Dynamic conductivity near T_c of $YBa_2Cu_3O_7$ films: Vortex-glass-like criticality in the absence of externally induced vortices

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(Received 5 September 1996)

Using a contactless method, we investigated the electrical ac conductivity $\sigma' + i\sigma''$ in the CuO_2 planes of thin Y-Ba-Cu-O films deposited on (001)-$SrTiO_3$ and -MgO substrates at zero magnetic field. The temperature variation of the phase angles, $\varphi(T;\omega) = \arctan(\sigma''/\sigma')$, measured between 30 Hz and 2 GHz, allows a precise definition of the superconducting transition T_c. Remarkably, the frequency-independent value $\varphi(T_c) = 74°$, obtained for both films, agrees with those observed previously for high vortex densities (0.1–19 T), where this fact was associated with the universality near the so-called vortex-glass transition line. Since the shape of the present scaling function for $\sigma(\omega)$ also proves to be identical with that found in a high field, we argue that the same fluctuations destroy superconductivity in zero field and in the presence of a dense vortex lattice. [S0163-1829(97)07709-6]

I. INTRODUCTION

For both applicational and fundamental reasons, investigations of the frequency variation of the Ohmic and reactive components of the linear conductivity, $\sigma' + i\sigma''$, represent one of the important approaches to elucidate the nature of high-T_c superconductivity. Quite remarkably, most of the existing experimental studies concentrated on the effect of external magnetic fields B on the superconducting transition which, according to the seminal hypothesis of Fisher and co-workers,[1] should remain continuous along a so-called vortex-glass (VG) line $T_g(B)$ located distinctly below the so-called mean-field line $T_{c_2}(B)$. The predicted scaling behavior of $\sigma(\omega)$ (Refs. 1 and 2) has been realized near T_g of $YBa_2Cu_3O_7$ (YBCO) thin films[3,4] and crystals.[5,6] However, its interpretation is still controversal. While uncorrelated weak pinning of vortices by point defects[1] is now thought to be insufficient to produce stable superconductivity,[7,8] it appears more likely that correlated pinning by extended defects[7] provides a relevant mechanism. Recently, a VG line determined from $\sigma(\omega)$ data on a YBCO film with a high density of columnar pins turned out to be in substantial agreement up to the matching field $B_\phi = 4$ T (Ref. 9) with the prediction for the so-called Bose localization of single vortices along the defects,[7] thus supporting this idea.

Here we present the first wide band study (30 Hz to 2 GHz) of the conductivity parallel to the CuO_2 planes of thin epitaxial YBCO films in the limit of vanishing induced vortex density where fluctuations of the superconducting order parameter rather than fluctuations and interactions in the vortex system[1] should dominate in $\sigma(\omega)$. It is our primary goal to determine the singular behavior of $\sigma(\omega)$ near the transition temperature T_c, arising from the critical divergences of the relevant correlation length, $\xi \sim |T - T_c|^{-\nu}$, and correlation time, $\tau \sim \xi^z$, and also the scaling function of $\sigma(\omega)$ characteristic for the continuous superconducting phase transformation. Recent scaling analyses of nonlinear I-V curves of thin films[10,11] revealed a yet unexplained increase of z from the ''universal'' high-field VG value, $z = 5.7(3)$,[4,12] and a corresponding decrease of ν from $\nu = 1.7(1)$ in fields below 0.1 T. On the other hand, no such changes have been observed by I-V measurements on granular YBCO (Ref. 13) down to 0.1 mT.

II. EXPERIMENT AND RESULTS

In order to check for possible effects of the substrate material on the ac conductivity near T_c, we have selected two different YBCO films for this study. The first was grown on (001)-$SrTiO_3$ (''STO film'') to a thickness of 250 nm by laser ablation[14] and shaped to a disk with radius 1.5 mm. The VG behavior of this sample between 0.1 T and 19 T has been evidenced recently.[4,12] The second, a 100 nm thick ''MgO film,'' was prepared by magnetron sputtering on (001)-MgO,[15] which provides a larger lattice mismatch (8%) than STO (1.2%) but which, due to the much smaller dielectric permeability, exhibits favorable microwave properties. For the present measurements we cut a 2×2 mm^2 square. Atomic force microscopy (AFM) pictures of the surfaces of both films revealed a rather similar lateral granularity of mean size of 100 nm.

