Paraconductivity and excess Hall effect in epitaxial $YBa_2Cu_3O_7$ films induced by superconducting fluctuations

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Measurements of the temperature dependence of the electrical resistivity and the Hall effect in epitaxial films of $YBa_2Cu_3O_7$ are reported. The resistivity is close to linear in T and the cotangent of the Hall angle proportional to T^2 at temperatures sufficiently above T_c. Deviations from this behavior near T_c are attributed to thermodynamic fluctuations and are analyzed in terms of theories for direct (Aslamazov-Larkin) and indirect (Maki-Thompson) fluctuation contributions in a layered superconductor. We obtain an unequivocal fit to both paraconductivity and excess-Hall-effect data with a *consistent* set of three parameters, i.e., the *c*-axis coherence length $\xi_c(0)=1.5$ Å, the phase-relaxation time $\tau_\phi=8.6\times10^{-14}$ s at 100 K, and the electron-hole asymmetry parameter $\beta=-0.17$. We observe a sign change of the excess Hall effect close to T_c, when the Aslamazov-Larkin process becomes dominant.

I. INTRODUCTION

Fluctuations of the superconducting order parameter have been a well-known effect in conventional superconductors and manifest themselves in all properties associated with the superconducting state, e.g., in the resistivity, susceptibility, specific heat, and several others.[1] Since the superconducting fluctuations are based on thermodynamic considerations, their magnitude is essentially governed by a characteristic length, the Ginzburg-Landau coherence length. It may be easily imagined that large coherence lengths in conventional superconductors make the fluctuations a small correction to the normal-state properties in a very limited temperature range above T_c. The situation is remarkably different in the cuprate high-temperature superconductors. The high-temperature scale of the critical temperature, the layered structure of the copper oxide planes, giving rise to a high anisotropy in the normal state and in the superconducting properties, and the extremely short in-plane coherence length are all favorable for the occurrence of superconducting fluctuations in a broad temperature range, which is conveniently accessible for detailed experimental analysis. In early investigations of the transport properties of high-temperature superconductors, a remarkable rounding of the superconducting transition was observed, and the question was raised whether this characteristic feature may be attributed to inhomogeneities in the ceramic samples or to highly enhanced superconducting fluctuations.[2] Recent, careful investigations on high-quality single crystals and thin films clearly show that these effects fit the predictions of the fluctuation theories in a certain temperature range.[3-8] Several questions, however, remain controversial and will be addressed in this paper.

There have been numerous reports on the *paraconductivity,* the enhancement of the electrical conductivity near T_c, in bulk samples, thin films, and single crystals. The analysis in $YBa_2Cu_3O_{7-\delta}$ was based either on the initial work by Aslamazov and Larkin[9] (AL), or on the formula of Lawrence and Doniach (LD) for two-dimensional, layered superconductors,[10] which interpolates between the limiting cases of two- and three-dimensional fluctuations in the AL theory. There is no complete agreement, however, whether the results are more favorable for two-dimensional superconductivity,[3,4] eventually crossing over to three-dimensional behavior close to T_c,[5-8] or for an isotropic case.[2] Another point of interest is whether the indirect fluctuation contribution, proposed by Maki[11] and Thompson[12] (MT) may be observed in the paraconductivity of high-temperature superconductors. This question is of remarkable importance, since the direct AL contribution can also be derived in the framework of the time-dependent Ginzburg-Landau theory[13] and therefore should represent a rather universal feature of any type of superconducting mechanism, whereas the MT process was calculated on a microscopic ansatz, which is expected to be strongly dependent on the pairing mechanism. The question if a MT contribution does generally exist in high-temperature superconductors, and whether its temperature dependence is the same as in BCS superconductors, is still open.

One of the reasons for rather diverging interpretations of otherwise similar experimental results of transport properties at temperatures above T_c is the procedure used to separate the fluctuation part from the normal-state properties. In contrast to conventional superconductors, it is impossible to suppress superconductivity and superconducting fluctuations by a technically available magnetic field. To get around this shortcoming, an extrapolation from the normal-state properties at elevated temperatures, where superconducting fluctuations

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disappear, was generally used,[2,3,6–8] or some model for the normal-state behavior was included in the parameters for the fit to the experimental data.[4,5]

Another approach to the problem is an analysis of the magnetoresistance in the fluctuation regime, an effect where the normal-state properties are cancelled, and consequently no assumption for their temperature dependence is needed.[14] The theory for the fluctuation magnetoresistance,[15] however, includes at least four different contributions with several parameters, which have to be fitted to a single physical property.

Investigations of the Hall effect in the presence of superconducting fluctuations, which may be called the *excess Hall effect,* are rare[16–21] and the theoretical predictions of the excess Hall effect are rather contradictory. Abrahams *et al.*[22] suggested that the fluctuation contributions usually should cancel out in the Hall effect, Fukuyama *et al.*[23] presented a detailed analysis based on a microscopic calculation, and recently more theoretical papers on the excess Hall effect have appeared.[24–26] A general consensus now seems to be achieved about the existence and about the temperature dependence of the excess Hall effect, but the prediction of the sign seems to be still controversial.

In this paper we present measurements of the paraconductivity and the excess Hall effect in epitaxial thin films of $YBa_2Cu_3O_7$ in low magnetic fields and an analysis of our results using the same set of parameters for both physical quantities. We also attempt to reduce the number of fit parameters as far as possible and present a fit to both measurements with a consistent set of three parameters.

II. EXPERIMENTAL TECHNIQUES

Thin films of $YBa_2Cu_3O_7$ were fabricated by pulsed-laser deposition using 248-nm KrF-excimer-laser radiation and stoichiometric ceramic targets. A schematic of the experimental setup has been shown in Ref. 27. The substrates employed were (100) MgO and (100) $SrTiO_3$. The laser fluence used in the experiments was $\Phi = 4$ J/cm^2 with a pulse repetition rate of 10 Hz and the oxygen background pressure $p(O_2) = 0.4$ mbar. Film growth took place at substrate temperatures around 710 °C. Depending on the number of laser pulses, the film thickness varied between 1000 Å and 4000 Å. The critical current density of films fabricated on (100) MgO was around $j_c(77\text{ K}) \geq 2\times 10^6$ A/cm^2. The x-ray diffraction spectra of the films deposited on both (100) MgO and (100) $SrTiO_3$ show only 1:2:3 phase, with the c axis oriented perpendicular to the substrate surface.

