downward bending of the curves at higher temperatures,  $\varepsilon > 0.2$ .

more pronounced than on the paraconductivity or the excess Hall effect. We have also found,<sup>48</sup> that the ALO contribution is the dominant fluctuation correction for magnetoconductivity in a large temperature range. Among the three contributions, the MTO and ALZ terms are only slightly enhanced in our model, and the MTZ term remains essentially unchanged.

### III. EXPERIMENTAL METHODS AND RESULTS

#### A. Sample preparation

The BSCCO films were fabricated according to our procedure developed for the  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$  (2223) phase formation, described in detail in Ref. 57. Briefly, the films were dc-magnetron sputtered from an off-stoichiometric, heavily Pb-doped BSCCO target (the exact target composition was  $\text{Bi}_2\text{Pb}_{2.5}\text{Sr}_2\text{Ca}_{2.15}\text{Cu}_{3.3}\text{O}_x$ ) on heated to  $150^\circ\text{C}$  single-crystalline MgO substrates. The as-deposited films were subsequently *ex situ* crystallized by annealing in air at  $870^\circ\text{C}$  for 1–2 h, then slowly cooled down to  $700^\circ\text{C}$  and quenched to room temperature. We found that the high initial Pb content and the slight excess of Ca and Cu, as well as precisely adjusted and controlled crystallization temperature ( $870^\circ\text{C}$ —1 to  $2^\circ\text{C}$  below the film melting point), were essential in obtaining films with the highest content of the 2223 phase.<sup>57</sup>

The annealed films showed a granular, "micalike" morphology with 10- to  $20\text{-}\mu\text{m}$ -long crystals, arranged parallel to the substrate surface. Figure 2 presents an x-ray-diffraction pattern of our BSCCO film annealed for 1 h. The spectrum contains reflections corresponding to 2223 and 2212 phases, labeled *H* and *L*, respectively. The fraction of the 2223 phase in the film, estimated from the intensity ratio of the H002 and L002 peaks, exceeded 90%. We note that no peaks related to other phases, except the one at  $2q = 17.6^\circ$  for the  $\text{Ca}_2\text{PbO}_4$ , were detected.  $\text{Ca}_2\text{PbO}_4$  forms at the initial crystallization stages of the Pb-doped BSCCO and is known to promote the 2223-phase formation, but it disappears rapidly during further annealing (with the escape of Pb from the film) and has not been detected in our films annealed for 2 h. We see that the only strong reflections in Fig. 2 originate from

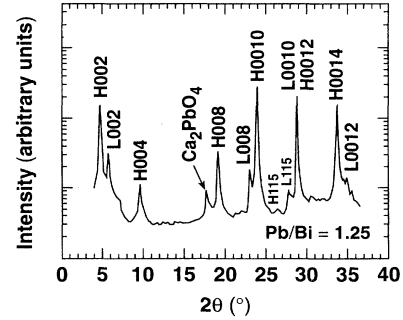


FIG. 2. X-ray-diffraction spectrum of the  $\text{Bi}_2\text{Pb}_{2.5}\text{Sr}_2\text{Ca}_{2.15}\text{Cu}_{3.3}\text{O}_x$  film deposited on MgO and annealed for 1 h at  $870^\circ\text{C}$ . The peaks of the 2223 and 2212 BSCCO phases are labeled *H* and *L*, respectively. Note that the intensity scale is logarithmic.

the (00*n*) BSCCO planes, indicating that the film was highly oriented with the *c* axis perpendicular to the substrate surface.

For the electrical transport measurements, our films were patterned using either conventional photolithography (with wet etching in diluted  $\text{H}_3\text{PO}_4$ ) or a laser ablation method.<sup>58</sup> In this latter case, the film was mounted on a computer-controlled two-dimensional translational stage with a 1  $\mu\text{m}$  minimum step size and the pattern was traced out by ablating away the unwanted material with a focused beam from a mode-locked Nd-YAG laser (laser fluence was few  $\text{J}/\text{cm}^2$ ). The ablation technique allowed us to fabricate our test structures with no need for the photomask (necessary in photolithography) and, what was even more important, without any degradation of the film properties by, e.g., edge undercutting and/or contacting the film with damaging chemicals. Our samples were patterned to a five-probe geometry suitable for transport measurements with typical strip dimensions of  $10 \times 1.5 \text{ mm}^2$ . Silver pads were evaporated on the arms of the patterned film and contacts were established with silver paste and gold wires.

The temperature dependencies of the normalized resistance and the critical current density  $j_c$  for the film of Fig. 2 is shown in Fig. 3. The measurement was done in a standard four-probe geometry on a 300- $\mu\text{m}$ -wide strip patterned in the film. The strip showed a well-defined superconducting transition with a zero-resistivity state at 105 K (see also Fig. 4). The  $j_c$  of the film (in a zero magnetic field) was about  $2 \times 10^2 \text{ A}/\text{cm}^2$  at 90 K and increases to  $2 \times 10^4 \text{ A}/\text{cm}^2$  at low temperature.<sup>59</sup> The temperature dependence of  $j_c$  showed a distinct irregularity at about 80 K—the superconducting transition temperature of the 2212 phase. Above 80 K,  $j_c$  was strongly suppressed and its temperature dependence was proportional to  $(T - T_c)^2$ , as is shown in the inset in Fig. 3. Such behavior of  $j_c$  in the sample with a predominantly 2223 phase suggests a nonuniform distribution of the superconducting phases in the film, with a preferential formation of the 2212 phase on the boundaries of the 2223 grains.<sup>60</sup> This, in turn, results in the presence of normal-

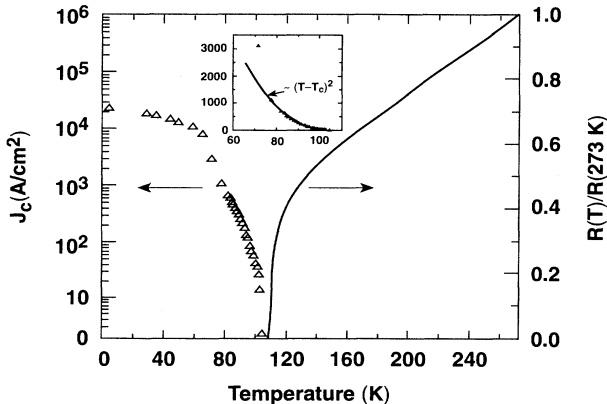


FIG. 3. The temperature dependencies of the normalized resistance and the critical current density for the film of Fig. 2. The inset shows in detail the  $j_c$  behavior close to  $T_c$ .