The evaluation of the linear ac conductivity parallel to the CuO_2 planes of thin films from the linear dynamic magnetic susceptibility $\chi(\omega)$, measured perpendicular to the film plane, has been outlined in some detail in Ref. 4. The exact inversion procedures used here for disks and squares have been based on the extensive works of Brandt.[16] Up to 50 MHz, the measuring technique for $\chi(\omega)$ was identical to that described in Ref. 4. Between 70 MHz and 2000 MHz, the real and imaginary parts of $\chi(\omega)$ were determined from shifts of the resonance frequency and the quality factor, respectively, of helical resonators[17,18] under the slow change of sample temperature.

The samples were mounted on sapphire holders which extended from the cold finger of a closed-cycle refrigerator (Oxford CC1000) to the resonators. All data shown below have been measured in ambient laboratory field of 50 μT. With a long copper solenoid wrapped around the cold cylinder we checked the insensitivity of $\chi(\omega)$ against small changes of the dc field in the order of ± 50 μT near T_c where the field effect should be the largest. From the independence of the ac response against variations on the order of the Earth field we infer that we study the zero-field limit.

As noted previously,[4,6] the most detailed information on the superconducting transition is not contained in the single components of $\sigma(\omega)=\sigma'+i\sigma''$, but in the phase angle $\varphi(\omega,T)=\arctan(\sigma''/\sigma')$. The results obtained near T_c of both films are displayed in Fig. 1, which for completeness shows also the moduli of $\sigma(\omega)$ as insets. A striking feature is the appearance of a well-defined temperature, where the phase angles of all isofrequency curves intersect and where $d\varphi/d\omega$ changes sign. This feature of $\sigma(\omega\rightarrow 0)$ marks unambiguously the transition to long-range superconductivity, which on a phenomenological level may be understood as follows: At low frequencies the metallic phase is characterized by the Drude dynamics $\sigma(\omega)=\sigma_0/(1-i\omega\tau)$, whence $d\varphi/d\omega>0$, while in the superconducting phase, according to the two-fluid model, the onset of bulk screening gives rise to $\sigma(\omega)=\sigma_0-1/i\omega\mu_0\lambda_L^2$. This implies $d\varphi/d\omega<0$, but the value of the frequency-independent phase angle $\varphi(T_c)$ at T_c, of course, cannnot be inferred from these simple arguments. Here we only emphasize that regardless of the different substrates, film thicknesses, and the different transition temperatures T_c=89.02(1) K and 90.97(2) K for the MgO and STO films, respectively, the critical phase angles are identical for both, $\varphi(T_c)=\arctan(\sigma''/\sigma')=74(1)°$. According to the Kronig-Kramers relation for the linear response, this result for the phase demands a power law for the modulus of the conductivity at T_c, $|\sigma(\omega,T_c)|\sim\omega^{-x}$, where $x=\varphi(T_c)/(\pi/2)$.[2] In fact, the data shown for both films by the insets to Fig. 1(a) and 1(b) obey the expected behavior, depicted by the solid lines in Fig. 1(c).