The thin film investigated in this study was deposited on a (100) MgO substrate and was patterned to a five-probe contact arrangement by photoresist and wet chemical etching in diluted phosphoric acid to a strip of 10×1 mm^2 in size. Comparison of the resistivity vs temperature curve with reference films not subjected to the etching procedure indicated no detoriation of the samples. Ag pads were evaporated on the arms of the patterned film and contacts were established with silver paste and gold wires. For the measurement of the Hall voltage, a potentiometer was connected across the two arms on the same side of the sample and was used for nulling the spurious transverse voltage in zero magnetic field due to contact misalignment.

The resistivity and the Hall-effect measurements were performed in a closed-cycle refrigerator with temperature control by a platinum resistor and precision temperature measurements with a calibrated $Ga_{1-x}Al_xAs$ diode placed close to the sample. The measurements were carried out with an ac current of frequency 89 Hz using lock-in technique for the detection of the resistive voltage drop along the sample and for the Hall voltage. A low current density of $j = 80$ A/cm^2 was used. The temperature variation of the longitudinal resistivity was measured in a slow downsweep both with a magnetic field $B = 0.7$ T, which was generated by an electromagnet, and without field, where care was taken to cancel the remanence of the magnet. The Hall effect was measured during an extremely slow temperature sweep (0.02 K/min), which proved to yield more stable conditions than waiting for thermal equilibrium before taking data. The polarity of the magnetic field B was reversed for every data point and the temperature accuracy was better than 10 mK.

III. RESULTS

The temperature variation of the longitudinal resistivity ρ_{xx} along the CuO_2 planes in zero magnetic field is shown in Fig. 1. The 10%–90% width of the superconducting transition is about 0.5 K. At temperatures be-

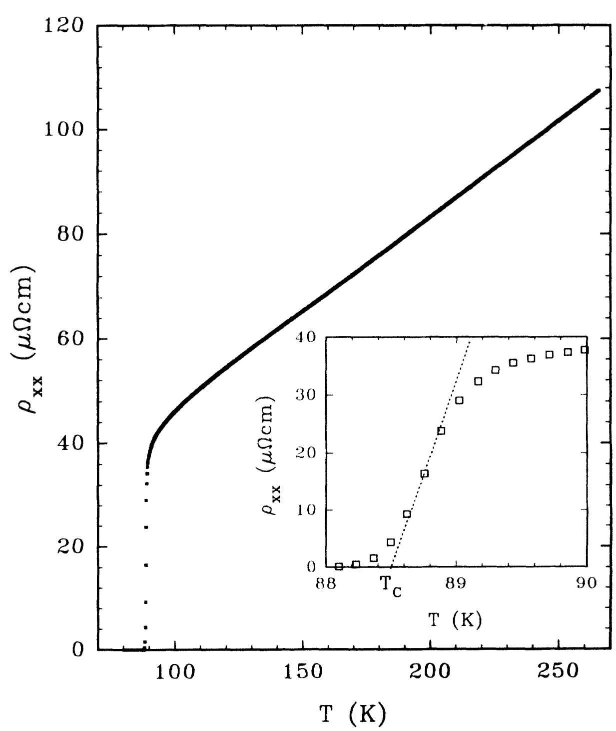

FIG. 1. Temperature variation of the resistivity ρ_{xx} of a $YBa_2Cu_3O_7$ film. The inset shows the region of the superconducting transition and the method for determining T_c.

tween 140 and 200 K a linear temperature dependence is observed. At still higher temperatures a slight upturn can be noticed. This change of slope was also observed in untwinned single crystals[18] and is of presently unknown origin. The transition region is shown enlarged in the insert of Fig. 1. For the analysis presented below, we have to determine the mean-field critical temperature T_c, which depends on the procedure applied. We chose the intersection point of the tangent to the inflection point of the transition curve with the T axis and obtained $T_c = 88.55$ K. The influence of this choice for the subsequent analysis, which will be confined to temperatures not too close to T_c, is insignificant.

A determination of the thickness of the thin films by mechanical probing turned out to be rather inaccurate. In addition it may be argued that small regions on the surface and at the interface to the substrate have considerably higher resistance. Similar considerations were brought up by Oh *et al.*,[7] who proposed a "C factor." We took a different approach and scaled our data to the resistivity value ρ_{xx} (100 K) $=45\ \mu\Omega$ cm, which generally is considered to be the intrinsic value for twinned single crystals of $YBa_2Cu_3O_7$,[28] and also is in good agreement with results from untwinned crystals.[18] This procedure avoids the introduction of an additional parameter in our analysis. The resistivity determined from our thickness measurement, which consequently represents an upper limit, was ρ_{xx} (100 K) $=80\ \mu\Omega$ cm.

The transverse resistivity $\rho_{yx} = R_H B$ as a function of temperature in a magnetic field $B = 0.7$ T is presented in Fig. 2. The Hall effect is positive (holelike) and remarkably temperature dependent, a feature which is considered universal in all cuprate superconductors.[29] Several kelvin above T_c, ρ_{yx} changes slope, which we attribute to superconducting fluctuations. At 88.7 K, ρ_{yx} changes sign and exhibits a negative maximum around 88 K. The origin of this sign change is a topic of current interest and will be discussed briefly in this paper.