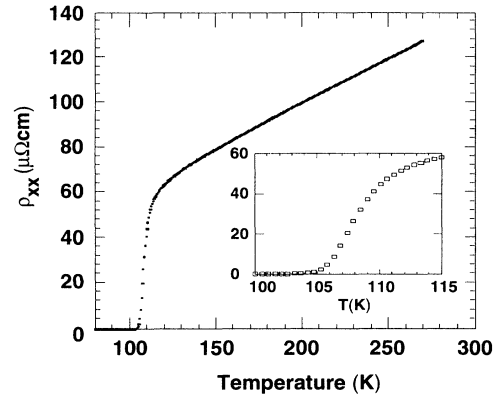


FIG. 4. Temperature variation of the in-plane resistivity  $\rho_{xx}$  of a 2223-BSCCO thin film in zero magnetic field. The inset shows the region of the superconducting transition on an expanded scale.

metal-like barriers between the superconducting 2223 grains, leading to the low value of  $j_c$  and its characteristic temperature dependence for superconductor-normal metal-superconductor junctions. The assumption of nonuniform phase distribution in our films was also confirmed by magnetically modulated microwave absorption measurements, which at temperatures 90–100 K revealed a film response characteristic for almost decoupled superconducting grains.<sup>60</sup> Below 80 K, the film  $j_c$  (Fig. 3) had a temperature dependence typical for an oriented high- $T_c$  film, reaching above  $2 \times 10^4 \text{ A}/\text{cm}^2$  at 4.2 K.

## B. Measurement techniques

All magnetotransport measurements were performed in a closed-cycle refrigerator with temperature controlled by a platinum resistor. Precision temperature measurements were made with a calibrated  $\text{Ga}_{1-x}\text{Al}_x\text{As}$  diode, placed close to the sample. The measurements were carried out with an ac current of 80-Hz frequency, using lock-in technique for the detection of the resistive voltage drop along the sample and for the Hall voltage. The current density was about  $800 \text{ A}/\text{cm}^2$ , calculated using the effective electrical thickness of the samples as discussed in Sec. III C.

The resistivity, the Hall effect, and the magnetoresistance were measured in subsequent runs on the same sample during a slow (0.02 K/min) downsweep of the temperature, which proved to yield more stable conditions than waiting for thermal equilibrium before taking data. Resistivity measurements were performed in the earth magnetic field; for the Hall effect and magnetoresistance measurements, which should be done in as low fields as possible, we used an electromagnet with  $B = 0.7 \text{ T}$ . The polarity of the magnetic field was reversed for every data point of the Hall effect and the magnetoresistance. The temperature accuracy was better than 10 mK.

## C. Results

An accurate knowledge of the physical sample dimensions is a prerequisite to determine the absolute values of

the various components of the magnetoconductivity tensor. The lateral dimensions are defined precisely by the patterning process, but the thickness of the film could only be estimated from the deposition conditions, since determination by mechanical probing turned out to be ambiguous due to surface roughness. In addition, we have to consider that thickness variations modify the film resistance and reduce the current-carrying cross section. To account for this problem, usually a  $C$  factor<sup>8</sup> is used to fit the data or the temperature derivative of the resistance is analyzed.<sup>6</sup> Alternatively, we have recently proposed a different approach,<sup>38</sup> by calculating an “effective thickness” from the comparison with single-crystal data. The effective-thickness method avoided the introduction of an additional fitting parameter and gave reliable results for YBCO thin films.

Since pure single crystals of the 2223 phase in BSCCO are hard to grow and no conductivity data is available, we compare the lowest values for  $\rho_{xx}(300\text{ K})=150\ \mu\Omega\text{ cm}$  in twinned YBCO single crystals<sup>61</sup> with the averaged  $a$ - $b$ -plane resistivity  $\rho_{xx}(300\text{ K})=140\ \mu\Omega\text{ cm}$  in the 2212 phase of BSCCO.<sup>62</sup> The similar values suggest that they are characteristic for the current transport along  $\text{CuO}_2$  planes. In fact, a comprehensive study on ceramic compounds with the nominal composition  $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_y$  with  $n=1,2,3$  revealed almost the same values for the room-temperature resistivity in the respective optimally doped compounds.<sup>63</sup> We, thus, concluded that the intrinsic room-temperature resistivity of the 2223 phase must be close to that “universal”  $\rho_{xx}(300\text{ K})=140\ \mu\Omega\text{ cm}$ , value and scaled our data accordingly. As a result, we found the effective thickness of our film to be 74 nm. The uncertainty introduced by this procedure affects only the absolute values of a few parameters, as most of our conclusions are based on the analysis of temperature dependencies.

In Fig. 4, the temperature variation of the resistivity  $\rho_{xx}$  of a 2223-BSCCO thin film in zero magnetic field is shown. Zero resistance is achieved below 105 K, the inflection point  $T_{ci}=108\text{ K}$ , and the 10–90% transition width of the superconducting transition is about 5 K. The curve exhibits a negative curvature from  $T_{ci}$  up to 180 K and flattens to a linear behavior at higher temperatures. In contrast to YBCO,<sup>38</sup> no upturn of the resistivity curve near room temperature was observed.

The transverse (Hall) resistivity  $\rho_{yx}=R_H B$  as a function of the temperature in a magnetic field  $B=0.7\text{ T}$  is shown in Fig. 5. The normal-state Hall effect is positive (holelike) and temperature dependent, as it is generally observed in most of HTS materials.<sup>64</sup> The room-temperature Hall coefficient  $R_H(300\text{ K})=1.4\times 10^{-4}\text{ cm}^3/\text{As}$  is about 1/4 of the value in YBCO.<sup>38</sup> The insert in Fig. 5 presents the temperature variation of the Hall number  $n_H=1/eR_H$ , which, in a conventional metal, would indicate the number of mobile carriers. Such an interpretation of  $n_H$  in HTS’s is, however, questionable due to their unusual temperature dependence of the Hall effect.

Above  $T_c$ ,  $\rho_{yx}$  exhibits a broad rounding, which we attribute to superconducting fluctuations. In the vortex state below  $T_c$ ,  $\rho_{yx}$  changes sign between 102 and 109 K.

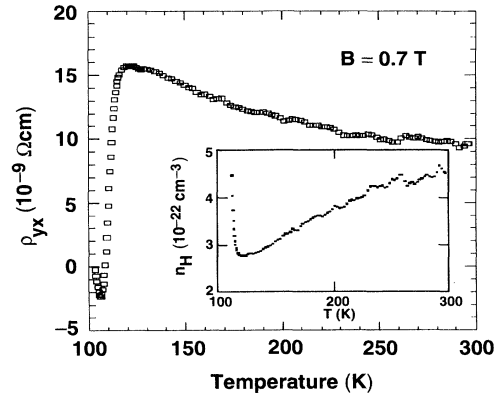


FIG. 5. Hall resistivity  $\rho_{yx}$  in 2223-BSCCO in a magnetic field  $B=0.7\text{ T}$  as a function of temperature. The inset shows the temperature variation of the Hall number  $n_H=1/eR_H$ .