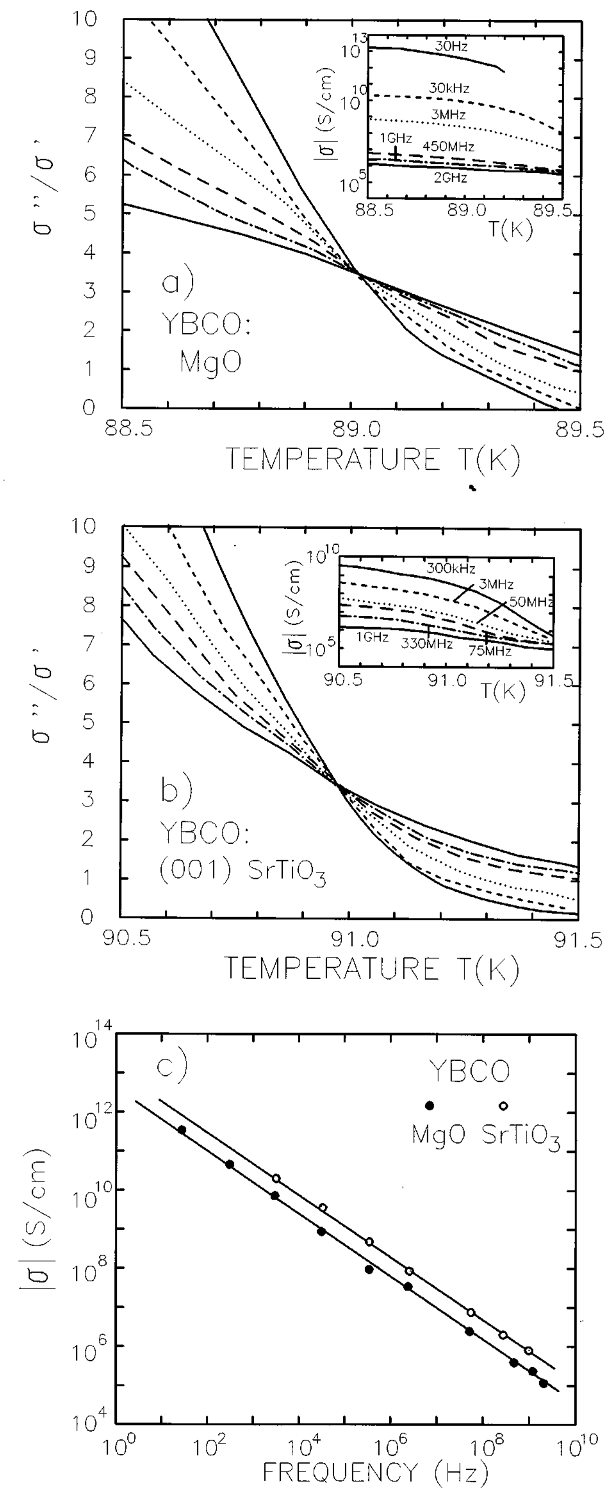

FIG. 1. Temperature variation of the phase and of the modulus (inset) of the dynamical conductivity of YBCO films on (a) MgO and (b) $SrTiO_3$ between 30 Hz and 2 GHz in zero applied field. (c) Frequency dependence of $|\sigma|$ at T_c and power laws, $|\sigma|\sim\omega^{-x}$, with $x=\varphi(T_c)/90°$ (see text).

III. VORTEX-GLASS-LIKE CRITICAL DYNAMICS

Based on pure relaxational, time-dependent, i.e., Ginzburg-Landau (TDGL) dynamics, for which $z=2$, the phase angle of $\sigma(\omega)$ at T_c of a D-dimensional homogeneous superconductor has been calculated by Dorsey[19] using for the superfluid density, $\rho_s\sim\xi^{2-D}$, and treating thermal fluctuations in the Gaussian approximation. By means of scaling arguments his result was generalized to include critical fluctuations[1] and also uncorrelated isotropic disorder:[2]

$$\varphi(T_c)=\frac{\pi}{2}\left(1-\frac{D-2}{z}\right). \quad (1)$$

For this so-called VG model the dynamic exponent increases, $z=2(2-\eta)$,[2] where η is zero in the mean-field approximation and may become negative in the critical regime, which would lead to $z>4$. A reliable theoretical estimate for η, however, is not yet available.

For correlated disorder, in the form of columnar defects, Wallin and Girvin[20] predict within the so-called Bose glass (BG) model[7] $z=6\pm0.5$ for currents perpendicular to the pins. This result is rather close to that observed here at

$B=0$ and also on YBCO films in large fields.[4,9] Because the existence of columnar pins is not obvious in pristine, epitaxial films, it appears more likely that their microtwin and granular network provides correlated pinning perpendicular to the current in the ab planes. Then the BG model would be valid if the vortex fluctuations were confined to the twin planes,[7] which appears not implausible.