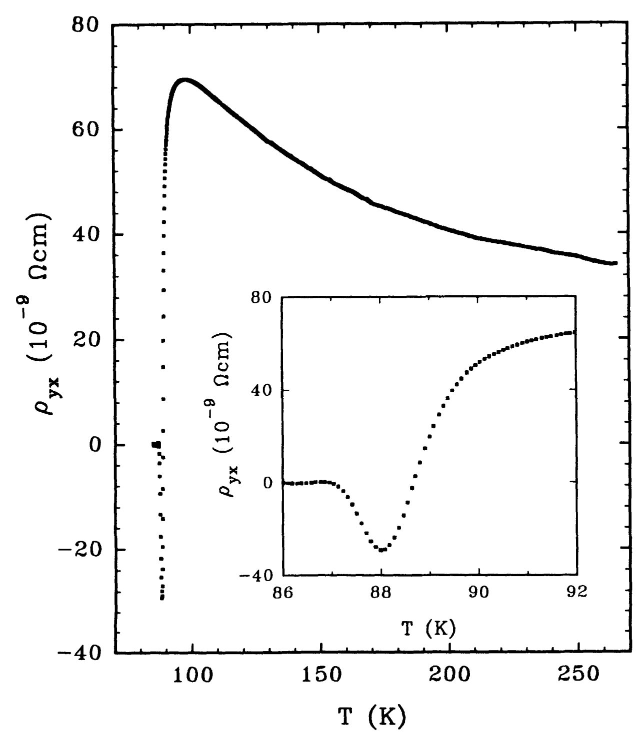

FIG. 2. Hall resistivity ρ_{yx} in a magnetic field $B = 0.7$ T as a function of temperature T. The inset shows the transition region and the sign change of ρ_{xy}.

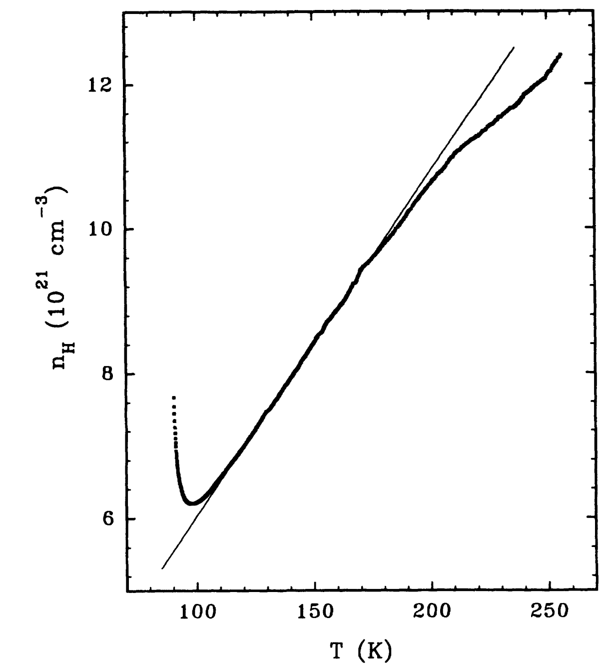

FIG. 3. Hall number $n_H = 1/eR_H$ as a function of temperature T. The straight line indicates a linear relation between n_H and T.

In Fig. 3, we plot the Hall number $n_H = 1/(eR_H)$ versus temperature. The interpretation of this quantity as a carrier density, as would result from a simple one-band model, is questionable. It can be recognized, however, that $1/R_H$ varies linearly with temperature in the range from around 120 to 175 K.

IV. ANALYSIS AND DISCUSSION

A. Paraconductivity

Now we compare the experimental results with the predictions of the theories of thermodynamic fluctuations of the superconducting order parameter. We ignore the anisotropy of the transport properties in a and b direction, due to CuO chains in $YBa_2Cu_3O_7$, since our thin films are twinned. The transport measurements therefore yield an average over all directions in the CuO_2 planes. Due to the geometry of our experiments, properties of the crystallographic c direction will contribute to these transport properties only as a result of misalignments in the films and represent a very small spurious component.

Above T_c, the diagonal component of the conductivity tensor $\sigma_{xx} = \sigma_{yy}$ is composed of contributions from the normal state and from superconducting fluctuations, σ_{xx}^N and $\Delta\sigma_{xx}$, respectively,

$$\sigma_{xx} = \sigma_{xx}^N + \Delta\sigma_{xx} \ . \tag{1}$$

For an evaluation of $\Delta\sigma_{xx}$, the normal-state conductivity σ_{xx}^N has to be subtracted from the measured quantity. No general consensus, however, exists about the functional form of the normal-state conductivity, but the linear temperature dependence of the resistivity over a broad temperature range is generally accepted. Anderson and Zou[30] proposed a formula for the normal-state resistivity

$$\rho_{xx}^N = A/T + BT \ , \tag{2}$$

where A, B are constants. The first term on the right-hand side of Eq. (2) describes the interplane hopping of carriers, the second one the effect of spinon-holon scattering. Although this interpretation is controversial, it supplies an excellent fit to the data, which are shown in Fig. 4 where $\rho_{xx}T$ is plotted as a function of T^2. The parameters used in the fit are $A = 1.048\times10^{-3}$ Ω cm K and $B = 3.897\times10^{-7}$ Ω cm/K. The standard deviation of the data points from the fitted values in the temperature range 160–265 K is 3.22×10^{-8}. We also examined a linear relationship $\rho_{xx} = CT + D$ fitted to the data in the temperature range from 160 to 240 K and found a standard deviation of 4.16×10^{-8}.

The initial work on the enhancement of the conductivity, the *paraconductivity,* by Aslamazov and Larkin[9] considered the acceleration of short-lived superconducting charged carrier pairs, which form in thermal nonequilibrium above T_c, in an electric field. The expressions depend on the dimensionality of the sample and have been extended for two-dimensional, layered superconductors, a situation very much resembling the crystal structure of $YBa_2Cu_3O_7$, by Lawrence and Doniach,[10]