The nature of this sign change, which is commonly observed in HTS’s, is presently unknown.

It has been pointed out by Anderson<sup>65</sup> that the key quantity for an analysis of the normal-state Hall effect is the Hall angle  $\tan\Theta_H^N=\rho_{yx}^N/\rho_{xx}^N=\sigma_{xy}^N/\sigma_{xx}^N$ , which exhibits a universal behavior in a variety of HTS systems, such as YBCO with various oxygen content,<sup>66,67</sup> YBCO with substitutions of magnetic atoms,<sup>68</sup> overdoped TBCCO,<sup>69,70</sup> and several others. The basic idea is the separation of the transport scattering time  $\tau_{tr}$  describing the relaxation of carrier motion normal to the Fermi surface from the Hall relaxation time  $\tau_H$  modeling the motion parallel to the Fermi surface. Anderson proposed two different quasiparticle excitations, namely “spinons” and “holons” with different relaxation times. Thus the electrical transport in zero magnetic field is governed by  $\tau_{tr}\propto T^{-1}$  due to scattering between holons and spinons. The spinons can interact with both a magnetic field and with magnetic impurities, yielding  $\tau_H\propto T^{-2}$  for the former process and a temperature-independent relaxation rate for the latter. Since  $\tau_{tr}$  cancels in the Hall angle, the universal expression in the above model is

$$\cot\Theta_H^N=AT^2+C, \quad (20)$$

where  $A$  is related to carrier density and spinon bandwidth and  $C$  is proportional to the impurity density.<sup>68</sup>

The dependence of the cotangent of the Hall angle as a function of  $T^2$  is shown in Fig. 6. The straight line represents a fit of the data to Eq. (20) in the temperature range from 130 to 185 K, using the parameters  $A=0.1855\text{ K}^{-2}$  and  $C=1429$ . Except for the temperature region close to  $T_c$ , where deviations from the linear relationship are attributed to the excess Hall effect, Eq. (20) describes the data remarkably well. At temperatures above 190 K, we observe a decrease of the slope  $A$ , in accordance with observations in YBCO.<sup>38,67</sup> Comparing the results for  $n_H$  and  $A$  with previous work on YBCO,<sup>38</sup> we realize that both the Hall number at room temperature and the slope of  $\cot\Theta_H^N$  are larger in 2223-BSCCO by about a factor of 4. This may indicate, assuming a similar spinon bandwidth in the two compounds, that the carrier density is substantially larger in the Bi-

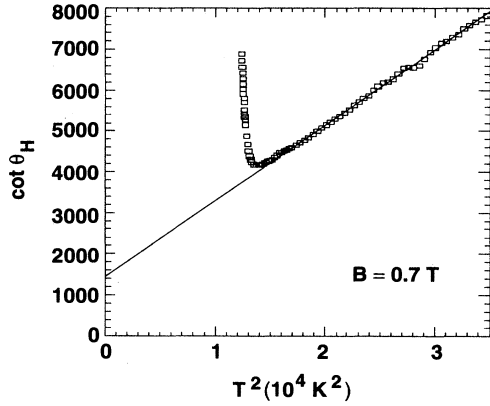


FIG. 6. Cotangent of the Hall angle  $\cot\theta_H$  as a function of  $T^2$ . The straight line represents a fit to Anderson's formula  $\cot\theta_H = \sigma_{xx}^N / \sigma_{xy}^N = AT^2 + C$  in the temperature range 130–185 K.

superconductor. Recently, a similar relation between the Hall angle slope and the carrier density was also verified in oxygen-depleted YBCO films.<sup>71</sup>

Finally, to complete our results, we present (Fig. 7) measurements of the transverse magnetoresistance (MR)  $[\rho_{xx}(B) - \rho_{xx}(0)] / \rho_{xx}(0)$  in a magnetic field  $B = 0.7$  T oriented parallel to the  $c$  axis. The MR spans a large range of magnitude in only a narrow temperature region, i.e., from 0.07 at 110 K to around  $10^{-5}$  at 130 K, where it drops below our experimental limits. The slope of the curve becomes remarkably steeper at  $T < 118$  K. Such a tremendous variation is far beyond any explanations, based on a conventional mechanism associated with the cyclotron orbits of charged carriers. In addition, using the relation  $\tan\theta_H \cong \omega_c \tau$  for a simple parabolic one-band model, where  $\omega_c$  is the cyclotron frequency and  $\tau$  is an average carrier scattering time, we may estimate an upper limit of the magnitude of the normal-state MR, viz.,  $[\rho_{xx}(B) - \rho_{xx}(0)] / \rho_{xx}(0) \leq (\omega_c \tau)^2$  and obtain  $6 \times 10^{-8}$  at 120 K, which is three orders of magnitude smaller than the observed value. We, thus, attribute the MR entirely to superconducting fluctuations.

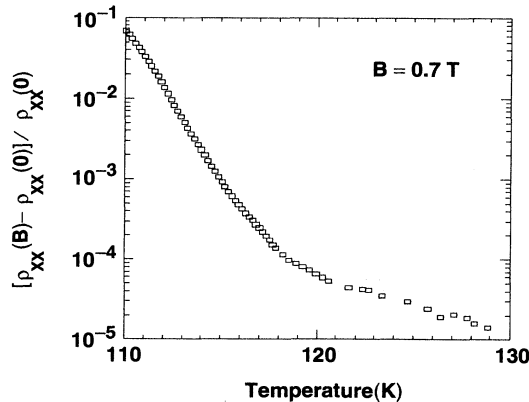


FIG. 7. Transverse magnetoresistance in 2223-BSCCO in a magnetic field  $B = 0.7$  T as a function of the temperature.

## IV. ANALYSIS AND DISCUSSION

### A. Paraconductivity

For an analysis of the paraconductivity we have to separate the normal-state contribution  $\sigma_{xx}^N(0)$  from the paraconductivity  $\Delta\sigma_{xx}(0)$ , according to Eq. (1). As mentioned in Sec. I, this is not a simple task, since no generally approved theoretical background for the temperature dependence of  $\sigma_{xx}^N(0)$  is available and the suppression of the fluctuation spectrum by the intense magnetic field is technically impossible in HTS's. On the other hand, a linear temperature dependence has been observed in a variety of cuprate materials in the optimum (highest- $T_c$ ) doping range. Since the anisotropy in BSCCO is considerably larger than in YBCO, we expect that in thin-film measurements any contributions from the  $c$ -axis resistivity are vanishingly small along the  $ab$  plane, and postulate a phenomenological relation  $\rho_{xx}^N(0) = AT + B$ . Actually we observed in our films such a behavior in the range from 180 to 273 K and determined the fitting parameters  $A = 4.01 \times 10^{-7} \text{ } \Omega \text{ cm K}^{-1}$  and  $B = 1.80 \times 10^{-5} \text{ } \Omega \text{ cm}$ . Although this procedure is widely used in literature, we point out that it naturally underestimates the paraconductivity at higher temperatures and may lead to a failure in the detection of the MT contribution.