As another possible origin of the observed critical dynamics we consider topological fluctuations, i.e., vortex-antivortex pairs formed within the CuO_2 planes. Their effect on the I-V curves and the linear resistivity below and above T_c has been reported for YBCO films.[21,22] The data were interpreted in terms of the jump of ρ_s at the pair unbinding Kosterlitz-Thouless (KT) transition, $T_{\rm KT}$, of thin [two-dimensional (2D)] superconducting films.[23] To our knowledge, a possible crossover to 3D critical dynamics has not yet been worked out.

In this context we should mention a critical dynamics discovered by Jonsson and Minnhagen,[24] who performed simulations for a 2D XY-type model with TDGL dynamics. Assuming a nearest-neighbor interaction of the form $U(\phi_{ij}) = 2J[1-\cos^{2p^2}(\phi_{ij}/2)]$, where ϕ_{ij} denotes the phase difference of the order parameter between sites i and j on a square lattice, they evaluated for the phase of the dynamic conductivity near $T_c=T_{\rm KT}$ essentially the same behavior as shown here in Figs. 1(a) and 1(b). Remarkably, the phase angle they found at the KT transition, $\varphi(T_{\rm KT})=75.1°\pm0.4°$, agrees almost exactly with our value $74°\pm1°$ which raises the challenging question of whether the topological 2D fluctuations considered in Ref. 24 determine the behavior of the YBCO films at T_c. One of the open questions concerning this simulation is to what extent the special choice of the exponent $p=2.5$, being larger than $p=1$ for the generic XY model and introduced to more easily excite vortices, has an influence on the phase angle at T_c.

On a phenomenological level, the most intriguing feature emerging from the critical behavior of $\sigma(\omega)$ at T_c is the coincidence of the phase angle obtained here (Fig. 1) in zero magnetic field with that determined in applied fields between 0.1 T and 19 T at the VG line $T_g(B)$ of the present STO film[4,12] and with that of almost all films reported in the literature (''universal'' z values, compiled, e.g., in Refs. 6 and 10). As noted in the Introduction, the linear dynamic conductivity displays scaling behavior near $T_g(B)$ which is characteristic for a continuous thermodynamic phase transition, and which has been hypothesized for both the VG (Ref. 1) and the BG.[7] This scaling property arises from the diverging correlation length $\xi\sim|1-T/T_g|^{-\nu}$ and time $\tau\sim\xi^z$ of the order parameter fluctuations. Then the phase and the amplitude of the dynamic conductivity must obey the forms[1,2]

$$\arctan(\sigma''/\sigma') = P_\pm(\omega\tau), \tag{2a}$$

$$|\sigma(\omega)| = \sigma_0 S_\pm(\omega\tau), \tag{2b}$$

where $P_\pm$ and $S_\pm$ are homogeneous functions defined above (+) and below (−) T_c.

Regarding the identical behavior of $\sigma(\omega)$ determined here for $B=0$ at T_c and for high vortex densities along the VG line $T_g(B)$, it is near at hand to search for the scaling feature also in the vicinity of T_c. The results for both films based on

