Surface resistance of epitaxial $YBa_2Cu_3O_{7-x}$ films on various substrates: Effects of pair condensation and quasiparticle scattering

Matthias Hein, Thomas Kaiser, and Günter Müller
Department of Physics, University of Wuppertal, D-42097 Wuppertal, Germany
(Received 30 June 1999)

The temperature dependent surface resistance $R_s(T)$ of eight high-quality epitaxial $YBa_2Cu_3O_{7-x}$ films on $LaAlO_3$(Y/L), MgO (Y/M), and CeO_2-buffered sapphire (Y/S) substrates was investigated at 19 GHz with a resolution of 20 $\mu\Omega$. The residual level $R_s(T\rightarrow 0)$ was (90–180) $\mu\Omega$ for all films. The slope $\partial R_s(T)/\partial T$ in the range (4–20) K decreased from (6–8) $\mu\Omega$/K for Y/L to (0–2) $\mu\Omega$/K for Y/M, and to slightly negative values for Y/S. This slope correlated with the transition temperature T_c, including published data of about (10–20) $\mu\Omega$/K for high-purity $YBa_2Cu_3O_{7-x}$ single crystals. The increase of R_s from 4.2 showed power-law behavior $\Delta R_s(T)\propto T^a$ up to 40 K with $a\approx 1.3$ for Y/L and Y/M. However, it was exponential for Y/S, $\Delta R_s(T)\propto \exp(-\delta\times T_c/T)$ with $\delta\approx 0.8$. The results can be described with the two-fluid model in terms of the quasiparticle density $n_N(T)$ and the scattering time $\tau(T)$. The high and reproducible residual resistance implies (magnetic) impurity scattering at a rate $\tau^{-1}(0)\propto n_N(0)$, i.e., proportional to the density of quasiparticles. In crystals and unstrained films, the pair condensation is gapless, and the power-law temperature dependences of τ and n_N are reflected in that of R_s. In contrast, in strained films, $n_N(T)$ displays activated behavior, and the low-temperature behavior of R_s changes to exponential. The formation of an energy gap is attributed to the interaction between the Cu-O planes and the chains, which is affected by strain. The existence of an energy range with zero density of states limits possible interpretations of the order parameter of YBaCuO. A two-band scenario with magnetic pairbreaking and two different order parameters ($2\Delta/kT_c=6-8$ for the planes and $\geqslant 0$ for the chains) with *s*-wave symmetry, at least in the chains, appears to be an adequate explanation.

I. INTRODUCTION

Until now, the pairing mechanism and the symmetry of the order parameter in high-temperature superconductors (HTS's) are not unambiguously resolved. Many experimental results such as a finite surface resistance $R_s(T\ll T_c)$, power-law behavior of the penetration depth $\lambda(T)$ at low temperatures,[1–5] spontaneous magnetization,[6–8] and anomalous Josephson behavior[9,10] revealed an unconventional character of the HTS materials. Phenomenological explanations of these and other observations assumed a *d*-wave symmetry of the superconducting order parameter. However, regarding the high transition temperatures, the required pairing mechanism consistent with this symmetry is presently not clear.[11] Furthermore, the observation of Cooper pair tunneling along the crystalline *c* direction proved the existence of a nonvanishing *s*-wave contribution to the pair state.[12–14] An alternative explanation of the unconventional behavior of HTS's was based on a two-band model.[15–17] This model considers two electronic subsystems, represented by the Cu-O planes and the charge reservoirs between these planes which, in the case of YBaCuO, are the Cu-O chains. Strong electron-phonon interaction of the in-plane carriers[18–20] and induced superconductivity in the reservoirs lead to two order parameters which could be of conventional, unconventional, or mixed symmetry. This model accounts for the crystal structure of the HTS compounds and is able to describe many experimental observations. A specific feature of the two-band model is intrinsic magnetic pairbreaking, which potentially leads to spontaneous magnetization at interfaces and to gapless superconductivity in the charge reservoirs of HTS's.[21,22]

According to the pairing mechanism and the symmetry of the *order parameter*, the energy dependent density of states $N(E)$ of HTS has been discussed controversially. Most models involving *d*-wave symmetry imply that $N(E)$, averaged over all directions across the Fermi surface, vanishes at $E=0$ similar to E^a with $a=1-2$.[2,7] In contrast, the existence of an *energy gap* $|\Delta|$, i.e., where $N(E)\equiv 0$ for an extended energy range $E<Re(\Delta)$, is unambiguously related to *s*-wave symmetry and is well known to result in thermally activated charge transport in terms of the reduced energy gap $\delta=|\Delta|/k_BT_c$.[18,23] In turn, the observation of exponential temperature dependences must be considered a proof against gapless superconductivity as in most *d*-wave scenario. It is worth noting that early reports indeed claimed an exponential temperature dependence of the surface resistance of well oxygenated epitaxial YBaCuO films and of single crystals.[24–26] However, many surface impedance data on HTS single crystals did not reveal such features later on.[2]

We report recent studies of the surface resistance $R_s(T)$ of high-quality epitaxial YBaCuO films on different substrates, part of which displayed a clear exponential temperature dependence of R_s. Based on an improved measurement technique and on statistical reliability, these data present an important confirmation of the earlier observations. They also provide deeper insight into the mechanisms which lead to the formation of an energy gap. These mechanisms are related to the crystalline perfection of the epitaxial films and hence to the type of substrates employed. The results bear important

 ©2000 The American Physical Society

TABLE I. Survey of selected properties of the investigated YBaCuO films. The sequence of the samples corresponds to the curves in Fig. 1(a), from top to bottom. The thermally evaporated film on $LaAlO_3$ (Y/L2) and all films on sapphire (Y/S1–3) were deposited after the substrate was covered with a CeO_2 buffer layer. The deposition techniques are explained in the text. The residual resistance and the slope $\partial R_s/\partial T$ were extracted from quadratic least-squares fits in the range (4–20) K as indicated in Fig. 1(b).