The experimental values of the paraconductivity as a function of the reduced temperature are compared to the LD theory and to the  $T_c$ -distribution model in Fig. 8. A fit to Eq. (4) involves two adjustable parameters, i.e., the mean-field critical temperature  $T_c = 109.2$  K and the  $c$ -direction coherence length  $\xi_c(0) = 0.14$  nm.  $T_c$  is somewhat larger than  $T_{ci}$  indicating that the lower temperature tail of the transition curve arouses primarily from vortex motion. Actually, the exact value of  $T_c$  has little influence on the analysis of the paraconductivity a few kelvin above the transition. Similarly,  $\xi_c$  is only significant for low  $\epsilon$ , since at higher temperatures we

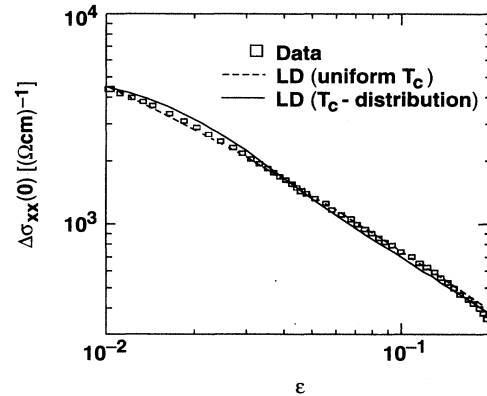


FIG. 8. Paraconductivity in a 2223-BSCCO thin film (squares) and the fits calculated for the LD contribution using a uniform  $T_c$  (broken line) and the  $T_c$ -distribution model (solid line). All data are plotted as a function of the reduced temperature  $\epsilon = \ln(T/T_c)$ . The parameters used are  $\xi_c(0) = 0.14$  nm,  $d = 1.85$  nm,  $T_c = 109.2$  K,  $\bar{T}_c = 108$  K,  $\delta T_c = 2.3$  K, and  $\epsilon_c = 2 \times 10^{-3}$ .



enter the 2D limit of the LD model, where the paraconductivity is entirely determined by the superconducting layer spacing  $d = c/2 = 1.85$  nm. Here  $c$  is the crystallographic unit cell parameter perpendicular to the  $\text{CuO}_2$  layers. Thus we emphasize that at  $\varepsilon \approx 0.1$ , the theoretical results are without any adjustable parameters, and from the different temperature dependencies in Eqs. (4) and (6), we may conclude that the MT process has no significant contribution to the paraconductivity in 2223-BSCCO. Additional support to the above conclusion also comes from the amplitude of the paraconductivity, which should be larger when MT fluctuations are present. Although this latter argument is frequently used, it must be regarded with some caution since it crucially depends on the choice of the normal-state background.

In Fig. 8, the excellent fit to the paraconductivity in the LD model seems to suggest an absence of an inhomogeneity of  $\delta T_c$ . For comparison, however, we have also presented in Fig. 8 the values for the paraconductivity (solid line) calculated, based on parameters that we will deduce later from the analysis of the excess Hall effect and the fluctuation magnetoresistance. We note that the  $T_c$ -distribution calculation also gives a fair description of the data. Thus, the paraconductivity analysis alone neither yields a very reliable determination of the essential parameters, nor provides a straightforward indication of critical temperature inhomogeneities in the sample.

### B. Excess Hall effect

For an analysis of the excess Hall effect, we again have to separate the normal-state contribution  $\sigma_{xy}^N$  from the fluctuation correction  $\Delta\sigma_{xy}$  according to Eq. (8). The procedure is analogous to the one for the paraconductivity and similar restrictions apply. Based on, at least phenomenological, validity of Eq. (20) for the normal-state Hall angle we calculate the off-diagonal normal-state conductivity  $\sigma_{xy}^N = (\rho_{xx}^N \cot\Theta_H^N)^{-1}$  from the fit in Fig. 6. The result is compared in Fig. 9 with the experimental data. Below 130 K, the Hall conductivity stays below the ex-

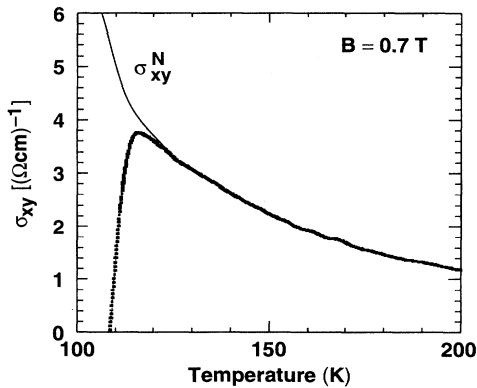


FIG. 9. The Hall conductivity  $\sigma_{xy} = (\rho_{xx} \cot\Theta_H)^{-1}$  as a function of the temperature. The solid line represents the normal-state contribution  $\sigma_{xy}^N$ , deduced from the fit in Fig. 6. The deviations from the extrapolated normal-state Hall conductivity are interpreted as fluctuation corrections to the Hall effect.

trapolated normal-state value, exhibits a maximum at about 117 K, and, at lower temperatures, rapidly fades. Thus,  $\Delta\sigma_{xy}$  is always negative and strongly temperature dependent. This observation is in contrast to the situation in YBCO, where a negative  $\Delta\sigma_{xy}$  was observed close to  $T_c$  and a positive  $\Delta\sigma_{xy}$  at higher temperatures.<sup>38,56,72</sup> Since Eq. (10) predicts that the MT contribution to the excess Hall effect is positive at any temperature, we conclude that the MT effect is negligible in 2223-BSCCO, supporting our previous findings from the paraconductivity analysis.

