

# Microwave measurements of the in-plane and $c$ -axis conductivity in $\text{HgBa}_2\text{CuO}_{4+\delta}$ : Discriminating between superconducting fluctuations and pseudogap effects

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An approach to microwave measurements is used in order to determine both, the in-plane and out-of-plane conductivity of the high- $T_c$  superconductor  $\text{HgBa}_2\text{CuO}_{4+\delta}$  near optimal doping. Unlike the  $ab$ -plane conductivity, the  $c$ -axis conductivity is highly sensitive to superconducting fluctuations. From a single  $c$ -axis data set, we can clearly discern the opening of the pseudogap at  $T^* = 185(15)$  K, the appearance of the superconducting fluctuations at a much lower temperature  $T' = 105(2)$  K, and the full transition to the superconducting state at the critical temperature  $T_c = 94.3$  K. Thus, with the present high sensitivity, we establish that the extent of the superconducting fluctuations is only about 10 K above  $T_c$ .

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## I. INTRODUCTION

The high-temperature cuprate superconductors exhibit anomalous charge and magnetic properties due to the opening of a normal-state gap—the “pseudogap”—at a temperature  $T^*$  above the superconducting (SC) transition temperature  $T_c$ . The pseudogap regime has been argued to be either a precursor state to superconductivity<sup>1–4</sup> or to be associated with some “hidden” order that competes with the superconductivity.<sup>5–8</sup> Indications for an intermediate superconducting fluctuation regime are observed from the vortex Nernst effect,<sup>9</sup> torque magnetometry,<sup>10</sup> NMR relaxation rates,<sup>11</sup> tunneling,<sup>12</sup> as well as dc (Ref. 13) and frequency-dependent conductivity measurements.<sup>14</sup> Nevertheless, the highest temperature at which the superconducting fluctuations can be observed, although of utmost importance for understanding of the possible connection between  $T^*$  and  $T_c$ , is still not unambiguously determined. A problem with many experimental techniques is the subtraction of nonsuperconducting contributions to the total signal. Usually, this procedure involves some assumptions about the normal-state behavior, and/or a theoretical model to extract the unknown variables. Thus, it has been recently shown that the Nernst effect, considered a pivotal experiment for the determination of the fluctuation regime, should be interpreted with care.<sup>15,16</sup> Consequently, an alternative experimental verification of the SC fluctuation regime is desirable.

In this paper, we apply a very sensitive method for measuring the microwave conductivity to nearly optimally doped  $\text{HgBa}_2\text{CuO}_{4+\delta}$  ( $\text{Hg1201}$ ) single crystals. The present approach has several advantages in comparison with other experimental techniques: (i) for the first time the same measurement unambiguously determines three distinct characteristic temperatures: the pseudogap temperature  $T^*$ , the temperature  $T'$  associated with the extent of the super-

conducting fluctuations, and  $T_c$ ; (ii) our method does not rely on any theoretical assumption or subtraction of different contributions, since the characteristic regimes are clearly apparent already from the raw data. The temperature at which SC fluctuations appear is furthermore confirmed by microwave measurements made in strong magnetic fields, which shift  $T_c$  to lower temperatures, but leave  $T'$  and the conductivity above  $T'$  unchanged. We find that  $T'$  is only 10 K above  $T_c$  and hence well below  $T^*$ , which demonstrates the clear distinction between the SC fluctuations and the processes governing the pseudogap.

## II. EXPERIMENTAL

Recent progress in the synthesis<sup>17</sup> and doping control<sup>18</sup> of  $\text{Hg1201}$  single crystals offers an exceptional opportunity for a systematic investigation of the intrinsic properties of the cuprate superconductors.  $\text{Hg1201}$  possesses the highest  $T_c$  at optimal doping of all single-layer cuprates, a simple tetragonal crystal structure with a large spacing between the  $\text{CuO}_2$  layers, and is thought to be relatively free of disorder effects.<sup>19</sup>

The samples studied here were slightly underdoped single crystals. They were characterized according to the procedure described in Ref. 18, and exhibit a single and narrow transition close to 95 K, with a zero-field/field-cooled susceptibility ratio exceeding 75%. The primary microwave measurements were made on a cleaved sample with dimensions  $1.9 \times 0.6 \times 0.1$  mm<sup>3</sup>, where the smallest direction coincides with the crystallographic  $c$  axis. Measurements on other samples with uncleaved surfaces and of different geometries are in agreement with results presented for the primary sample.