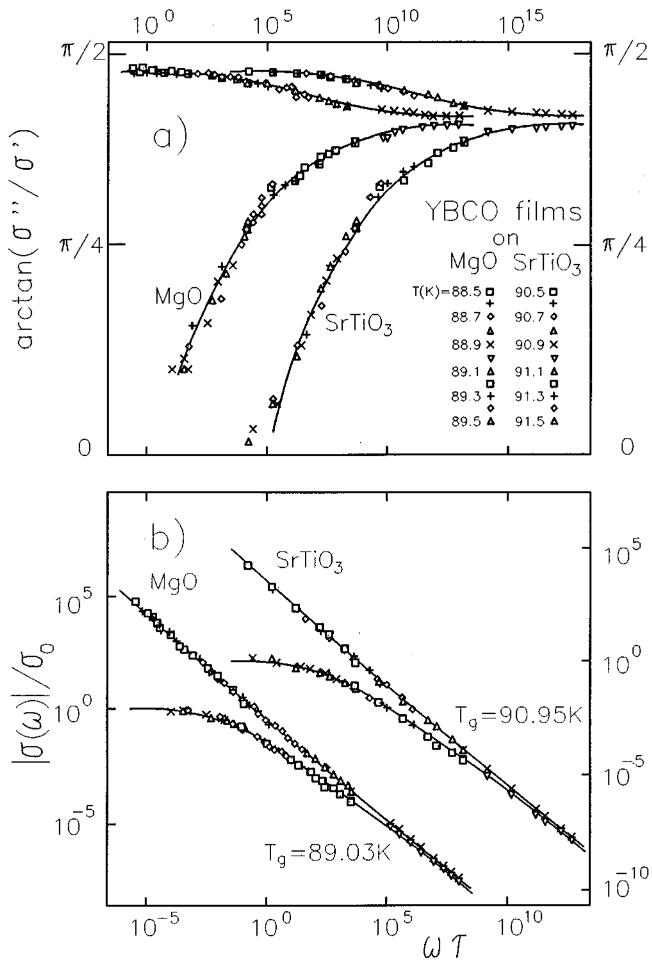

FIG. 2. Scaling functions of (a) the phase and (b) the modulus of the dynamical conductivity of both films (upper and left scales refer to the MgO film); the solid line represents the ''universal'' scaling function near the vortex-glass transition, obtained between 0.4 and 19 T in Refs. 4 and 12.

the data of Figs. 1(a) and 1(b), i.e., from the temperature interval $|1-T/T_c|\leqslant0.006$, are shown in Fig. 2. In fact, on either side of T_c we find two pairs of curves for the phase $P_\pm(\omega\tau)$ and the scaled modulus $S_\pm(\omega\tau)$, on which all data collapse. The scaling feature is perhaps less surprising for the zero-field phase transition than that near the VG line $T_g(B)$. However, as the most striking result we consider the fact that also the *shapes of the scaling functions* $P_\pm(\omega\tau)$ and $S_\pm(\omega\tau)$, determined for both films, are identical with those determined near the VG line of the STO film for a wide range of vortex densities.[4] These ''VG scaling functions'' are drawn as solid lines in Fig. 2.

The relaxation rates τ^{-1}, which provide the scaling of the frequency, and the length $\lambda_s(T)\equiv(\tau(T)/\sigma_0(T)\mu_0)^{1/2}$, derived from $\tau(T)$ and the scaling parameter of the modulus, σ_0, are depicted in Fig. 3. The temperature variation of both quantities can be described by the well-known power law singularities

$$\tau^{-1}(T) = \tau^{-1}|1-T/T_c|^{\nu z}, \tag{3}$$

$$\lambda_s(T) = \lambda_s|1-T/T_c|^{-\nu/2}. \tag{4}$$

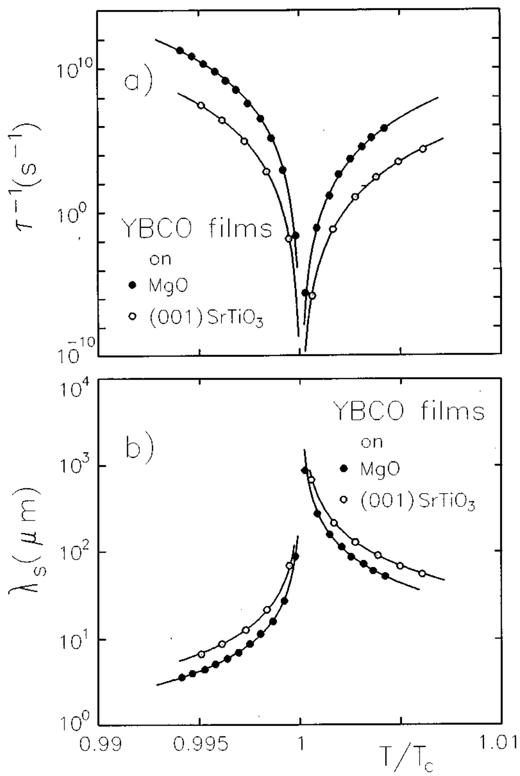

FIG. 3. Critical behavior of (a) the relaxation rates and (b) the screening lengths obtained from the scaling parameters σ_0 and τ for the modulus and the frequency used in Fig. 2.