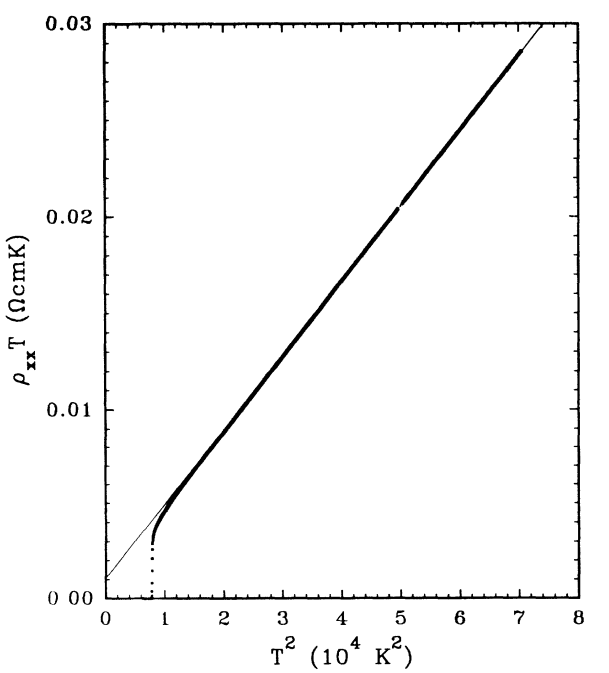

FIG. 4. Plot of $\rho_{xx}T$ versus T^2 to verify the formula $\rho_{xx} = A/T + BT$. The straight line represents the fit to the experimental data.

$$\Delta\sigma_{xx}^{\mathrm{LD}} = (e^2/16\hbar d)(1+2\alpha)^{-1/2}\varepsilon^{-1} \ , \tag{3}$$

where e is the electron charge, $\hbar$ is Planck's constant, d the distance between adjacent layers, and $\varepsilon = \ln(T/T_c) \approx (T-T_c)/T_c$ is a reduced temperature. The coupling parameter

$$\alpha = \frac{2\xi_c^2(T)}{d^2} = \frac{2\xi_c^2(0)}{d^2\varepsilon} \tag{4}$$

models the crossover from the two-dimensional limit at high temperatures to the three-dimensional behavior close to T_c, when the temperature-dependent coherence length $\xi_c(T)$ along the stacking direction exceeds the layer distance.

Maki[11] and Thompson[12] considered the scattering of normal-state quasiparticles by superconducting carrier pairs as an additional mechanism for the paraconductivity in order to account for enlarged fluctuation effects observed in pure lead and alumina samples.[1] This process is limited by strong inelastic scattering and by other pair-breaking interactions, like magnetic impurities or intrinsic magnetic moments. The model has been recently extended for layered superconductors by Hikami and Larkin[31] and Maki and Thompson[32] and yields

$$\Delta\sigma_{xx}^{\mathrm{MT}} = \frac{e^2}{16\hbar d}\,\frac{2}{\varepsilon-\delta}\ln\left\{\frac{\varepsilon}{\delta}\,\frac{1+\alpha+(1+2\alpha)^{1/2}}{1+\alpha\varepsilon/\delta+(1+2\alpha\varepsilon/\delta)^{1/2}}\right\} , \tag{5}$$

where a pair-breaking parameter

$$\delta = \frac{\pi\hbar}{8k_B T\tau_\phi} \tag{6}$$

is introduced and τ_ϕ is the phase-relaxation time of the quasiparticles as given by Patton.[33] Finally the paraconductivity is $\Delta\sigma_{xx} = \Delta\sigma_{xx}^{\mathrm{LD}} + \Delta\sigma_{xx}^{\mathrm{MT}}$.

The paraconductivity $\Delta\sigma_{xx}$ and the values calculated from Eqs. (3) and (5) are plotted in Fig. 5 for a range $0.01 < \varepsilon < 0.1$. The fit contains only one adjustable parameter $\xi_c(0)$ in the LD expression and in addition a second one δ for the MT term. For the interlayer distance d we took the c-axis lattice parameter $d = 11.7$ Å of $YBa_2Cu_3O_7$. The results from the fit are $\xi_c(0) = 1.5$ Å and $\delta = 0.35$. Considering that only two adjustable parameters were used, the fit yields a remarkably good agreement with the experimental data. We did not try to evaluate data for $\varepsilon < 0.01$, since we expect that both a distributions of T_c's in the sample as well as the breakdown of the Gaussian approximation in the theories will invalidate any analysis too close to the critical temperature. On the other hand, an onset of disagreement between experimental and calculated values is discernible when $\varepsilon > 0.07$. Basically, three different sources may account for the observation of a paraconductivity which is smaller than expected from fluctuation theories.

Hagen *et al.*[4] pointed out that the two-dimensional AL-type paraconductivity is proportional to T^{-1} in the limit of high temperatures and therefore is indistinguishable from the normal-state behavior. Any procedure based on the subtraction of σ_{xx}^N will consequently yield a

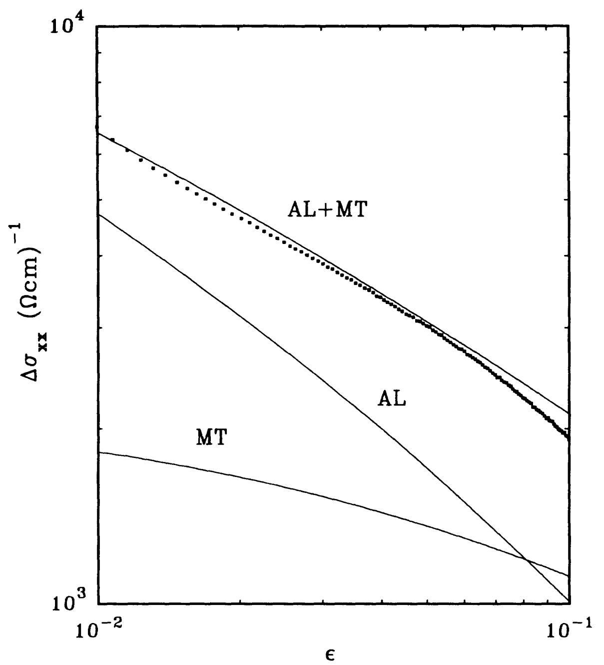

FIG. 5. Paraconductivity in $YBa_2Cu_3O_7$ as a function of the reduced temperature ε. The solid lines represent the results from the fit of the direct (AL) and the indirect (MT) fluctuation processes and the sum of these two contributions (AL+MT) using the parameters specified in the text.

too small value for $\Delta\sigma_{xx}$. To approach this problem it was suggested[4,5] to include a linear function for the normal-state resistivity with two additional parameters in the fitting procedure. This procedure, however, leads *intrinsically* to a perfect fit to the two-dimensional AL term at high temperatures as long as the normal-state resistivity obeys the linear relation.