The microwave loss measurements [ $1/(2Q)$ ] were made in an elliptic cavity using  ${}_{\epsilon}\text{TE}_{112}$  and  ${}_{\epsilon}\text{TE}_{211}$  resonant modes

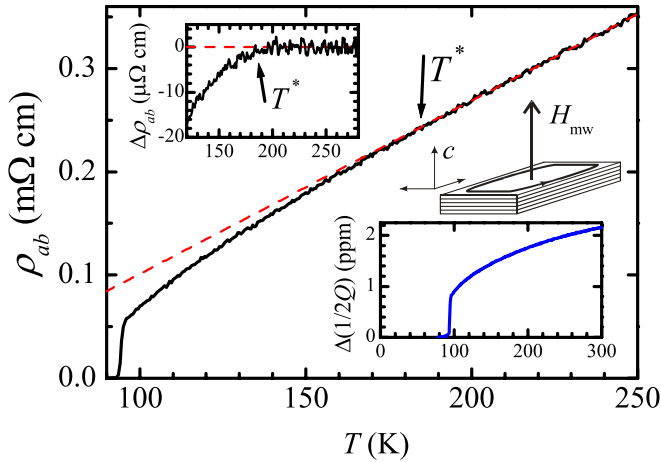


FIG. 1. (Color online) Temperature dependence of  $\rho_{ab}$  in the single crystal of nearly optimally doped  $\text{HgBa}_2\text{CuO}_{4+\delta}$  (full line) calculated from the measured microwave absorption (lower inset, with  $H_{\text{mw}} \parallel c$ ). The dashed line represents extrapolated linear behavior above 200 K. Upper inset: difference between  $\rho_{ab}$  and the linear behavior extrapolated to low  $T$  clearly reveals  $T^*$ .

at frequencies of 13.14 and 15.15 GHz, respectively. The high sensitivity in measuring changes in the relatively low  $Q$  factor of the copper cavity loaded with the sample was achieved using audio modulation of microwaves followed by a harmonic detection in the output signal, as described previously.<sup>20</sup> The use of a (nonsuperconducting) copper cavity had the benefit of allowing measurements in applied high magnetic fields without loss of sensitivity.

The samples were mounted on a sapphire cold finger in the cavity center, where the microwave magnetic field  $H_{\text{mw}}$  has its antinodal position for both modes used in the experiment. The polarizations of  $H_{\text{mw}}$  in the two modes are mutually orthogonal, which allowed the measurement of two distinct signals with one and the same sample mount.

### III. MICROWAVE ABSORPTION IN $ab$ -PLANE

First, we present the temperature dependence of the microwave absorption in the configuration  $H_{\text{mw}} \parallel c$ , where the induced currents flow only in the  $ab$  plane (lower inset of Fig. 1, resonant mode  ${}_e\text{TE}_{211}$ ). From the room-temperature resistivity  $\rho_{ab}(300 \text{ K}) \approx 0.44 \text{ m}\Omega \text{ cm}$ , the skin depth in this configuration can be estimated to be less than  $10 \mu\text{m}$  at all temperatures, which is much less than the sample dimensions. In this case, one can use the classical skin-effect relation  $\Delta[1/(2Q)] \propto \sqrt{\rho_{ab}}$ , in order to extract  $\rho_{ab}$  from the measured data. Its temperature dependence is shown in the main panel of Fig. 1. The observed behavior of  $\rho_{ab}$  is reminiscent of the well-known in-plane dc transport results. Following well established procedure,<sup>21</sup> we determine the pseudogap temperature  $T^* = 185(15) \text{ K}$  from the deviation of the planar resistivity from the linear, high-temperature metallic behavior characteristic of the cuprates (upper inset of Fig. 1).

It is worth noting that, in the normal state, microwave measurements should yield the same results as dc resistivity measurements. One advantage of the microwave technique is

that no electrical contacts are required, so that problems related to an inhomogeneous injection of the current are avoided.