Here we have presented the length λ_s rather than σ_0, because this length has a physical meaning on both sides of T_c: λ_s represents the distance along which a normal (or superconducting) fluctuation diffuses within its lifetime τ below (or above) the transition temperature. The critical exponents fitted to Eqs. (3) and (4), $z=5.6(1)$ and $\nu=1.7(1)$, agree excellently with the ''universal'' values of YBCO films, determined previously[4,10] in high fields. Moreover, inserting $z=5.6$ in Eq. (1) and supposing $D=3$, the phase angle measured at T_c, $\varphi(T_c)=74°$, is reproduced.

IV. CONCLUSIONS

From an empirical point of view, the almost perfect agreement between the critical dynamics near the transitions to stable superconductivity in zero field and in the presence of a high density of externally induced vortices suggests that the fluctuations driving these transitions have a common origin and that vortex-lattice effects are absent. This holds at least for YBCO films, whereas single crystals are characterized by smaller values, $z\approx 3$,[6] and, hence, belong to a different universality class of critical behavior. Because this ''film universality'' occurs in fields up to at least 19 T,[12] corresponding to mean vortex distances down to 12 nm, the symmetry of the fluctuations responsible for the VG criticality of the conductivity parallel to the ab planes should be the same. Discussing the phase angle at T_c in Sec. III, we have considered the possibilities of 3D and 2D fluctuations driving the YBCO films to the normal state.

Accordingly, the 3D scenario turned out to be acceptable if correlated pinning of vortex fluctuations by linear defects or microtwins would prevail. Such fluctuations could consist of vortex loops, the vortex and/or antivortex segments of which are localized at the pins extending in the c direction. Below T_c, both are connected by two Josephson strings, which then unbind at T_c. Of course, in the presence of externally induced vortices such loops would act as dislocations in the pinned vortex system[1] and would give rise to the melting at $T_g(B)$. With increasing B this is achieved by loops of decreasing mean lateral diameter, $\sqrt{\phi_0/B}$, which are more easily excited so that the unbinding occurs at lower temperature.

Recently, the possibility of 2D fluctuations in thin YBCO films was proposed by Matsuda *et al.*,[22] who studied the thickness dependence of T_c in zero magnetic field. They argued that vortex-antivortex strings piercing a multilayered film are favored over the small loops formed by small vortex-antivortex pairs belonging to the same CuO_2 plane. Since such string pairs are also coupled by the long-range logarithmic interaction, they can give rise to a KT-like unbinding transition. Perhaps, the nucleation of such strings in films of thickness 200 nm is facilitated by the pressure of microtwin networks in the films. This 2D-string picture is consistent with the recent detection of a highly correlated motion of flux-noise sources on the opposing sides of thin YBCO films and crystals in zero magnetic field.[25] The authors related their observation to a hopping of rigid vortices and antivortices between pinning sites. Remarkably, Lee *et al.*[25] found the flux noise to decrease somewhat weaker than $1/\omega$. Due to the correspondence between the spectral density of the flux noise and the dissipative part of $\sigma(\omega)$,[26] this result is consistent with $\sigma'(T_c)\sim\omega^{-1+1/z}$ obtained here. If such quasi-2D string-antistring excitations can be reproduced by the modified 2D-XY Hamiltonian used by Jonsson and Minnhagen,[24] the agreement between the value of the phase angle $\varphi(T_c)$ predicted by them and that observed by us would indicate the dominance of 2D fluctuations. Extending this string-antistring model to finite fields, smaller string pairs should become relevant for a depinning of the vortex system for increasing B. Then unbinding should occur at correspondingly lower temperatures, which is also in qualitative agreement with the decrease of the VG temperature.

In conclusion, we have studied the singularities of the linear dynamic conductivity of thin YBCO films at T_c and the scaling functions of $\sigma(\omega)$ on both sides of T_c. The perfect agreement between this critical behavior and that observed in high external fields implies that the same kind of fluctuations destroys the equilibrium superconductivity also in the presence of a vortex system, and that the elastic properties,[1] i.e., the vortx-vortex interactions, have no impact on the transition. The question of whether 2D or 3D vortex fluctuations are responsible for these phenomena remains to be settled.

ACKNOWLEDGMENTS

The authors are indebted to D. Fay (Hamburg) for a critical reading of the manuscript. This work has been supported by the Deutsche Forschungsgemeinschaft under Contract No. Ko 800-1/5.

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