We now compare (Fig. 10)  $\Delta\sigma_{xy}$  with the calculations based on the UD model. At temperatures above 117 K ( $\varepsilon > 0.07$ ),  $\Delta\sigma_{xy} \propto \varepsilon^{-2}$  as anticipated from Eq. (9). Toward lower temperatures, however, the excess Hall effect raises to a steeper temperature dependence and exceeds the theoretical predictions (broken line) by a factor of 2. The theoretical values in Fig. 10 for the UD model were calculated, using the same parameters as previously for the paraconductivity, and, in addition, the electron-hole asymmetry parameter was  $\beta = -0.38$ . The calculation including an inhomogeneous  $T_c$  (solid line) provides a much better fit to the data, although it also somewhat underestimates the excess Hall effect in the vicinity of  $T_c$ . The shape of the curve, however, is very well reproduced by the  $T_c$ -distribution model of Eq. (18). We note, however, that the most crucial constraint for the parameters characterizing the distribution of  $T_c$ 's in the sample:  $\bar{T}_c$ ,  $\delta T_c$ , and  $\varepsilon_c$ , is the fit to the fluctuation magnetoconductivity presented later.

The excess Hall effect is governed by the electron-hole asymmetry parameter  $\beta$ . Since  $\beta < 0$ , the normal-state Hall angle is reduced close to  $T_c$  by superconducting fluctuations. It should be emphasized that the Hall effect itself remains positive above  $T_c$ , since  $\Delta\sigma_{xy}$  is only a small correction to  $\Delta\sigma_{xy}^N$ . Our previous calculations do not apply in the critical fluctuation region very close to  $T_c$ ,

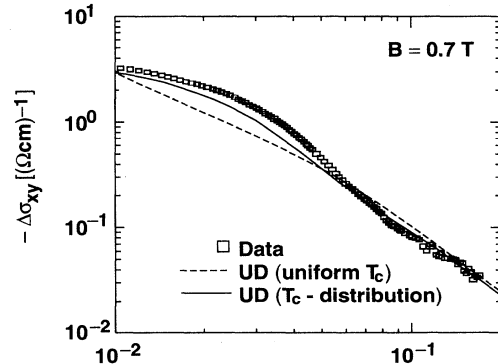


FIG. 10. Excess Hall effect in a 2223-BSCCO thin film (squares) as a function of the reduced temperature  $\varepsilon = \ln(T/T_c)$  and the fits calculated for the UD contribution assuming a uniform  $T_c$  (broken line) and a distribution of  $T_c$ 's (solid line) models. The excess Hall effect is negative in the entire temperature range. The same parameters as for the fit to the paraconductivity in Fig. 8 were used and, additionally,  $\xi_{ab}(0) = 1.6$  nm and the electron-hole asymmetry parameter  $\beta = -0.38$ .

where  $\Delta\sigma_{xy}$  becomes comparable to  $\Delta\sigma_{xy}^N$ . In a recent paper<sup>38</sup> we discussed a possible connection between a negative  $\beta$  and the occurrence of a negative Hall effect in the flux-flow region below  $T_c$ . Comparing the present data with those previous results, we note that  $\beta$  in 2223-BSCCO has the same (negative) sign and is approximately twice as large as in YBCO. Thus, in Bi-based materials, the imaginary part of the complex relaxation time in the time-dependent Ginzburg-Landau theory is larger,<sup>52</sup> yielding to a larger Hall effect due to vortex motion, according to Dorsey<sup>54</sup> and Kopnin, Ivlev, and Kalatsky.<sup>55</sup> In fact, the negative anomaly in the Hall effect is generally more pronounced in the bismuth cuprates.<sup>73</sup>

### C. Fluctuation magnetoconductivity

Finally, we present an analysis of the magnetoresistance results with regard to the superconducting fluctuation effect. From a comparison of Eqs. (11) and (14) and taking into account the previously determined parameters it can easily be seen that the Zeeman effect has no significance at the magnetic field  $B=0.7$  T, used in this study. Thus, we may confine the analysis to the orbital terms alone. Since no background subtraction procedure is necessary, we can directly calculate the fluctuation magnetoconductivity  $\Delta\sigma_B = -\rho_{xx}(B)^{-1}[\rho_{xx}(B) - \rho_{xx}(0)]/\rho_{xx}(0)$  from the magnetoresistance results.

In Fig. 11, the fluctuation magnetoconductivity is presented as a function of  $\epsilon$ . Above 118 K ( $\epsilon > 0.08$ ),  $\Delta\sigma_B \propto \epsilon^{-3}$ , in accordance with the 2D limit of the ALO contribution in Eq. (11). Since a significant MTO contribution would inevitably change the temperature dependence of the magnetoconductivity, results in Fig. 11 are another evidence of a negligible MT fluctuation contribution in 2223-BSCCO. An estimation of the largest MT contribution, which is still consistent with our experimental data, yields for the pair-breaking parameter  $\delta > 1.0$  in Eq. (13).

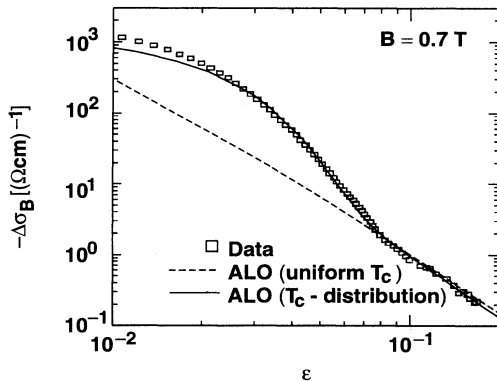


FIG. 11. Magnetoconductivity  $\Delta\sigma_B$  as a function of the reduced temperature  $\epsilon = \ln(T/T_c)$  of a 2223-BSCCO thin film (squares) in a magnetic field  $B=0.7$  T. The broken line represents a fit to the ALO contribution of the fluctuation magnetoconductivity using an uniform  $T_c$  and the solid line a respective fit to the  $T_c$ -distribution model with the same parameters as in Figs. 8 and 10.

Similar to the excess Hall effect, but even more pronounced, there is a tremendous difference between our experimental data and the ALO model (broken line) at temperatures below 118 K. The data exceeds the ALO contribution of Eq. (11) up to a factor of 8, which again was calculated using the same parameters as in the paraconductivity and excess Hall-effect fits. A similar observation has been also reported for the magnetoconductivity of TBCCO thin films<sup>74</sup> but was absent in several studies performed on YBCO single crystals<sup>32-34,61</sup> and thin films.<sup>35,36</sup> It should be mentioned, however, that the anomaly decreases in higher magnetic fields<sup>74,75</sup> and our investigations were performed in  $B$  lower than in any other work. A corresponding anomaly has been also detected in the fluctuation diamagnetism of 2212 and 2223 BSCCO.<sup>76</sup>

We note that the  $T_c$ -distribution model (solid line in Fig. 11) provides an excellent fit to the data over a wide temperature range. Only very close to  $T_c$  a small discrepancy can be observed, resulting from the cutoff procedure in Eq. (17), which somewhat underestimates the amplitude of local fluctuations. It is noteworthy to mention that the observed anomaly of the magnetoresistance exhibits a distinct onset and that the slope is significantly changed from  $\epsilon^{-3}$  (in the 2D limit of the ALO term) to approximately  $\epsilon^{-5.3}$ . Changing in the calculations the width of the distribution function  $\delta T_c$  does not significantly alter the slope of the anomaly, but rather moves its onset. Consequently, the enhanced temperature dependence of the magnetoconductivity provides a fingerprint of an inhomogeneous  $T_c$  in a given sample.