The appearance of SC fluctuations, which bring about excess conductivity, reduces the microwave absorption as  $T_c$  is approached from above. This effect is also included in the behavior observed in Fig. 1, but cannot be separated from the processes that reduce the resistivity below  $T^*$ . The SC fluctuations will be better seen in the  $c$  axis conductivity presented in the next Section.

From the data in Fig. 1, we determine  $T_c = 94.3 \text{ K}$  as the midpoint of the superconducting transition. The width of the transition is  $\Delta T_c = 1.2 \text{ K}$ , as determined by 10–90 % criterion. Using this value of  $T_c$ , we note that the pseudogap temperature obtained here is consistent with the linear trend established for underdoped samples from polarized neutron diffraction<sup>22</sup> and dc transport.<sup>23</sup>

### IV. $c$ -AXIS CONTRIBUTION TO MICROWAVE ABSORPTION

The data analysis is more complex when  $H_{\text{mw}}$  lies in the  $ab$  plane, since the induced microwave currents flow partly along the  $c$  direction, and partly in the  $ab$  plane. In this case, there are two relevant skin depths:  $\delta_c = \sqrt{2\rho_{ab}/(\mu_0\omega)}$  for the microwave penetration along the  $c$  axis with the shielding currents in the  $ab$  plane, and  $\delta_{ab} = \sqrt{2\rho_c/(\mu_0\omega)}$  for the microwave penetration along  $a$  and  $b$  directions, which depends on the current path associated with the  $c$  axis. The latter plays the dominant role in the presently observed microwave absorption signal because the anisotropy  $\rho_c/\rho_{ab}$  in  $\text{Hg}1201$  is on the order of 1000.<sup>24</sup> Moreover, the value of  $\rho_c$  is large enough so that the skin depth  $\delta_{ab}$  can be comparable to the width of the sample, leading to two distinct measurement geometries.

If the sample is wider than  $2\delta_{ab}$ , i.e., the microwave current does not fully penetrate the sample (opaque sample), the microwave losses increase as expected with an increase in  $\rho_c$ . However, if the sample is narrower than  $2\delta_{ab}$ , the microwave field penetration is achieved throughout the whole sample volume (transparent sample), in which case a counterintuitive effect is observed: when  $\rho_c$  increases, the microwave losses diminish. Namely, the microwave shielding currents are reduced, and the sample becomes even more transparent, with less energy being dissipated in the sample. Obviously, the microwave absorption reaches its maximum between the two regimes, seen as a so-called “loss-peak” in the absorption temperature dependence, when the sample width becomes comparable to  $2\delta_{ab}$ .<sup>25,26</sup> On each slope of the loss peak one obtains an unusually high sensitivity of the absorption for a small change in the  $c$ -axis conductivity.

We have used this effect to our advantage on a sample of convenient geometry, and observed two completely different microwave absorption signals simply by rotating the sample with respect to  $H_{\text{mw}}$ . In Fig. 2 we show two measurements of the temperature-dependent microwave absorption with  $H_{\text{mw}}$  in the  $ab$  plane (resonant mode  ${}_e\text{TE}_{211}$ ). In the first, the sample was oriented so that its shorter edge was aligned with  $H_{\text{mw}}$  [Fig. 2(a)]. The penetration of the microwaves then oc-