A similar deviation has also been observed in the paraconductivity of three-dimensional amorphous superconductors[34] and was attributed to a high-energy cutoff in the fluctuation spectrum, where the slow-variation approximation of the Ginzburg-Landau theory is no longer valid. A recent analysis showed,[8] however, that the cutoff can only give a qualitative explanation, since the paraconductivity even falls below this prediction.

As a third possibility, we point out that the deviations become discernible at temperatures where the MT process gains importance. At lower temperatures, the paraconductivity is governed by the AL contribution, but at $\varepsilon=0.8$, a crossover takes place and the MT process becomes the dominant one. Since the MT calculations are based on a detailed microscopic model (namely the BCS model), it is likely that the MT process exhibits at least a different temperature dependence in the cuprate superconductors. We emphasize, however, that we observe a paraconductivity larger than the bare AL contribution and attribute this to a MT process.

There has been much controversy as to whether the paraconductivity in $YBa_2Cu_3O_7$ indicates three- or two-dimensional superconductivity. The LD model predicts a crossover from the three-dimensional limit close to T_c to two-dimensional behavior at high temperatures at $\varepsilon_0=2(\xi_c(0)/d)^2$. The crossover is a rather broad one and takes place at $\varepsilon_0\approx0.03$. Hence, the temperature region of our analysis, $0.01<\varepsilon<0.1$, covers both sides of the crossover temperature and does not extend to the pure three- and two-dimensional limits. From our results it becomes clear that an analysis based on either the two- or the three-dimensional formulas will fail in the experimentally accessible temperature range.

B. Excess Hall effect

Like the paraconductivity, the excess Hall effect is an additional contribution to the *Hall conductivity* $\sigma_{xy}=\rho_{yx}/(\rho_{xx}^2+\rho_{yx}^2)\approx\rho_{yx}/\rho_{xx}^2$. The total Hall conductivity σ_{xy} may be expressed as

$$\sigma_{xy}=\sigma_{xy}^N+\Delta\sigma_{xy} , \tag{7}$$

where σ_{xy}^N is the normal-state Hall conductivity and $\Delta\sigma_{xy}$ is the excess Hall conductivity. For an evaluation of $\Delta\sigma_{xy}$ we have to subtract the normal-state contribution σ_{xy}^N. The nature of the temperature dependence of the Hall effect in the normal state is a matter of intense discussion. Several approaches have been put forward to explain this very unusual property, which seems to be a universal feature of all cuprate superconductors. The suggestions span a large variety from very narrow bands of width $\approx k_BT$,[35] magnetic skew scattering,[36] and two- or multiple-carrier conduction.[37] More recently, Ong *et al.*[38] and Anderson[39] suggested that the Hall effect may be governed by a *Hall relaxation time* $\tau_H\propto T^{-2}$, which has a different temperature dependence from the transport relaxation time $\tau_{tr}\propto T^{-1}$, unlike the situation in ordinary metals. This unusual situation is attributed to a two-dimensional Luttinger-liquid nature of the carrier system in the cuprates. It has been shown[38,40,41] that in fact many measurements on various high-temperature superconductors may be explained using Anderson's formula,[39]

$$\cot\theta_H^N=AT^2+C(x) , \tag{8}$$

where A is a constant related to the doping degree and $C(x)$ is a magnetic impurity concentration.

In Fig. 6 the cotangent of the Hall angle, $\cot\theta_H=\rho_{xx}/\rho_{yx}=\sigma_{xx}/\sigma_{xy}$, is plotted as a function of T^2. The straight line represents the fit to the data using Eq. (8) and the parameters $A=0.0483\ K^{-2}$ and $C(x)=183.6$ in the temperature range 120–175 K. Besides the low-temperature region, where deviations from the linear relationship may be attributed to the excess Hall effect, the semiempirical formula of Eq. (8) is perfectly well obeyed up to 175 K. At this temperature a rather abrupt change of slope is observed. Surprisingly, no corresponding change in the smooth ρ_{xx} behavior can be noticed. We now obtain the Hall conductivity in the normal state, $\sigma_{xy}^N=(\rho_{xx}^N\cot\theta_H^N)^{-1}$, from the fits to Eqs. (2) and (8).

The fluctuation Hall effect was initially studied based on the time-dependent Ginzburg-Landau equation by Abrahams *et al.*,[22] who pointed out that in the limits they applied, the excess Hall effect should vanish, and by Fukuyama *et al.*[23] in a microscopic, BCS-based calculation. The latter work included both the AL and MT con-

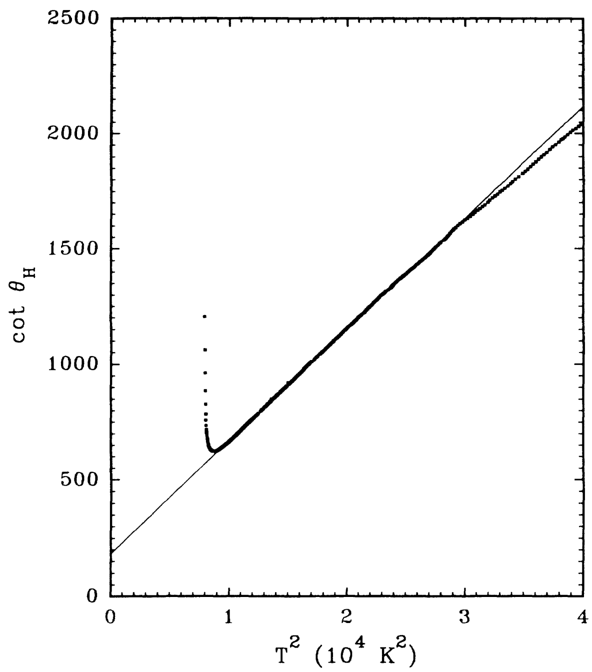

FIG. 6. Cotangent of the Hall angle $\cot\theta_H$ as a function of T^2. The straight line represents the fit of $\cot\theta_H = AT^2 + C(x)$ to the data in the temperature range 120–175 K.