Sample ID	Substrate	Deposition technique	Film thickness d_F, nm	Critical temperature T_c, K	Residual resistance $R_s(0)$, $\mu\Omega$	Slope $\partial R_s/\partial T$, $\mu\Omega$/K
Y/L 1	$LaAlO_3$	LD[28]	260	90.1	165	6.0
Y/L 2	$LaAlO_3$	TC[32]	400	>87	107	8.2
Y/L 3	$LaAlO_3$	DS[30]	330	89.6	88	6.6
Y/M 1	MgO	EC[31]	350	89.9	108	2.4
Y/M 2	MgO	EC[31]	350	92.0	127	0.0
Y/S 1	Sapphire	TC[32]		86.4	187	−3.3
Y/S 2	Sapphire	MS[29]	400	87.9	147	−3.4
Y/S 3	Sapphire	MS[29]	550	87.5	115	−2.0

constraints for possible interpretations of the pair state in HTS's. Section II describes the microwave measurement technique and the investigated films. The results for the temperature dependence of R_s are summarized in Sec. III, and discussed in terms of a two-fluid model in Sec. IV. The paper is concluded in Sec. V by a discussion of the major findings.

II. EXPERIMENTAL TECHNIQUES

The surface resistance of the investigated YBaCuO films was measured at 19 GHz using a Nb-shielded dielectric resonator excited in the edge-current free TE_{011} mode.[27] The dielectric was a high-purity sapphire with a loss-tangent below 4×10^{-8} at 4.2 K. The unloaded quality factor of the host resonator, when terminated with a pure Nb sample, exceeded 3×10^7 at 2 K, indicating residual losses equivalent to a surface resistance of as low as 20 $\mu\Omega$. The sample under investigation was thermally and electrically isolated by a (100–300) μm wide gap from the sapphire and the Nb shield, which were both permanently kept at 4.2 K. Heating the Cu support of the HTS film allowed us to study the temperature dependence of its surface resistance with high precision up to temperatures close to T_c. The microwave antennas into and out of the host resonator were adjusted to low coupling factors <1%, and the source was kept at the low power level of −10 dBm during such measurements. The surface resistance of the sample was deduced from the quality factor Q of the resonator, including the corrections for the (weak) coupling. The $Q(T)$ data were acquired with a computer controlled network analyzer during warming the sample from 4.2 K up to T_c within typically five hours. Further details and representative results obtained with this system were described previously.[27]

Three groups of high-quality epitaxial YBaCuO films were investigated in the present study (see Figs. 1 and 3). Group 1 consisted of three films deposited onto CeO_2-buffered sapphire substrates (referred to as Y/S) from two suppliers (see references below and in Table I). Group 2 contained two films on MgO (Y/M) from a single supplier, and group 3 three films on $LaAlO_3$(Y/L) from three different suppliers. Selected properties of all samples are summarized in Table I. The films were prepared by pulsed laser ablation[28] (LD), magnetron sputtering[29] (MS), high oxygen-pressure dc sputtering[30] (DS), electron beam coevaporation[31] (EC), and thermal coevaporation[32] (TC). The EC samples covered an area of 1 in. by 1 in., while the other films were 2 in. in diameter. All films, except for sample Y/M2 which was near optimal oxygen doping, were slightly overdoped ($x\leqslant0.12$) $YBa_2Cu_3O_{7-x}$ of correct (within 3%) cation stoichiometry. The thickness d_F of the films varied between 260 and 550 nm, corresponding to a ratio $d_F/\lambda(0)$ of about 1.5–3. The resulting film thickness correction of the surface resistance remains below 10% up to temperatures about $T\approx T_c/2$, above which $\lambda(T)$ starts to increase appreciably.[33] This correction could therefore be safely neglected for the following analysis, given the lack of sufficiently precise $\lambda(T)$ data for the investigated films. The critical temperatures of the films were inductively measured, or deduced from the surface resistance data, with a resulting uncertainty of 0.5 K. The $R_s(0)$ and $\partial R_s/\partial T$ data were extracted from quadratic least-squares fits in the temperature range between 4 and 20 K as discussed below [see Fig. 1(b)].

III. TEMPERATURE DEPENDENCE OF THE SURFACE RESISTANCE

A. Residual resistance and slope $\partial R_s/\partial T$

Figure 1(a) compares the as-measured $R_s(T)$ data of the investigated YBaCuO films in the temperature range from 4.2 to 40 K. The data for Y/M and Y/L were offset by +100 $\mu\Omega$ and +200 $\mu\Omega$ for clarity, as indicated by the arrows. At 4.2 K, all films displayed surface resistance values around 150 $\mu\Omega$, independent of the type of substrate. This low level, which corresponds to 40 $\mu\Omega$ if scaled quadratically with frequency to 10 GHz, is indicative of a high sample quality, and is comparable to published data on high-purity single crystals.[34] However, the $R_s(T)$ data can be distinguished from the steepness with which the surface resistance increases at $T>4.2$ K. The Y/S films display almost constant R_s up to about 15 K, and a weak increase at higher temperatures. The surface resistance of the Y/M films starts to increase above 4.2 K with a steeper slope and with significant curvature, while the $R_s(T)$ data of the Y/L films increase still