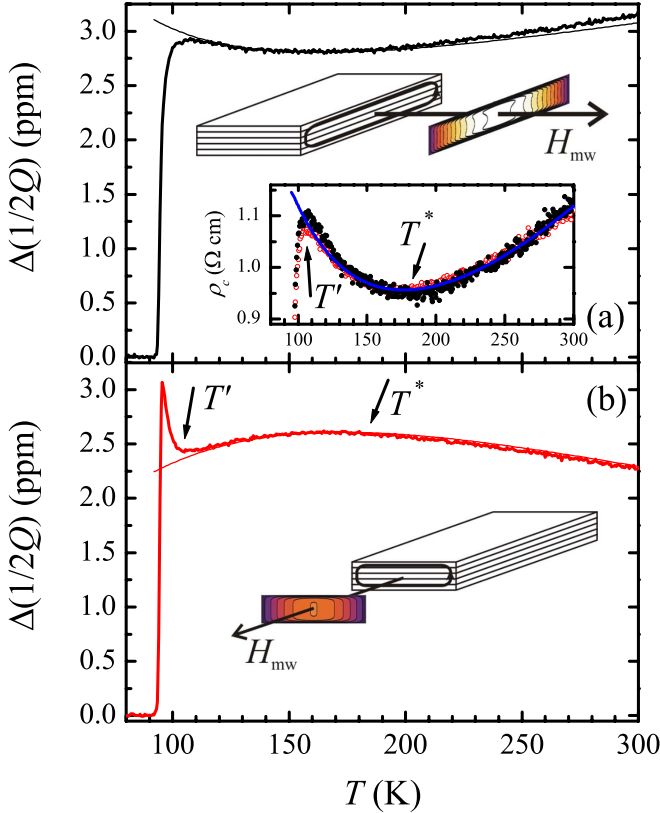


FIG. 2. (Color online) Microwave absorption for two geometries with  $H_{mw}$  parallel to the  $ab$  plane. Screening currents have in-plane and out-of-plane contributions. The conductivity along the  $c$  axis is more than  $10^3$  times smaller than that parallel to the  $ab$  planes and consequently the skin depth is  $10^{3/2}$  times larger. Thus we distinguish two cases: (a) when  $H_{mw}$  is aligned with the sample's shorter edge, the perpendicular longer edge acts as the sample's width which is then larger than  $2\delta_{ab}$ , so that the microwave field is completely screened deeply in the sample bulk, (b) when  $H_{mw}$  is along the sample's longer edge, it is the sample's shorter edge, which turns out to be smaller than  $2\delta_{ab}$ , that acts as its width. Consequently, the microwave currents flow throughout the sample volume and do not completely screen the field, i.e., the sample is transparent to some degree. These two experimental configurations are shown in the insets of the panels (a) and (b), where profiles of the microwave magnetic field across the sample are schematically represented by the gradual change in the color intensity. The lower inset of panel (a) shows  $\rho_c$  obtained from both measured orientations, black and light gray (red) symbols, respectively. The dark gray (blue) line is the fit to the model described in the text. Thin lines in the main panels (a) and (b) are calculated temperature dependencies of the microwave absorption as explained in the text.

curs along the longer edge, and does not reach the center of the sample. In the second case, the sample holder was rotated to align the longer edge of the sample with  $H_{mw}$  [Fig. 2(b)], so that full penetration of the microwaves into the sample is achieved due to the shortness of the perpendicular edge. It is in this geometry that we obtain the highest sensitivity of the microwave absorption to small changes of the  $c$ -axis conductivity.

Unlike the microwave absorption with  $H_{mw} \parallel c$  as shown in Fig. 1, the geometry of Fig. 2(b) allows us to clearly discern the SC fluctuations as a change in the sign of the slope of the microwave absorption: upon cooling toward  $T_c$ , the absorption abruptly increases below  $T' = 105(2)$  K.

The data in Fig. 2(b) furthermore exhibit an absorption maximum slightly below  $T^*$ , where an increasing contribution to  $\rho_c$  starts to develop upon cooling. In the present geometry, the microwave measurement has the distinct advantage that enables the simultaneous identification of the three characteristic temperatures  $T_c$ ,  $T'$ , and  $T^*$ . In contrast to other experimental techniques, these temperatures are here deduced from the very same experimental curve, without any data subtraction, and/or analysis based on theoretical models. We find that  $T'$  is close to  $T_c$  and well below  $T^*$ .

## V. ANALYSIS OF THE $c$ -AXIS RESISTIVITY

Besides the very identification of the three characteristic temperatures, it is interesting to analyze the temperature dependence of  $\rho_c$ . According to the model developed by Gough and Exon,<sup>25</sup> the microwave absorption in a rectangular rod of a cross section  $a \times c$  can be expressed by

$$\frac{1}{2Q} \propto \text{Im} \sum_{nm} \frac{1}{n^2 m^2} \frac{\Lambda_{nm}}{\Lambda_{nm} + i\omega}, \quad (1)$$

where

$$\Lambda_{nm} = \frac{\omega \pi^2}{2} \left[ \frac{n^2}{(a/\delta_c)^2} + \frac{m^2}{(c/\delta_{ab})^2} \right], \quad (2)$$