### V. CONCLUSIONS AND SUMMARY

There are several conclusions that can be drawn from our combined investigations of the paraconductivity, the excess Hall effect, and the fluctuation magnetoconductivity in 2223-BSCCO.

(1) We were able to fit the paraconductivity, the excess Hall effect, and the fluctuation magnetoconductivity with a *consistent* set of parameters, resolving this way some ambiguities, introduced by the extrapolated procedures used for the normal state resistivity and the Hall angle. Simultaneously, the agreement with the magnetoconductivity results supports our method for estimating the BSCCO normal-state properties close to  $T_c$ .

(2) It is impossible to achieve a consistent fit to our results in the framework of the conventional fluctuation theories. To illustrate this we have replotted in Fig. 12 the data of Figs. 8, 10, and 11, together with fits to Eqs. (4), (9), and (11) restricted to the temperature region where the anomalies in the Hall conductivity and the magnetoconductivity were observed. It can be noted that data deviate from the fits at  $\epsilon > 0.08$  and, most striking, there is no agreement in the paraconductivity. Additionally, we had to assume  $T_c = 112$  K, which seems unlikely high. On the other hand, considering a Gaussian distribution of  $T_c$ 's resolves the discrepancies without a change to the essential parameters  $\xi_{ab}$ ,  $\xi_c$ , and  $\beta$ .

(3) Using our model for inhomogeneous  $T_c$ , we also estimated the possible influence of a 10% fraction of the

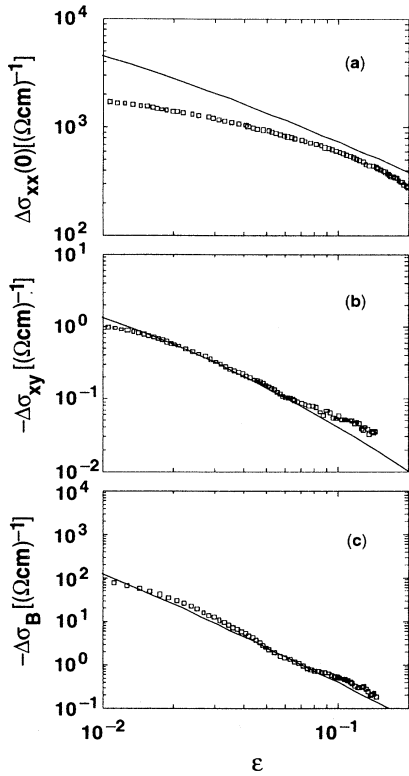


FIG. 12. Comparison of the (a) paraconductivity, (b) excess Hall effect, and (c) magnetoconductivity with the fits to the conventional fluctuation theories focused on the anomalies near  $T_c$ . The lines represent the calculations, using the parameters  $T_c = 112$  K,  $\xi_c(0) = 0.14$  nm,  $\xi_{ab}(0) = 1.3$  nm, and  $\beta = -0.16$ .

2212 phase ( $T_c = 80$  K) in our samples on the fluctuation spectrum of the dominant 2223 phase, and found no changes, except for a small (approx. 9%) enhancement of  $\xi_{ab}$  and 8% increase of  $\beta$  in the fits. Thus, small amounts of the 2212 phase are irrelevant for the analysis of fluctuations in BSCCO samples superconducting above 100 K.

(4) From the LD theory and Eq. (4), the crossover temperature  $\varepsilon_0 = \ln(T_0/T_c)$  from 2D to 3D superconducting behavior in the vicinity of  $T_c$  can be estimated,  $\varepsilon_0 = 2\xi_c^2(0)/d^2$ . Thus, 2223-BSCCO exhibits essentially 2D properties at temperatures  $|T - T_c| > 1.3$  K. We emphasize, however, that a determination of  $\xi_c$  in the LD model, which is dependent on the change of slope near  $T_c$ , is only approximate due to a distinct influence of an inhomogeneous  $T_c$  in that temperature region. Thus, recent results that indicate a pure 2D behavior and an absence of the crossover<sup>11,12,15,19</sup> or a considerable lower  $\varepsilon_0$  (Refs. 13 and 17) have to be cautioned.

(5) If we compare the physical parameters deduced

from our results to the previous work, we recognize that  $\xi_c(0) = 0.14$  nm is similar to the value for YBCO (Ref. 38), indicating that the three neighboring  $\text{CuO}_2$  layers in 2223-BSCCO are coupled together, acting as a joint superconducting sheet. The in-plane coherence length  $\xi_{ab}(0) = 1.6$  nm is somewhat larger than in YBCO (Refs. 32, 35, and 61) and close to a recent estimation from the upper critical field slope in 2212-BSCCO.<sup>77</sup>

(6) Now we turn to the additional parameters deduced from the  $T_c$ -distribution model. From a comparison of  $T_c = 109.2$  K and  $\bar{T}_c = 108$  K we see that the high-temperature side of the  $T_c$ -distribution dominates the fluctuations, resulting in an apparent higher  $T_c$  in the fits to the conventional models. The width of the distribution,  $2\delta T_c = 4.6$  K is close to the 10–90% transition width of the resistivity. Finally, using the argument for the introduction of the low-temperature cutoff, we estimate a typical length scale of the  $T_c$  variation to be about 36 nm, which is 67 times the width of the elementary cell along the  $a$  direction, but is simultaneously significantly lower than a diameter of an average crystallite in our films.

(7) Contrary to investigations in YBCO,<sup>31,32,35,38,56</sup> we find no evidence for the MT process in 2223-BSCCO, although a rather small contribution cannot be ruled out from the data. The most straightforward conclusion comes from the excess-Hall-effect data, where a significant MT process should be easily detectable due its different sign. Since the basic structural element, the  $\text{CuO}_2$  plane, is the same in YBCO and BSCCO we do not expect enhanced pair breaking due to magnetic impurities in BSCCO. The MT contribution may be suppressed by a larger impurity density, such a behavior, however, would be a distinct indication for non- $s$ -wave superconductivity according to Yip.<sup>30</sup>

In summary, we have presented a comprehensive study of superconducting fluctuations in 2223-BSCCO probed by magnetotransport measurements. A complementary investigation of the paraconductivity, the excess Hall effect, and the magnetoconductivity allowed us for an unambiguous determination of both the  $c$ -axis and in-plane coherence lengths, and the electron-hole asymmetry parameter in the time-dependent Ginzburg-Landau theory. Anomalies of the excess Hall effect and the magnetoconductivity are shown to be caused by inhomogeneities of the critical temperature of our sample.