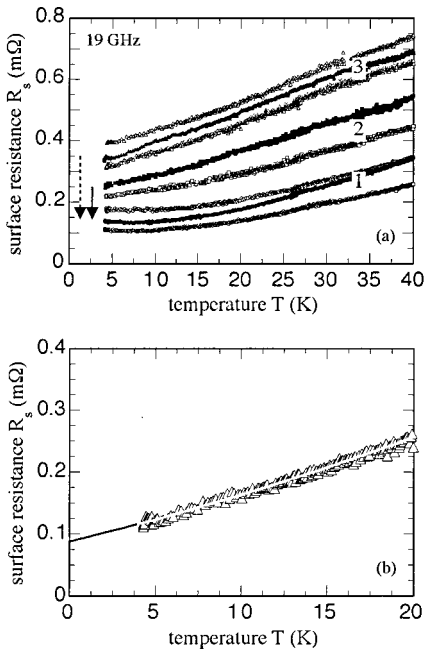

FIG. 1. (a) Temperature dependence of the surface resistance of the YBaCuO films listed in Table I: Y/S (group 1, circles, three different samples), Y/M (group 2, squares, two samples), Y/L (group 3, triangles, three samples). The arrows indicate an offset of 0.1 mΩ (solid line, for Y/M) and of 0.2 mΩ (dashed, for Y/L). (b) Selected data for sample Y/L3 in the temperature range (4–20) K (symbols) together with a quadratic least-squares fit (line).

more steeply. These features are independent of deposition technique and film thickness, as can be concluded from comparing the different films within each group, and can therefore be correlated with the type of substrates. Similar results were recognized recently also for data communicated in a commercial advertisement, and can hence be considered representative.[35,36]

In order to evaluate the substrate dependent behavior quantitatively, the $R_s(T)$ curves in Fig. 1(a) were parameterized in the low-temperature range (4–20) K by the polynomial

$$R_s(T) = R_s(0) + \frac{\partial R_s}{\partial T} \times T + O(T^2). \tag{1}$$

The lowest order required to reproduce the curvature of the $R_s(T)$ curves was two. $R_s(0)$ is the extrapolated residual resistance at zero temperature, and $\partial R_s/\partial T$ represents the linear slope of $R_s(T)$. The quality of a typical fit is demonstrated in Fig. 1(b) for the film Y/L3. The $R_s(0)$ and $\partial R_s/\partial T$ data evaluated for all films are listed in Table I. In accordance with the discussion of Fig. 1(a), $R_s(0) \approx (90-180)\ \mu\Omega$ is comparable for all films, while $\partial R_s/\partial T$ accumulates at high values of (6–8) $\mu\Omega$/K for the films on $LaAlO_3$, at intermediate values of (0–2) $\mu\Omega$/K for the films on MgO, and slightly below zero for the films on sapphire.

In an attempt to understand the influence of a substrate on the temperature dependence of the surface resistance of the film deposited onto it, the slope $\partial R_s/\partial T$ was tried to be correlated with the crystal structure of the films. It is generally known that a mismatch of lattice parameters between substrate and film induces strain[37] and, potentially, related

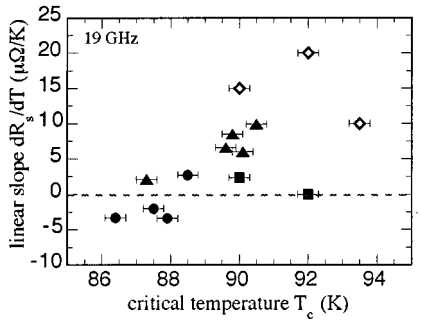

FIG. 2. Slope dR_s/dT as obtained from the quadratic fit in Fig. 1(b), versus critical temperature for the films on sapphire (filled circles), MgO (squares), and $LaAlO_3$ (triangles), and for single-crystals (open diamonds, Refs. 43, 44). The horizontal line marks constant $R_s(T)$.

defects such as microcracks.[38,39] This fact led originally to the development of buffer layers such as CeO_2.[38,40] It is worth noting that the existence of a CeO_2 buffer in sample Y/L2 did not lead to a corresponding deviation of $dR_s/\partial T$ from those observed for the unbuffered $LaAlO_3$ substrates [Fig. 1(a)]. In accordance with this observation, the grouping of steep, intermediate, and shallow slopes of $R_s(T)$ for $LaAlO_3$, MgO, and sapphire anticorrelated clearly with the lattice misfits between YBaCuO and the unbuffered substrates of 0.02, 0.09, and about 0.11.[38]

Alternatively, the crystal structure of $YBa_2Cu_3O_{7-x}$ can be parametrized in terms of the *c*-axis lattice parameter or, e.g., via the oxygen deficiency *x*, in terms of the critical temperature T_c.[41–43] As shown in Fig. 2 for a variety of films including those listed in Table I (filled symbols), the slope $\partial R_s/\partial T$ is obviously correlated with T_c. [It is worth noting that the right-most square represents sample Y/M2. Due to being close to optimal oxygen doping (Sec. II), its T_c deviates from the body of data towards higher values.] Also included in the figure are data on YBaCuO single-crystals extracted from the literature[44,45] (open diamonds), which fit to the overall trend. The scatter of the data represents the influence of further microstructural or electronic peculiarities like inhomogeneities, impurities or granularity on T_c and/or on $\partial R_s/\partial T$.

Altogether, the temperature dependence of the surface resistance of YBaCuO becomes steeper with increasing crystalline perfection of the samples. A similar conclusion was drawn for YBaCuO single crystals doped with Ni and Zn.[45] According to Fig. 2, there seems to be no general difference between the microwave properties of films and crystals. The observed behavior is therefore relevant for the explanation of the pair state.