and the sum is over odd integers.  $\delta_c$  is given by the in-plane resistivity  $\rho_{ab}$  known from Fig. 1. Thus, from the measurements in Fig. 2, having the defined sample geometry, and taking into account the demagnetization effect, one can extract the out-of-plane resistivity  $\rho_c = \mu_0 \omega \delta_{ab}^2 / 2$  for each orientation. These  $\rho_c$  values numerically obtained from the two measurements are plotted in the inset of Fig. 2(a). A very similar result for the temperature dependence and absolute value of  $\rho_c$  was obtained for a second sample with  $T_c \approx 95$  K.

The obtained  $\rho_c$  is approximately 2000 times larger than  $\rho_{ab}$ . It is important to note that  $\rho_c$  exhibits its minimum roughly at the same temperature  $T^*$  at which  $\rho_{ab}$  starts to deviate from linearity. Consequently, even though interplanar disorder affects the  $c$ -axis transport, the semiconductorlike behavior of  $\rho_c$  below  $T^*$  is associated with the opening of the pseudogap.<sup>13,21,24,27,28</sup> The temperature dependence of the out-of-plane resistivity can be modeled by  $\rho_c = (a + bT^\alpha) / n_c(T)$  where  $n_c(T) = n_{c0} + n_{c1} e^{-T^*/T}$  is the effective number of charge carriers contributing to the  $c$ -axis transport.<sup>29</sup> It is worth noting that the data obtained from apparently different curves measured in two different sample orientations coincide. The fit shown by the thin line in the inset of Fig. 2(a) yields the parameters  $\alpha = 1.66$ ;  $n_{c0}/n_{c1} = 0.18$ ;  $a/n_{c1} = 0.3$   $\Omega$  cm; and  $b/n_{c1} = 0.04$  m $\Omega$  cm K<sup>-1.7</sup>. While a good description of the normal-state data is obtained at high temperatures, deviations occur in a narrow region above  $T_c$ : a strong downturn at  $\approx 105$  K [inset of Fig. 2(a)],



which coincides with the value of  $T'$  obtained directly from the data in Fig. 2(b).

In order to analyze the distinct features in the data shown in Fig. 2, we used the fit for  $\rho_c(T)$  shown in the inset of Fig. 2(a) to calculate the expected absorption  $\Delta(1/2Q)$  for each of the two sample orientations. The resulting curves are plotted as thin lines in the main panels of Fig. 2. At higher temperatures the calculated curves coincide well with the data in both crystal orientations. The deviations occur at  $T' \approx 105$  K in both cases, with the experimentally measured curves changing the sign of their respective slopes. It is clear that a new physics emerges at this temperature, and we ascribe it to the appearance of the SC fluctuations.

The discussion of the effect of superconductivity on the overall microwave absorption signal requires some additional consideration. Namely, as long as only the “normal” electrons contribute to the transport properties, we can equally well consider either the resistivity  $\rho$  or the conductivity  $\sigma = 1/\rho$  because they are both real quantities. However, when Cooper pairs are formed, the “normal” and “superconducting” channels act in parallel and the formalism involving the complex conductivity  $\tilde{\sigma} = \sigma_1 - i\sigma_2$  is required.<sup>30</sup>

The remarkable peak observed in the microwave absorption shown in Fig. 2(b) (transparent sample), can be described in a simple manner. Upon cooling from room temperature,  $\sigma_c$  first increases giving rise to a larger absorption. Below  $T^*$ ,  $\sigma_c$  starts to decrease, which is seen as a gradual decrease in absorption. However, this trend is inverted at  $T' \approx 105$  K, because  $\sigma_c$  sharply increases. We attribute this sudden increase of conductivity to the appearance of sizeable superconducting phase correlations detectable at our probe frequency of 15.15 GHz. In other words, the real part of the conductivity  $\sigma_{1c}$  grows due to these fluctuations. The imaginary part  $\sigma_{2c}$  also emerges,<sup>31</sup> but in the fluctuation regime above  $T_c$  it is much smaller than  $\sigma_{1c}$ . Only below  $T_c$ , where the long-range correlation in the phase of the Cooper pairs is established, does the imaginary part of the conductivity prevail giving rise to a sharp drop in absorption.