tributions to the excess Hall conductivity and found that the MT process yields a positive contribution $\Delta\sigma_{xy}^{MT} > 0$, whereas the AL term may be either positive or negative, depending on the energy derivative of the density of states at the Fermi energy. The results of Fukuyama *et al.* were given for the limits of two- and three-dimensional superconductors, and the temperature dependence of the AL contribution was predicted to be more divergent than the paraconductivity, i.e., $\Delta\sigma_{xy}^{AL} \propto \varepsilon^{-2}$ in the two-dimensional case. More recently, Ullah and Dorsey[24] (UD) presented a theory in the phenomenological context of the time-dependent Ginzburg-Landau equation in a Hartree approximation, which extends to temperatures close to T_c. In the two- and three-dimensional limits and at temperatures where the Gaussian approximation is valid and fluctuations may be regarded as noninteracting, it reproduces the results of Fukuyama *et al.*[23] There seems to be an agreement about the AL contribution, and an identical temperature dependence for $\Delta\sigma_{xy}^{AL}$ was also predicted by another microscopic calculation of Varlamov and Livanov.[25] We know of no detailed calculation of the MT contribution for the excess Hall effect in the layered superconductor model, but based on the results in the two- and three-dimensional limits,[23] it has been speculated[18] that the nondiagonal MT process also scales with the MT paraconductivity contribution

$$\Delta\sigma_{xy}^{MT} = 2(\sigma_{xy}^N/\sigma_{xx}^N)\Delta\sigma_{xx}^{MT} \tag{9}$$

in the crossover region. For our further analysis we use the result of UD for the AL process, converted to the appropriate numerical prefactors, which yield the formulas of Fukuyama *et al.*[23] in the two- and three-dimensional limits,[42] and Eqs. (5) and (9) for the MT contribution.

$$\Delta\sigma_{xy}^{UD} = \frac{e^2}{16\hbar d}\frac{\sigma_{xy}^N}{\sigma_{xx}^N}\beta\frac{\pi d}{72\xi_c(0)}\frac{1+1/\alpha}{(1+1/2\alpha)^{3/2}}\varepsilon^{-3/2}, \tag{10}$$

$$\Delta\sigma_{xy}^{MT} = \frac{e^2}{16\hbar d}\frac{\sigma_{xy}^N}{\sigma_{xx}^N}\frac{4}{\varepsilon-\delta} \times \ln\left\{\frac{\varepsilon}{\delta}\frac{1+\alpha+(1+2\alpha)^{1/2}}{1+\alpha\varepsilon/\delta+(1+2\alpha\varepsilon/\delta)^{1/2}}\right\}, \tag{11}$$

and finally the excess Hall effect is $\Delta\sigma_{xy} = \Delta\sigma_{xy}^{UD} + \Delta\sigma_{xy}^{MT}$. The parameter β in Eq. (10) reflects an asymmetry between electron and holes[43] and has different interpretations, depending on the model used. Fukuyama *et al.* found from microscopic considerations that β depends on the energy derivative of the electron density of states at the Fermi energy,[23] and Ullah and Dorsey introduced a similar parameter as the imaginary part of the relaxation rate in the time-dependent Ginzburg-Landau equation.[24]

In Fig. 7, we compare σ_{xy} determined from the measurements of the Hall effect with the normal-state Hall conductivity σ_{xy}^N, which was extrapolated from the fit to Eq. (8). At temperatures below 120 K, σ_{xy} begins to surpass the values expected from the normal-state extrapolation, exhibits a maximum at around 93 K, and at still lower temperatures rapidly falls below σ_{xy}^N. This behavior clearly indicates two different, counteracting fluctuation contributions with differing temperature dependencies. Obviously the contribution which reduces

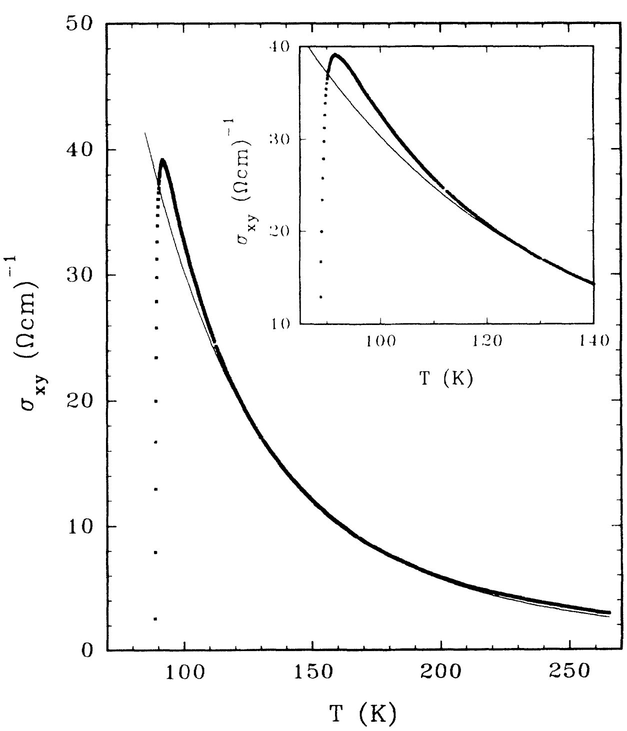

FIG. 7. The Hall conductivity σ_{xy} as a function of the temperature T. The solid line represents the normal-state Hall conductivity σ_{xy}^N deduced from the fit in Fig. 6. Deviations from this line are interpreted as fluctuation contributions to the Hall effect. The inset shows a part of the data on an expanded scale.

σ_{xy} has a higher divergency towards T_c and becomes dominant only close to the superconducting phase transition.