B. Increase of R_s above its low-temperature level

The analysis of $R_s(T)$ in Sec. III A. showed that the residual resistance was independent of the employed substrates, in contrast to the slope $\partial R_s/\partial T$. It is therefore important to analyze the temperature induced increase of R_s,

$$\Delta R_s(T) = R_s(T) - R_s(0), \tag{2}$$

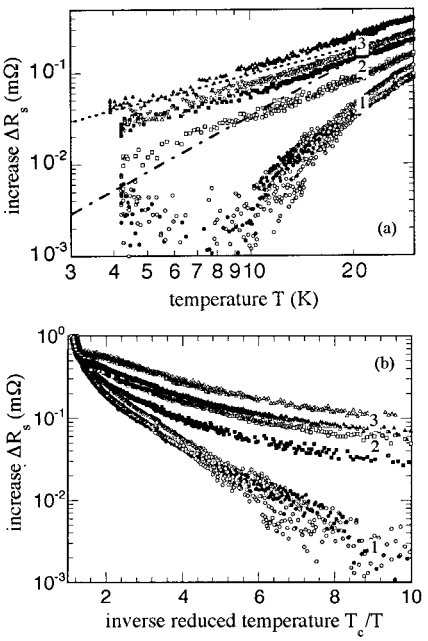

FIG. 3. Temperature dependence of $\Delta R_s = R_s(T) - R_s(0)$ in double-logarithmic scaling (a), and in an Arrhenius plot (b). The notation is the same as in Fig. 1. The dotted and dash-dotted lines in (a) represent the linear ($\alpha = 1$) and the quadratic dependences ($\alpha = 2$) of $\Delta R_s \propto T^{\alpha}$.

in greater detail. The results for the samples discussed along with Fig. 1 are summarized in Fig. 3 using the same notation. Figure 3(a) displays the temperature dependence of $\Delta R_s(T)$ in a double-logarithmic scaling, in comparison with the power-law behavior T^a, $a=1$ (dotted line) and $a=2$ (dash-dotted line). The reproducibility and the grouping of the data according to the three different types of substrates is similar as in Fig. 1. The data for Y/L and for Y/M are consistent with a slope $a \approx 1.1-1.4$ over the entire temperature range up to 40 K, which is in accordance with other reports.[2,4] In contrast, the data for Y/S at $T > 10$ K cannot be described by a single power law, rather than by a series of power-laws with different exponents. The data scatter below 10 K reflects an uncertainty of about 10 $\mu\Omega$ for the $R_s(0)$ levels.

Figure 3(b) is a representation of the same data as in Fig. 3(a), but scaling ΔR_s logarithmically versus the inverse reduced temperature T_c/T. This scaling is naturally applied to conventional superconductors whose surface resistance displays thermally activated behavior as discussed in Sec. I. Plotting $\Delta R_s(T)$ data then yields, at $T_c/T > 2$, a straight line, the slope of which is a measure of the reduced energy gap δ. The experimental data for Y/L and Y/M display a plateau in ΔR_s close to the transition temperature, which is well known to reflect strongly temperature dependent quasiparticle scattering,[1–5] but no linear behavior in any temperature range. In contrast, the plateau is suppressed in the films on sapphire, and $\Delta R_s(T)$ follows a pronounced activated behavior at $T_c/T > 2$ with a slope $\delta \approx 0.8$. The magnitude of this slope agrees with earlier reports on $R_s(T)$ of YBaCuO films and crystals,[24–26] and with the observation of a small energy gap of break junctions in $YBa_2Cu_3O_{7-x}$ and $YbBa_2Cu_3O_{7-x}$ single crystals.[46]

The observation of an exponential temperature dependence of $R_s(T)$ is an undoubtful proof of existence of an extended energy range where the density of states is identical to zero. It consequently limits possible interpretations of the pair state in YBaCuO such as the coexistence of a dominant *d*-wave contribution with a subdominant *s*-wave contribution, since such scenarii imply a finite angle-averaged density of states extending down to zero energy.

IV. TWO-FLUID MODEL OF THE SURFACE RESISTANCE

A. Basic features of the model

The surface impedance of high-temperature superconductors can be described phenomenologically with the two-fluid model in the local limit, in terms of the temperature dependent complex conductivity, as discussed in numerous publications:[1–3,17]

$$Z_s(T) = \left[\frac{i\mu_0\omega}{\sigma_1(T) - i\sigma_2(T)} \right]^{1/2}. \tag{3}$$

Assuming a Drude model for the quasiparticle conductivity with a frequency independent scattering time τ allows us to express σ_1 and σ_2 in terms of the number densities of unpaired and paired charge carriers, n_N and n_S:

$$\sigma_1(T) = \frac{e^2}{m^*} n_N(T) \frac{\tau(T)}{1 + [\omega\tau(T)]^2} \tag{4a}$$

and

$$\sigma_2(T) = \frac{1}{\mu_0\omega\lambda^2(T)} = \frac{e^2}{m^*\omega} \left\{ n_S(T) + n_N(T) \frac{[\omega\tau(T)]^2}{1 + [\omega\tau(T)]^2} \right\}, \tag{4b}$$

where the temperature dependences of all relevant quantities are explicitly indicated, e is the electronic unit charge, and m^* the effective quasiparticle mass. The first part of Eq. (4b) defines the microwave penetration depth $\lambda(T)$ as usual. The density $n_S(T)$ is related to $n_N(T)$ by

$$n_S(T) + n_N(T) = n_F, \tag{4c}$$

with n_F the number density of charge carriers at the Fermi level. At low temperatures, where $\sigma_1(T) \ll \sigma_2(T)$ holds, Eqs. (3) and (4) can be simplified to yield the surface resistance:

$$R_s(T) = \mu_0^2\omega^2/2 \times \lambda^3(T) \times \sigma_1(T). \tag{5}$$

In order to parametrize the surface resistance according to Eqs. (4) and (5), models are required for the temperature dependences of $n_N(T)$ and $\tau(T)$. The general form of $n_N(T)$, normalized to the residual fraction $n_0 > 0$ of unpaired charge carriers, can be considered to be

$$\eta_n(t) \equiv \frac{n_N(t)}{n_0} = 1 + (v-1)F(t) \tag{6}$$

with $t = T/T_c$ the reduced temperature and $v = n_F/n_0$. The temperature dependence of $n_N(T)$ is contained in the normalized function $F(T)$, with $F(0) = 0$ and $F(1) = 1$. An appreciable body of data (see, e.g., review in Ref. 3) pointed to the empirical behavior

$$F(t) = F_1(t) = t^2, \tag{7a}$$

while for a finite reduced energy gap δ follows

$$F(t)=F_2(t)=\exp\left(-\frac{\delta}{t}\right). \tag{7b}$$

The reduced energy gap $\delta(t)$ is generally temperature dependent such that $\delta(1)=0$, but it can be considered constant at $t<1/2$.