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- <sup>1</sup>For a review, see W. J. Skocpol and M. Tinkham, *Rep. Prog. Phys.* **38**, 1049 (1975).
- <sup>2</sup>For a review, see M. Ausloos, S. K. Patapis, and P. Clippe, in *Physics and Materials Science of High Temperature Superconductors II*, edited by R. Kossowsky, B. Raveau, D. Wohlleben, and S. K. Patapis (Kluwer, Dordrecht, 1992), p. 755.
- <sup>3</sup>For a review, see Y. Iye, in *Physical Properties of High Temperature Superconductors III*, edited by D. M. Ginsberg (World Scientific, Singapore, 1992), p. 285.
- <sup>4</sup>L. G. Aslamazov and A. I. Larkin, *Sov. Phys. Solid State* **10**, 875 (1968).
- <sup>5</sup>W. E. Lawrence and S. Doniach, in *Proceedings of the 12th International Conference on Low Temperature Physics, Kyoto, 1970*, edited by E. Kanda (Keigaku, Tokyo, 1971), p. 361.
- <sup>6</sup>M. Ausloos and Ch. Laurent, *Phys. Rev. B* **37**, 611 (1988).
- <sup>7</sup>S. J. Hagen, Z. Z. Wang, and N. P. Ong, *Phys. Rev. B* **38**, 7137 (1988).
- <sup>8</sup>B. Oh, K. Char, A. D. Kent, M. Naito, M. R. Beasley, T. H. Geballe, R. H. Hammond, A. Kapitulnik, and J. M. Graybeal, *Phys. Rev. B* **37**, 7861 (1988).
- <sup>9</sup>P. P. Freitas, C. C. Tsuei, and T. S. Plaskett, *Phys. Rev. B* **36**, 833 (1987).
- <sup>10</sup>F. Vidal, J. A. Veria, J. Maza, J. J. Ponte, F. Garcia-Alvarado, E. Moran, J. Amador, C. Cascales, A. Castro, M. T. Casais, and I. Rasines, *Physica C* **156**, 807 (1988).
- <sup>11</sup>G. Balestrino, A. Nigro, R. Vaglio, and M. Marinelli, *Phys. Rev. B* **39**, 12 264 (1989).
- <sup>12</sup>V. V. Gridin, T. W. Krause, and W. R. Datars, *J. Appl. Phys.* **68**, 675 (1990).
- <sup>13</sup>J. A. Veira and F. Vidal, *Phys. Rev. B* **42**, 8748 (1990).
- <sup>14</sup>P. Mandal, A. Poddar, A. N. Das, B. Gosh, and P. Choudhury, *Physica C* **169**, 43 (1990).
- <sup>15</sup>S. Ravi and V. S. Bai, *Physica C* **182**, 345 (1991).
- <sup>16</sup>G. Balestrino, M. Marinelli, E. Milani, L. Reggiani, R. Vaglio, and A. A. Varlamov, *Phys. Rev. B* **46**, 14 919 (1992).
- <sup>17</sup>R. Ivens, R. Wernhardt, M. Rosenberg, S. Löscher, and S. Kemmler-Sack, *Supercond. Sci. Technol.* **5**, 16 (1992).
- <sup>18</sup>P. Mandal, A. Poddar, P. Choudhury, J. B. Mandal, S. Keshri, A. N. Das, and B. Gosh, *Indian J. Pure Appl. Phys.* **30**, 531 (1992).
- <sup>19</sup>S. Labdi, S. Megtert, and H. Raffy, *Solid State Commun.* **85**, 491 (1993).
- <sup>20</sup>V. Calzona, M. R. Cimberle, C. Ferdeghini, G. Grasso, D. V. Livanov, D. Marré, M. Putti, A. S. Siri, G. Balestrino, and E. Milani, *Solid State Commun.* **87**, 397 (1993).
- <sup>21</sup>D. H. Kim, A. M. Goldman, J. H. Kang, K. E. Gray, and R. T. Kampwirth, *Phys. Rev. B* **39**, 12 275 (1989).
- <sup>22</sup>H. M. Duan, W. Kiehl, C. Dong, A. W. Cordes, M. J. Saeed, D. L. Viar, and A. M. Hermann, *Phys. Rev. B* **43**, 12 925 (1991).
- <sup>23</sup>K. Winzer, G. Kumm, P. Maab, H. Thomas, E. Schwarzmann, A. Aghaie, and F. Ladenberger, *Ann. Phys.* **1**, 479 (1992).
- <sup>24</sup>M. Ausloos, P. Clippe, and Ch. Laurent, *Phys. Rev. B* **41**, 9506 (1990).
- <sup>25</sup>K. Maki, *Prog. Theor. Phys.* **39**, 897 (1968).
- <sup>26</sup>R. S. Thompson, *Phys. Rev. B* **1**, 327 (1970).
- <sup>27</sup>E. Abrahams and J. W. F. Woo, *Phys. Lett.* **27A**, 117 (1968); *A. Schmid, Z. Phys.* **215**, 210 (1968).
- <sup>28</sup>T. R. Lemberger and L. Coffey, *Phys. Rev. B* **38**, 7058 (1988).
- <sup>29</sup>S. Hikami and A. I. Larkin, *Mod. Phys. Lett. B* **2**, 693 (1988).
- <sup>30</sup>S. K. Yip, *Phys. Rev. B* **41**, 2612 (1990).
- <sup>31</sup>M. Ausloos, F. Gillet, Ch. Laurent, and P. Clippe, *Z. Phys. B* **84**, 13 (1991).
- <sup>32</sup>K. Semba, T. Ishii, and A. Matsuda, *Phys. Rev. Lett.* **67**, 769 (1991).
- <sup>33</sup>M. Hikita and M. Suzuki, *Phys. Rev. B* **41**, 834 (1990).
- <sup>34</sup>N. Overend and M. A. Howson, *J. Phys. Condens. Matter* **4**, 9615 (1992).
- <sup>35</sup>Y. Matsuda, T. Hirai, S. Komiyama, T. Terashima, Y. Bando, K. Iijima, K. Yamamoto, and K. Hirata, *Phys. Rev. B* **40**, 5176 (1989).
- <sup>36</sup>J. Sugawara, H. Iwasaki, N. Kobayashi, H. Yamane, and T. Hirai, *Phys. Rev. B* **46**, 14 818 (1992).
- <sup>37</sup>J. P. Rice and D. M. Ginsberg, *Phys. Rev. B* **46**, 12 049 (1992).
- <sup>38</sup>W. Lang, G. Heine, P. Schwab, X. Z. Wang, and D. Bäuerle, *Phys. Rev. B* **49**, 4209 (1994), and references therein.
- <sup>39</sup>W. Lang, W. Kula, and R. Sobolewski, *Physica B* **194-196**, 1643 (1994).
- <sup>40</sup>A. G. Aronov, S. Hikami, and A. I. Larkin, *Phys. Rev. Lett.* **62**, 965 (1989); **62**, 2336(E) (1989).
- <sup>41</sup>K. Maki and R. S. Thompson, *Phys. Rev. B* **39**, 2767 (1989).
- <sup>42</sup>J. B. Bieri and K. Maki, *Phys. Rev. B* **42**, 4854 (1990).
- <sup>43</sup>J. B. Bieri, K. Maki, and R. S. Thompson, *Phys. Rev. B* **44**, 4709 (1991).
- <sup>44</sup>K. Kadowaki, J. N. Li and J. J. M. Franse, *J. Magn. Magn. Mater.* **90&91**, 678 (1990).
- <sup>45</sup>C. M. Fu, W. Boon, Y. S. Wang, V. V. Moschalkov, and Y. Bruynseraede, *Physica C* **200**, 17 (1992).
- <sup>46</sup>H. Fukuyama, H. Ebisawa, and T. Tsuzuki, *Prog. Theor. Phys.* **46**, 1028 (1971).
- <sup>47</sup>J. Maza and F. Vidal, *Phys. Rev. B* **43**, 10 560 (1991).
- <sup>48</sup>W. Lang, *Physica C* **226**, 267 (1994).
- <sup>49</sup>R. A. Craven, G. Thomas, and R. D. Parks, *Phys. Rev. B* **4**, 2185 (1971).
- <sup>50</sup>R. Balian and N. R. Werthamer, *Phys. Rev.* **131**, 1553 (1963).
- <sup>51</sup>E. Abrahams, R. E. Prange, and M. J. Stephen, *Physica* **55**, 230 (1971).
- <sup>52</sup>S. Ullah and A. T. Dorsey, *Phys. Rev. B* **44**, 262 (1991).
- <sup>53</sup>A. A. Varlamov and D. V. Livanov, *Zh. Eksp. Teor. Fiz.* **99**, 1816 (1991) [*Sov. Phys. JETP* **72**, 1016 (1991)].
- <sup>54</sup>A. T. Dorsey, *Phys. Rev. B* **46**, 8376 (1992).
- <sup>55</sup>N. B. Kopnin, B. I. Ivlev, and V. A. Kalatsky, *J. Low. Temp. Phys.* **90**, 1 (1993).
- <sup>56</sup>J. P. Rice, J. Giapintzakis, D. M. Ginsberg, and J. M. Mochel, *Phys. Rev. B* **44**, 10158 (1991).
- <sup>57</sup>W. Kula, R. Sobolewski, J. Gorecka, and S. J. Lewandowski, *J. Appl. Phys.* **70**, 3171 (1991).
- <sup>58</sup>P. H. Ballentine, A. M. Kadin, M. A. Fisher, D. S. Mallory, and W. R. Donaldson, *IEEE Trans. Magn.* **MAG-25**, 950 (1989).
- <sup>59</sup>The values of  $j_c$  were calculated assuming the film thickness of  $1 \mu\text{m}$ , as measured for the as-deposited film. For discussion on the thickness of annealed BSCCO films, see Sec. III C.
- <sup>60</sup>W. Kula and R. Sobolewski, *IEEE Trans. Appl. Supercond.* **AS-3**, 1446 (1993).
- <sup>61</sup>K. Winzer and G. Kumm, *Z. Phys. B* **82**, 317 (1991).
- <sup>62</sup>S. Martin, A. T. Fiory, R. M. Fleming, L. F. Schneemeyer, and J. V. Waszczak, *Phys. Rev. Lett.* **60**, 2194 (1988).
- <sup>63</sup>A. Maeda, M. Hase, I. Tsukada, K. Noda, S. Takebayashi, and K. Uchinokura, *Phys. Rev. B* **41**, 6418 (1990).
- <sup>64</sup>For a review, see N. P. Ong, in *Physical Properties of High Temperature Superconductors II*, edited by D. M. Ginsberg (World Scientific, Singapore, 1990), p. 459.
- <sup>65</sup>P. W. Anderson, *Phys. Rev. Lett.* **67**, 2092 (1991).
- <sup>66</sup>J. M. Harris, Y. F. Yan, and N. P. Ong, *Phys. Rev. B* **46**,