## VI. MAGNETIC FIELD DEPENDENCE

Superconducting fluctuations add in parallel with the normal-state conductance. Given that the  $c$ -axis conductivity  $\sigma_c$  is 2000 times smaller than  $\sigma_{ab}$ , a very small fluctuation conductivity could be a large percentage increase in  $\sigma_c$ , but a negligible increase in  $\sigma_{ab}$ . That makes  $\sigma_c$  the transport parameter most sensitive to the superconducting fluctuations.

In order to show that the increased conductivity between 95 and 105 K is indeed related to superconductivity, we performed measurements with an external dc magnetic field applied parallel to the sample's  $c$  axis. The resulting absorption curves are displayed in Fig. 3. As seen from this figure, already a 3 T magnetic field strongly affects microwave absorption close to  $T_c$ . Significantly, the three curves perfectly coincide at high temperatures but separate below 105 K. Clearly, the absorption near the pseudogap temperature  $T^*$  is insensitive to fields as large as 8 T. In contrast, the superconducting fluctuation conductivity, which sets in below  $T'$ , is strongly affected.

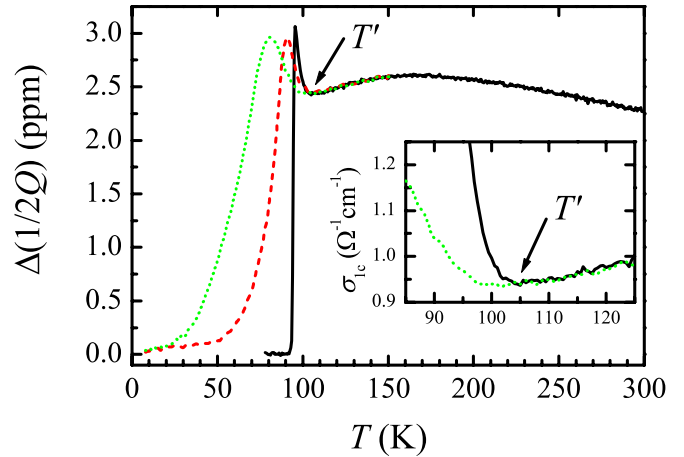


FIG. 3. (Color online) Microwave absorption measured with the external dc magnetic field applied in the  $c$  direction, in the same geometry as in the case of Fig. 2(b). The applied fields were 0, 3, and 8 T, plotted in full (black), dashed (red) and dotted (green) lines, respectively. Inset: temperature dependence of  $c$ -axis conductivity calculated from the microwave absorption data for zero field (full black line) and for 8 T (dotted green line).

The calculated  $c$ -axis conductivity is plotted in the inset of Fig. 3. We note that the appearance of the fluctuation conductivity shifts by about 1K/T, as expected for SC fluctuations. Hence, the measurements in dc magnetic fields give decisive proof that the superconducting fluctuations extend only up to  $T'$ .

We note that the narrow temperature range of superconducting fluctuations ( $T' - T_c \approx 10$  K) is in agreement with Josephson tunneling measurements on YBCO(Co),<sup>12</sup> as well as the recent Nernst effect measurements on YBCO (Ref. 32) and LSCO doped with Nd and Eu.<sup>15</sup> Although these measurements did not allow precise determination of  $T'$ , they indicate that the SC fluctuation regime is much narrower than previously reported.

## VII. CONCLUSIONS

We have demonstrated that our microwave approach constitutes a highly sensitive and reliable transport measurement. For the model superconductor Hg1201, our measurement allows the direct identification of the superconducting transition temperature  $T_c$ , the pseudogap temperature  $T^*$ , as well as a third characteristic temperature  $T'$  in zero magnetic field. By applying a uniform magnetic field, we have shown that  $T'$  is associated with the superconducting fluctuations. Since the  $c$ -axis conductivity is three orders of magnitude more sensitive to fluctuations than the  $ab$ -plane conductivity,  $T'$  can be identified as the upper limit of fluctuations. It seems unlikely that any other transport parameter would be more sensitive to fluctuations than  $\sigma_c$ . Hence, we have shown that fluctuations do not extend up to  $T^*$ , and that they are confined to a very small region above  $T_c$ .

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