The quasiparticle scattering rate $\tau^{-1}(T)$ of YBCO was found to drop exponentially below T_c, and to approach with a cubic power-law the constant value τ_0 at low temperatures.[2,5] Since here we are mainly interested in the temperature dependence of R_s at $t<\frac{1}{2}$, it follows that

$$\frac{1}{\tau(t)}=\frac{1}{\tau_0}+\frac{1}{\tau_T(t)}=\frac{1}{\tau_0}+\frac{t^3}{\tau_T(1)}. \tag{8a}$$

The total expression for the scattering time becomes in reduced units, using $\theta=[\tau_0/\tau(1)-1]$:

$$\eta_\tau(t)\equiv\frac{\tau(t)}{\tau_0}=\frac{1}{1+\theta\times t^3}. \tag{8b}$$

Combining Eqs. (4)–(8) gives the surface resistance in terms of the temperature dependent functions $\eta_n(t)$ and $\eta_\tau(t)$:

$$\frac{R_s(t)}{\mu_0\omega\lambda_p/2}=\nu^{1/2}\varphi_0\frac{\eta_n(t)\times\eta_\tau(t)}{1+[\varphi_0\eta_\tau(t)]^2}\left\{\nu-\frac{\eta_n(t)}{1+[\varphi_0\eta_\tau(t)]^2}\right\}^{-3/2} \tag{9}$$

with $\lambda_p=m^*/\mu_0 n_F e^2$ the plasma wavelength ($\lambda_p\approx 160$ nm in YBCO).[5] The constant parameters contained in Eq. (9) are ν (quantifying the inverse fraction of unpaired charge carriers at zero temperature, $\nu\to\infty$ in an ideal superconductor), the reduced energy gap δ (which is determined by measured data), θ (quantifying the steepness of the drop of the scattering rate below T_c), and $\varphi_0\equiv\omega\tau_0$ (which is given by the residual scattering rate). The parameters ν and φ_0 are related to the residual resistance $R_s(0)$ by

$$\frac{R_s(0)}{\mu_0\omega\lambda_p/2}=\nu^{1/2}\frac{\varphi_0}{1+\varphi_0^2}\left(\nu-\frac{1}{1+\varphi_0^2}\right)^{-3/2}. \tag{10}$$

B. Results for the surface resistance

Starting with a discussion of the residual resistance, Eq. (10) can be simplified since $\nu\gg 1$ (typically[20] $n_0/n_F=1/\nu<0.05$) and $\varphi_0\ll 1$ (typically[5] $\omega\tau_0\approx 0.2$ at $\omega/2\pi=19$ GHz) are realistic approximations, resulting in

$$\frac{R_s(0)}{\mu_0\omega\lambda_p/2}\approx\frac{\varphi_0}{\nu}\propto n_0\tau_0. \tag{11}$$

The observation that $R_s(0)$ is nearly independent of the type of substrate (Sec. III A) implies therefore that the residual scattering rate τ_0^{-1} scales in proportion to the number density n_0 of unpaired charge carriers. Obviously, τ_0 and n_0 have a similar (or even the same) origin, which could be intrinsic to the superconductor. In *s*-wave (respectively: *d*-wave) superconductors, the most likely mechanism to cause an appreciable fraction of unpaired charge carriers and, at the same time, scattering is magnetic (respectively: nonmagnetic) impurity scattering. However, regarding the obvious similarity of the $R_s(0)$ values of high-purity crystals and strained Y/S films, the second explanation seems inappropriate, at least for YBCO.

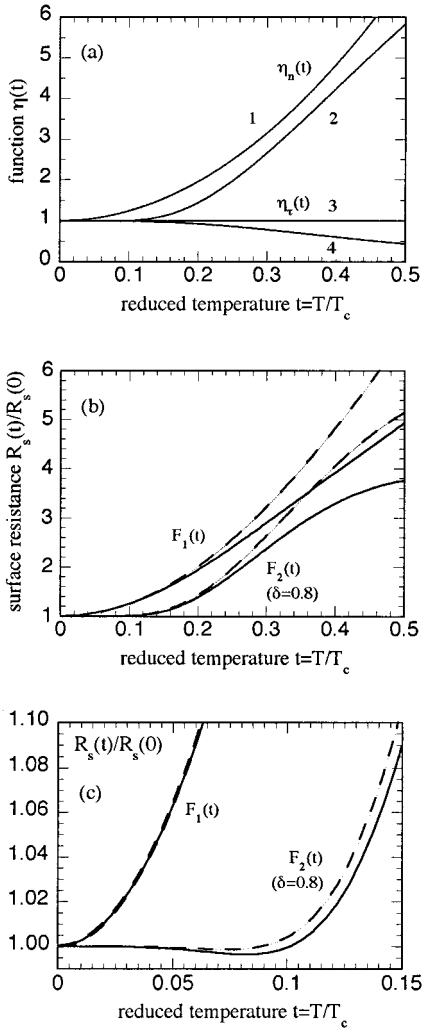

FIG. 4. (a) Temperature dependences of the reduced quasiparticle density $\eta_n(t)$ [curve 1: $F_1(t)$, curve 2: $F_2(t)$ with $\delta=0.8$, see Eqs. (7)] and of the reduced scattering time $\eta_\tau(t)$ [curve 3: $\theta=0$, curve 4: $\theta=10$, see Eq. (8b)]. (b) Results for the reduced surface resistance $R_s(t)/R_s(0)$ for gapless (F_1) and gapped (F_2, $\delta=0.8$) pair condensation and for $\theta=5$ (dashed) and $\theta=10$ (solid curves), as calculated from Eqs. (9) and (10). (c) Same as in (b), on an expanded scale.