We now compare the excess Hall effect $\Delta\sigma_{xy}$ to the theoretical predictions of Eqs. (10) and (11). There are two adjustable parameters, $\xi_c(0)$ and β, for the direct contribution $\Delta\sigma_{xy}^{\mathrm{UD}}$ and also two, $\xi_c(0)$ and δ, for the indirect contribution $\Delta\sigma_{xy}^{\mathrm{MT}}$. Two of them, $\xi_c(0)$ and δ, have been determined already in the paraconductivity analysis and are used as additional constraints, leaving β as the only adjustable parameter for the analysis of the excess Hall effect. In Fig. 8 we show $\Delta\sigma_{xy}$ in a plot together with the direct (AL) and the indirect (MT) contribution calculated from the fit, using $\beta=-0.17$. The sum of the two fluctuation processes (AL+MT) yields a good fit to the experimental values, including the maximum of $\Delta\sigma_{xy}$ at 93 K ($\varepsilon=0.05$) and the sign change at 90.3 K ($\varepsilon=0.02$). As for the paraconductivity, the experimentally observed excess Hall effect falls below the theoretical values when T exceeds 96 K ($\varepsilon>0.08$).

As may be expected from the theory, the indirect contribution $\Delta\sigma_{xy}^{\mathrm{MT}}$ is positive and dominates at higher temperatures. We clearly observe a negative contribution from the AL process, which governs $\Delta\sigma_{xy}$ close to T_c and also is responsible for the sign change of the fluctuation Hall effect $\Delta\sigma_{xy}$ at 90.3 K. It may be further speculated that this tremendous negative fluctuation Hall effect which we observe even exceeds the positive normal-state Hall effect when approaching T_c from the high-temperature side, leading to the sign change of the Hall conductivity σ_{xy} and consequently of the Hall resistivity ρ_{yx} in Fig. 2. We note, however, that the crossover appears at $T=88.7$ K, a temperature at which critical fluctuations are supposed to become important and Eq. (10) is no longer valid. Generally, the fluctuation contributions in the critical regime interpolate smoothly between the Gaussian fluctuation region above T_c and the flux-flow region below.[24] Thus, a negative fluctuation Hall effect $\Delta\sigma_{xy}$ means that the Hall conductivity σ_{xy} is reduced close to T_c, but does not necessarily imply a sign change of the Hall effect.

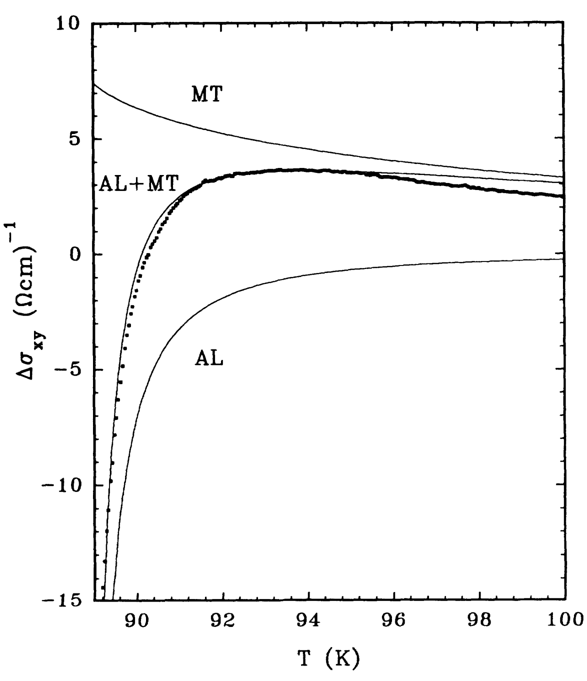

FIG. 8. Excess Hall effect $\Delta\sigma_{xy}$ in $YBa_2Cu_3O_7$ as a function of the temperature T. The line labeled AL+MT is the result from the fit to the sum of the direct (AL) and the indirect (MT) fluctuation contributions, which are also shown separately.

There is another way to make connections between the fluctuation Hall effect and the observation of a negative Hall effect in the flux-flow region. Dorsey[44] and Kopnin *et al.*[45] calculated the Hall effect arising from vortex motion in type-II superconductors in the framework of the time-dependent Ginzburg-Landau theory, and Dorsey and Fisher[46] investigated the disappearance of the Hall effect near the vortex-glass transition. The particle-hole asymmetry is introduced into the Ginzburg-Landau theory by assuming a complex relaxation time of the order parameter, $\gamma_1+i\gamma_2$. Since Ullah and Dorsey[24] also calculated the fluctuation Hall effect in the same framework, we notice that our parameter β is proportional to the imaginary part of the complex relaxation time and find from our results that $\gamma_2<0$. There is, however, some ambiguity in the theoretical papers as to whether or not a negative γ_2 can lead to a negative Hall effect in the flux-flow region.[44,45] Clearly more work is needed to clarify this question.

We now wish to compare our results with previous work. A negative sign of the excess Hall effect was also reported by Iye *et al.*[16] in $ErBa_2Cu_3O_{7-\delta}$ thin films, but they did not observe a positive contribution. We point out, that the MT process is very sensitive to the quality of the material, and that we did not observe the MT process either in the paraconductivity or in the Hall effect in several Y-Ba-Cu-O films with a higher resistivity at 300 K. Rice *et al.*[18] did not observe a negative excess Hall effect in untwinned $YBa_2Cu_3O_7$ single crystals, but their analysis is limited to $\varepsilon>0.07$, where the positive MT term dominates. From the parameters which they used to fit their Hall data, it can be concluded, however, that $\Delta\sigma_{xy}^{\mathrm{UD}}<0$. Very recently, Samoilov[21] found an excess Hall effect in $YBa_2Cu_3O_7$ films similar to our results, but neglected the indirect contribution and interpreted the region where $\Delta\sigma_{xy}>0$ as positive AL-type fluctuations, which reverse sign when entering the critical fluctuation region. Aronov and Rapoport[26] calculated the contribution of the AL process to the excess Hall effect $\Delta\sigma_{xy}^{\mathrm{AL}}\propto\partial\ln T_c/\partial\ln\mu$, where μ is the Fermi level, and concluded that $\Delta\sigma_{xy}^{\mathrm{AL}}>0$ in $YBa_2Cu_3O_{7-x}$. We note, however, that if $x\approx0$, no further enhancement of T_c by hole doping is observed and it has been argued that $YBa_2Cu_3O_7$ is already slightly overdoped,[47] which in turn implies $\partial\ln T_c/\partial\ln\mu<0$. Hence we do not expect a large positive $\Delta\sigma_{xy}^{\mathrm{AL}}$ in fully oxygenated $YBa_2Cu_3O_7$.