- 14 293 (1992).
- <sup>67</sup>B. Wuyts, E. Osquiquil, M. Maenhoudt, S. Libbrecht, Z. X. Gao, and Y. Bruynseraede, *Phys. Rev. B* **47**, 5512 (1993).
- <sup>68</sup>T. R. Chien, Z. Z. Wang, and N. P. Ong, *Phys. Rev. Lett.* **67**, 2088 (1991).
- <sup>69</sup>Y. Kubo, Y. Shimakawa, T. Manako, and H. Igarashi, *Phys. Rev. B* **43**, 7875 (1991).
- <sup>70</sup>P. S. Wang, J. C. Williams, D. Rathanyaka, B. Hennings, D. G. Naugle, and A. B. Kaiser, *Phys. Rev. B* **47**, 1119 (1993).
- <sup>71</sup>B. Wuyts, E. Osquiquil, M. Maenhoudt, S. Libbrecht, Z. X. Gao, and Y. Bruynseraede, *Physica C* **222**, 341 (1994).
- <sup>72</sup>A. V. Samoilov, *Pis'ma Zh. Eksp. Teor. Fiz.* **57**, 43 (1993) [*JETP Lett.* **57**, 44 (1993)]; *Phys. Rev. B* **49**, 1246 (1994).
- <sup>73</sup>N. V. Zavaritsky, A. V. Samoilov, and A. A. Yurgens, *Physica C* **180**, 417 (1991).
- <sup>74</sup>D. H. Kim, K. E. Gray, R. T. Kampwirth, and D. M. McKay, *Phys. Rev. B* **43**, 2910 (1991).
- <sup>75</sup>W. Lang, *Z. Phys. B* (to be published).
- <sup>76</sup>W. C. Lee, J. H. Cho, and D. C. Johnston, *Phys. Rev. B* **43**, 457 (1991).
- <sup>77</sup>H. C. Ri, R. Gross, F. Gollnik, A. Beck, and R. P. Huebener, *Phys. Rev. B* **50**, 3312 (1994).