The temperature dependence of the surface resistance is given in the framework of the two-fluid model developed in Sec. IV A by that of $\eta_n(t)$ and of $\eta_\tau(t)$ [Eqs. (6)–(9)]. Figure 4(a) illustrates $\eta_n(t)$ using $F_1(t)$ (curve 1) and $F_2(t)$ ($\delta=0.8$, curve 2), and $\eta_\tau(t)$ for $\theta=0$ (curve 3) and $\theta=10$ (curve 4). The two-fluid approximation $F_1(t)=t^2$ for the pair breaking leads to an increase of $\eta_n(t)$ starting at the lowest temperatures while the assumption of a finite, though small, energy gap results in a constant number of unpaired charge carriers $n_N(t)=n_0$ at low temperatures such that $\delta/t\gg 1$ (here: $t\leqslant 0.1$). The reduced scattering time $\eta_\tau(t)$ stays constant or decreases with increasing temperature, depending on

the choice of θ. The variation of η_n with temperature is much stronger and opposite to that of η_τ. Figures 4(b) and 4(c) display the reduced surface resistance $R_s(t)/R_s(0)$ calculated from Eqs. (9) and (10) for different sets of parameters. According to the previous discussion, the parameter $\varphi_0 = \omega\tau_0$ was fixed at 0.2, and the residual fraction of unpaired charge carriers at $v = 25$. The temperature dependence of R_s modeled for the gapless case [function $F_1(t)$] obeys a power-law as expected, with the steepness depending on the parameter θ [dashed curves in Figs. 4(b) and 4(c): $\theta = 5$, solid curves: $\theta = 10$]. In contrast, assuming a reduced energy gap $\delta = 0.8$ [function $F_2(t)$] leads to an almost constant $R_s(t)$ at temperatures $t < 0.15$. In detail, as magnified in Fig. 4(c), the competition between pair breaking (η_n) and scattering (η_τ) [see curves 2 and 4 in Fig. 4(a)] results in a shallow minimum of $R_s(t)$ slightly below $t = 0.1$.

V. CONCLUDING DISCUSSION

In summary, the measured residual resistance of differently prepared high-quality epitaxial YBaCuO films was independent of the employed substrates and quantitatively reproducible, $R_s(0) \approx 130\,\mu\Omega$ at 19 GHz. The temperature dependence of R_s, in terms of the slope $\partial R_s/\partial T$, became stronger with increasing crystalline perfection, as parametrized here by the critical temperature, from films on sapphire to MgO and further to $LaAlO_3$ substrates. This correlation extrapolated from epitaxial films even to high-purity single-crystals. Up to about 40 K, the increase $\Delta R_s(T) \propto T^a$ above the zero-temperature value $R_s(0)$ was between linear and quadratic (with an exponent $a \approx 1.3$) for the films on $LaAlO_3$ and MgO in accordance with other reports.[1,2] However, it was exponential for the films on CeO_2-buffered sapphire and corresponded to a reduced energy gap $\delta \approx 0.8$. This observation proves the existence of an extended energy range where the average density of quasiparticle states must be zero.

The observed $R_s(T)$ behavior was related in terms of a two-fluid model to the opposing temperature dependences $\eta_n(T)$ and $\eta_\tau(T)$ of the pair breaking and the quasiparticle scattering, respectively. The model involved the fixed parameters δ (set either to 0 or to 0.8) and $\varphi_0 \equiv \omega\tau_0 = 0.2$,[5] and the variables $v = n_F/n_0$ and $\theta = [\tau_0/\tau(1) - 1]$. The universal $R_s(0)$ values implied that the residual scattering rate τ_0^{-1} scaled in proportion to the residual fraction n_0 of unpaired charge carriers. Magnetic impurities were concluded to be the most likely source of pair-breaking and, at the same time, of scattering in YBaCuO, independent of the type of samples. The $R_s(0)$ level could be explained with a reasonable value of $n_0 \approx n_F/25$,[20] which turned out to be independent of the slope $\partial R_s/\partial T$. The exponent $a = \partial \ln(\Delta R_s)/\partial \ln(T)$ agreed with the data measured for the Y/L and Y/M films when assuming $\delta = 0$ and $\eta_n(T) \propto T^2$, as suggested by numerous penetration depth data.[2–4] In contrast, in the case of activated behavior with $\delta = 0.8$, the decrease of the scattering time $\eta_\tau(t)$ became competitive to the weak exponential increase of $\eta_n(T)$ at low temperatures, leading to a shallow minimum of $R_s(T)$ around 10 K (assuming $T_c \approx 90$ K), in accordance with the data measured for the Y/S films [Fig. 1(a)].

While the simple model can explain various features, the remaining questions concern the origin of the correlation of $\partial R_s/\partial T$ with T_c (Fig. 2), and the related turnover from a power law to an exponential temperature dependence of η_n as a function of the employed type of substrate. Recent reports[42,43] on significant enhancements of the critical temperature in strained LaSrCuO films on SrLaAlO demonstrated that lattice deformations associated with strain can fundamentally modify the energy scales governing the pair condensation. As concluded for the case of YBaCuO films, the separation of two adjacent CuO_2 planes from one another due to in-plane compressive strain (known as the ''Poisson effect'') is expected to reduce T_c. Such a behavior could explain the lower critical temperatures of the Y/S films compared to Y/L (Table I). Another consequence of an elongation of the lattice along the crystalline c direction is a modification of the electronic interactions between the CuO_2 planes and the CuO chains, which are suspected to induce superconductivity in the chains.[21,22,47] Whether the chains display a vanishing or a finite energy gap was argued in the two-band model to depend on the hole doping level (i.e., the oxygen deficiency), but also on magnetic interactions between the out-of-plane (''apex'') oxygen sites and the CuO chains. Increasing the plane-chain distances could therefore render the chains from a gapless into a weakly gapped state. This proposed mechanism would provide a natural explanation for activated behavior to occur predominantly in strained films rather than in high-purity crystals.