Our analysis yields a value of $\delta=0.35$ for the Maki-Thompson pair-breaking parameter from which a quasiparticle phase-relaxation time $\tau_\phi=8.6\times10^{-14}$ s follows according to Eq. (6). Hence $\hbar/\tau_\phi(100\ \mathrm{K})\ =\ 7.7$ meV $\approx k_B T_c$, indicating moderately strong pair breaking. According to the usual picture of the pair-breaking action,[1] this suggests the suppression of the critical temperature from a hypothetical value of $T_{c_0}=136$ K in the absence of pair-breaking effects to the experimentally observed value.

The energy associated with the dephasing time, $\hbar/\tau_\phi$, is of substantial magnitude compared to the zero-temperature energy gap $\Delta(0)\approx 20$ meV in $YBa_2Cu_3O_7$.[48] One thus might expect some characteristic features of pair breaking in the tunneling spectra. Actually the smearing of the gap structure in high-temperature superconductors, which exceeds the thermal broadening, has been attributed to a strong damping of the normal-state quasiparticles[48] and the characteristic energy of this process was found to be 4 meV at 60 K.[49] Taking into account that our value for $\hbar/\tau_\phi$ was determined at 100 K this is in remarkably good agreement.

The influence of static pair breaking by paramagnetic impurities is well known and can lead to a suppression of T_c and to gapless superconductivity.[50] In addition, electron-electron and electron-phonon inelastic scattering as well as other processes violating time-reversal symmetry give rise to dynamic pair breaking, which has been shown to be of crucial influence for the temperature dependence of the superconducting energy gap,[51] thus providing an explanation for observed deviations from BCS behavior. An estimation of the critical depairing time $\tau_{\phi c}$ for which T_c goes to zero, using the theory of Abrikosov and Gor'kov, yields $\tau_{\phi c}=2\hbar/\Delta(0)=6.6\times 10^{-14}$ s. Hence $\tau_{\phi c}/\tau_\phi=0.78$ is in a range where gapless superconductivity has been observed in conventional superconductors.[50] Consequently it may be argued that the difficulties with the measurements of the superconducting gap in high-temperature superconductors are also associated with the rather strong pair breaking in these compounds.

Finally we compare τ_ϕ to the transport scattering time in the normal state, τ_{tr}. From extrapolation of the normal-state Hall angle presented in Fig. 6, a Hall mobility $\mu_H(100\text{ K})=(\cot\Theta_H B)^{-1}=21\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ can be deduced. Assuming $\mu\approx\mu_H$ and taking the effective mass ratio $m^*/m_0=4.2$ from previous results[52] on similar thin films of $YBa_2Cu_3O_7$, we get $\tau_{tr}(100\text{ K})=\mu_H m^*/e=5.1\times 10^{-14}$ s. Since $\tau_\phi\approx 1.7\tau_{tr}$, which lies between earlier estimations from magnetoresistance measurements $\tau_\phi\approx\tau_{tr}$ (Ref. 53) and $\tau_\phi\approx 2\tau_{tr}$ (Ref. 54), we conclude that the scattering process causing the quasiparticle dephasing also is a major contribution to the scattering in the normal state, but may be complemented by other scattering processes.

V. CONCLUSIONS

We have presented measurements of the resistivity and the Hall effect in thin epitaxial films of $YBa_2Cu_3O_7$ on MgO substrates. The normal-state resistivity in the CuO_2 planes follows a temperature dependence that can be very well described by the formula proposed by Anderson and Zou and is close to a linear relationship. The cotangent of the Hall angle is proportional to T^2 as was proposed for a Luttinger liquid. Our study is based on the assumption that these two models provide at least a phenomenological description of the normal-state transport properties. Deviations, which are discernible in a limited temperature range above T_c, were interpreted as additional contributions from thermodynamic fluctuations of the superconducting order parameter and were analyzed in terms of the theories for two-dimensional layered superconductors, including the direct (Aslamazov-Larkin) and the indirect (Maki-Thompson) contributions.

We obtain a good fit to the data in the temperature range $1.01T_c<T<1.1T_c$ but note a discrepancy between theory and experiment arising at higher temperatures. It is noteworthy that this disagreement becomes significant when the indirect contribution is predominant, indicating a possible disagreement between the temperature dependence of the Maki-Thompson process calculated in a microscopical BCS model and our experiment. A similar conclusion was also proposed by Rice and Ginsberg.[55] We have shown that both paraconductivity and excess Hall effect can be fitted unequivocally by a *consistent* set of three parameters, i.e., the c-axis coherence length $\xi_c(0)$, the Maki-Thompson pair-breaking parameter δ, and the electron-hole asymmetry parameter β. In contrast to Rice *et al.*,[18] we did not observe a general discrepancy between the fits to the paraconductivity and to the excess Hall effect, which may provide an indication that our method of separating the fluctuation contributions from the normal-state properties is well applicable.

The fits to our data yield $\xi_c(0)=1.5$ Å, which is in fair agreement with the results from other determinations[7,8,18] and indicates that every two neighboring CuO_2 planes in $YBa_2Cu_3O_7$ are coupled together at all temperatures, whereas the double planes are separated, resulting in two-dimensional superconducting properties at temperatures $|T-T_c|>3$ K. The quasiparticle phase relaxation time is $\tau_\phi=8.6\times 10^{-14}$ s at 100 K and was found to be dependent on the room-temperature resistivity of the samples, which may explain the rather different values obtained in several studies.[18,28,53] The results in samples with low room-temperature resistivity provide the estimation $\tau_\phi\approx 1.7\tau_{tr}$, indicating that scattering responsible for the dephasing of quasiparticles is a significant, but probably not the sole, contribution to the normal-state scattering.

The fluctuation contributions to the Hall conductivity near T_c are dominated by the Aslamazov-Larkin process and are more divergent than the respective influence on the conductivity. The sign of this contribution is negative and reduces the Hall angle close to T_c. It is an interesting question, open to further investigations, whether a large negative fluctuation Hall effect due to the AL process can be connected with the sign change of the Hall effect in the mixed state of high-temperature superconductors.

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