An alternative explanation of the occurrence of gapped superconductivity in YBaCuO could be scattering at nonmagnetic impurities, which are likely to be present in strained films, and which are known to reduce the critical temperature of d-wave superconductors.[7,48] It is speculated that defects and interfaces in these highly anisotropic short-coherence-length materials lead to a frustration of the d-wave symmetry, and to the appearance of a subdominant s-wave component extending over a few coherence lengths.[49–51] However, it is questionable if this scenario can account for the observed exponential temperature dependence of the surface resistance, given that this quantity integrates over the entire sample area, and over all quasiparticle energies and momentum directions.[1,3] Furthermore, the small energy gap concluded here from the variation of R_s at low temperatures is not sufficient to describe the steep drop of R_s near T_c, but requires the assumption of a second, much larger, energy gap (approximately $2\Delta/k_B T_c \approx 6$) as discussed in Ref. 5.

In conclusion, the presented data contain serious constraints for any interpretation of the pair state in YBaCuO, if not in all cuprate superconductors. They indicate the coexistence of two superconducting bands. The occurrence of a finite energy gap defines a reference, and illustrates the necessity, for any theoretical approach in terms of d-wave or mixed d/s-wave symmetries to evaluate the resulting density of states. On the experimental side, the data connect the behavior of films to that of single-crystals, which were believed so far to be the only type of material to study intrinsic properties of cuprate superconductors. The proposed model can and will be checked with further microwave measurements on different cuprates and in different doping levels.

ACKNOWLEDGMENTS

The supply of HTS films from M. Getta (Wuppertal), R. Humphreys (DERA Malvern), H. Kinder (Munich), M. Lorenz (Leipzig), B. Stritzker (Augsburg), and R. Wördenweber (Jülich) is gratefully acknowledged. Valuable discussions with L. Alff, J. Halbritter, H. Hilgenkamp, H. Kinder, and V. Z. Kresin are greatly appreciated. This work was funded in part by the German BMBF (Grant No. 13N6833).

[1] J. R. Waldram, *Superconductivity of Metals and Cuprates* (IOP, Bristol, 1996).

[2] W. N. Hardy, S. Kamal, and D. A. Bonn, in *The Gap Symmetry and Fluctuations in High T_c Superconductors*, Vol. 371 of *NATO Advanced Study Institute, Series B: Physics*, edited by J. Bok, G. Deutscher, D. Pavuna, and S. A. Wolf (Plenum Press, New York, 1998); D. A. Bonn, S. Kamal, R. Liang, W. N. Hardy, C. C. Homes, D. N. Basov, and T. Timusk, Czech. J. Phys. **46**, 3195 (1996).

[3] M. A. Hein, *High-temperature Superconductor Thin Films at Microwave Frequencies*, Vol. 155 Springer Tracts of Modern Physics (Springer, Heidelberg, 1999).

[4] A. P. Mourachkine and A. R. F. Barel, in *Studies of High-temperature Superconductors*, edited by A. Narlikar (Nova Sciences, New York, 1996), Vol. 17, p. 221.

[5] S. Hensen, G. Müller, C. Rieck, and K. Scharnberg, Phys. Rev. B **56**, 6237 (1997).

[6] C. C. Tsuei, J. R. Kirtley, C. C. Chi, L. S. Yu-Jahnes, A. Gupta, T. Shaw, J. Z. Sun, and M. B. Ketchen, Phys. Rev. Lett. **73**, 593 (1994).

[7] D. J. Scalapino, Phys. Rep. **250**, 329 (1995).

[8] D. J. Van Harlingen, Rev. Mod. Phys. **67**, 515 (1995).

[9] J. Mannhart, H. Hilgenkamp, B. Mayer, Ch. Gerber, J. R. Kirtley, K. A. Moler, and M. Sigrist, Phys. Rev. Lett. **77**, 2782 (1996).

[10] E. Il'ichev, V. Zakosarenko, R. P. J. IJsselsteijn, V. Schultze, H.-G. Meyer, H. E. Hoenig, H. Hilgenkamp, and J. Mannhart, Phys. Rev. Lett. **81**, 894 (1998).

[11] J. R. Schrieffer, J. Low Temp. Phys. **99**, 397 (1995); Bull. Am. Phys. Soc. **41**, 74 (1996).

[12] A. G. Sun, A. Truscott, A. S. Katz, and R. C. Dynes, Phys. Rev. B **54**, 6734 (1996).

[13] M. Mößle and R. Kleiner, Phys. Rev. B **59**, 4486 (1999).

[14] Ya. G. Ponomarev, C. S. Khi, K. K. Uk, M. V. Sudokova, S. N. Tchesnokov, M. A. Lorenz, M. A. Hein, G. Müller, H. Piel, B. A. Aminov, A. Krapf, and W. Kraak, Physica C **315**, 85 (1999).

[15] V. Z. Kresin and S. A. Wolf, Phys. Rev. B **46**, 6458 (1992); V. Z. Kresin, H. Morawitz, and S. A. Wolf, *Mechanisms of Conventional and High-T_c Superconductivity* (Oxford University Press, New York, 1993).

[16] A. A. Golubov, M. R. Trunin, A. A. Zhukov, O. V. Dolgov, and S. V. Shulga, J. Phys. I **6**, 2275 (1996); Pis'ma Zh. Eksp. Teor. Fiz. **62**, 477 (1995) [JETP Lett. **62**, 496 (1995)]; A. A. Golubov (unpublished).

[17] H. Srikanth, Z. Zhai, S. Sridhar, A. Erb, and E. Walker, Phys. Rev. B **57**, 7986 (1998).

[18] J. P. Carbotte, Rev. Mod. Phys. **62**, 1027 (1990).

[19] X. Leyronas and R. Combescot, Phys. Rev. B **54**, 3482 (1996).

[20] A. C. Cassinese, M. A. Hein, S. Hensen, and G. Müller (unpublished).

[21] Yu. N. Ovchinnikov and V. Z. Kresin, Phys. Rev. B **54**, 1251 (1996); V. Z. Kresin, S. A. Wolf, and Yu. N. Ovchinnikov, J. Supercond. **9**, 431 (1996).

[22] V. Z. Kresin, A. Bill, S. A. Wolf, and Yu. N. Ovchinnikov, in *The Gap Symmetry and Fluctuations in High-T_c Superconductors* (Ref. 2).

[23] S. B. Nam, Phys. Rev. **156**, 470 (1967); **156**, 487 (1967).

[24] N. Klein, N. Tellmann, H. Schulz, K. Urban, S. A. Wolf, and V. Z. Kresin, Phys. Rev. Lett. **71**, 3355 (1993).

[25] S. M. Anlage and D. H. Wu, J. Supercond. **5**, 395 (1992).

[26] V. A. Gasparov, M. R. Mkrtchyan, M. A. Obolensky, and A. V. Bondarenko, Physica C **231**, 197 (1994).

[27] T. Kaiser, W. Diete, M. Getta, M. A. Hein, G. Müller, M. Perpeet, and H. Piel, Part. Accel. **60**, 171 (1998).

[28] T. Bollmeier, W. Biegel, B. Schey, B. Stritzker, W. Diete, T. Kaiser, and G. Müller, J. Alloys Compd. **251**, 176 (1997).

[29] A. G. Zaitsev, G. Ockenfuss, D. Guggi, R. Wördenweber, and U. Krüger, J. Appl. Phys. **81**, 3069 (1997); A. G. Zaitsev, R. Kutzner, R. Wördenweber, T. Kaiser, M. A. Hein, and G. Müller, J. Supercond. **11**, 361 (1998).

[30] G. Müller, B. Aschermann, H. Chaloupka, W. Diete, M. Getta, M. Hein, S. Hensen, F. Hill, M. Lenkens, S. Orbach-Werbig, T. Patzelt, H. Piel, J. Rembesa, H. Schlick, T. Unshelm, and R. Wagner, IEEE Trans. Appl. Supercond. **5**, 1729 (1995).

[31] N. G. Chew, J. A. Edwards, R. G. Humphreys, J. S. Satchell, S. W. Goodyear, B. Dew, N. J. Exon, S. Hensen, M. Lenkens, G. Müller, S. Orbach-Werbig, IEEE Trans. Appl. Supercond. **5**, 1167 (1995).

[32] H. Kinder, P. Berberich, B. Utz, W. Prusseit, IEEE Trans. Appl. Supercond. **5**, 1575 (1995).

[33] M. Lancaster, *Passive Microwave Device Applications of High-temperature Superconductors* (Cambridge University Press, Cambridge, 1997).

[34] A. Hosseini, R. Harris, S. Kamal, P. Dosanjh, J. Preston, R. Liang, W. N. Hardy, and D. A. Bonn, Phys. Rev. B **60**, 1349 (1999).

[35] DuPont Superconductivity report (unpublished).

[36] M. A. Hein (unpublished).

[37] A. H. Cottrell, in *Physics of Metals* edited by P. B. Hirsch (Cambridge University Press, Cambridge, 1975), Vol. 2.

[38] A. G. Zaitsev, G. Ockenfuss, and R. Wördenweber, in *Applied Superconductivity 1997*, Inst. Phys. Conf. Ser. No 158, edited by H. Rogalla and D. H. A. Blank (IOP Publishing, Bristol, 1997), Vol. 1, p. 25.

[39] G. Kästner, C. Schäfer, St. Senz, T. Kaiser, M. A. Hein, M. Lorenz, H. Hochmuth, and D. Hesse, Supercond. Sci. Technol. **12**, 366 (1999).

[40] X. D. Wu, R. C. Dye, R. E. Muenchhausen, S. R. Foltyn, M. Maley, A. D. Rollett, A. R. Garcia, and N. S. Nogar, Appl. Phys. Lett. **58**, 2165 (1991).

[41] J. D. Jorgensen, B. W. Veal, A. P. Paulikas, L. J. Nowicki, G. W. Crabtree, H. Claus, and W. K. Kwok, Phys. Rev. B **41**, 1863 (1990).

[42] J.-P. Locquet, J. Perret, J. Fompeyrine, E. Mächler, J. W. Seo, and G. VanTendeloo, Nature (London) **394**, 453 (1998).

[43] I. K. Schuller, Nature (London) **394**, 419 (1998).
[44] K. Zhang, D. A. Bonn, S. Kamal, R. Liang, D. J. Baar, W. N. Hardy, D. Basov, and T. Timusk, Phys. Rev. Lett. **73**, 2484 (1994); A. Hosseini, S. Kamal, D. A. Bonn, R. Liang, and W. N. Hardy, *ibid.* **81**, 1298 (1998).
[45] D. A. Bonn, S. Kamal, K. Zhang, R. Liang, D. J. Baar, E. Klein, and W. N. Hardy, Phys. Rev. B **50**, 4051 (1994).
[46] B. A. Aminov, M. A. Hein, G. Müller, H. Piel, D. Wehler, Ya. G. Ponomarev, K. Rosner, and K. Winzer, J. Supercond. **7**, 361 (1994).
[47] V. Z. Kresin, S. A. Wolf, and Yu. N. Ovchinnikov, Phys. Rev. B **53**, 11 831 (1996).
[48] P. J. Hirschfeld and N. Goldenfeld, Phys. Rev. B **48**, 4219 (1993).
[49] C. R. Hu, Phys. Rev. Lett. **72**, 1526 (1994).
[50] M. Matsumoto and H. Shiba, J. Phys. Soc. Jpn. **64**, 1703 (1995); **64**, 3384 (1995); **64**, 4867 (1995).
[51] A controversial discussion of this topic can be found, e.g., in *Proceedings of the ''Euroconference on Polarons: Condensation, Pairing, Magnetism,''* Erice, Italy, June, 1998 [J. Supercond. **12** (1999)].