

## Large Dynamical Fluctuations in the Microwave Conductivity of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ above $T_c$

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(Received 27 June 1996)

We report a systematic investigation of dynamical fluctuation effects in the frequency-dependent microwave conductivity (45 MHz–45 GHz) of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films for  $T \geq T_c$ . Our measurements directly yield a dynamical critical exponent  $z$  in the range 2.3–3.0, and the fluctuation lifetime  $\tau^{\text{fl}}$ , which diverges more quickly than Gaussian theory predicts as the temperature approaches  $T_c$  from above, independent of sample quality. In addition, both the temperature and the frequency dependence of the fluctuation conductivity  $\sigma^{\text{fl}}$  exhibit scaling behavior for temperatures 1–2 K above  $T_c$ , and can be collapsed onto the same universal curve. [S0031-9007(96)01664-X]

PACS numbers: 74.25.Nf, 74.40.tk

Dynamic fluctuation effects have become an intriguing subject of research in statistical and condensed matter physics, as they depend on the equations of motion of the system, and are not simply determined by the equilibrium distribution of particles at a given instant of time. Over the years, there have been both theoretical and experimental investigations of the static and dynamic fluctuations of many different types of systems. In the high  $T_c$  superconductors (HTSC), measurements of fluctuation effects reveal unusual behavior, such as 3D XY critical behavior observed in the microwave penetration depth  $\lambda(T)$  over a range of temperature 5–10 K wide below  $T_c$  [1]. Heat capacity measurements are also consistent with 3D XY critical fluctuations for  $T = T_c \pm 10$  K [2]. In contrast to these results, temperature-dependent dc conductivity measurements have been interpreted in terms of simple 2D or 3D Gaussian fluctuation theory for  $T > T_c$  [3,4]; also, low frequency penetration depth measurements have been interpreted as consistent with Gaussian fluctuations [5]. Furthermore, analysis of the nonlinear current-voltage characteristics near  $T_c$  in the HTSC yield a wide range of values for the relevant critical exponents [6,7].

In this Letter we report a systematic experimental study of the microwave fluctuation conductivity  $\sigma^{\text{fl}}$  obtained from thin films and a single crystal of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO). The study provides direct information about the fluctuation lifetime  $\tau^{\text{fl}}$ , which is one of the important parameters for the identification of the universality class of the phase transition, and which describes the relevant dynamics of the fluctuations in the HTSC. We employ a swept-frequency technique (45 MHz–45 GHz) to measure both the frequency and temperature dependence of the conductivity of YBCO thin films in the vicinity of  $T_c$  with nominally zero external magnetic field. The extra experimental degree of freedom afforded by access to the measured conductivity over three decades in frequency provides essential additional information about the dynamics of the system not available to temperature dependent measurements alone. Previous experiments have

exploited frequency dependent techniques to verify the Gaussian dynamics of fluctuations in thin films of conventional superconductors [8,9]. More recently, frequency-dependent complex resistivity measurements have been used at lower frequencies in finite magnetic fields to investigate the dynamical properties of the phase transition of the mixed state [10–13].

Sample fabrication and the measurements are carried out as described in detail elsewhere [13,14]. The thin film under study is used to terminate an 0.086 inch diameter coaxial cable in a Corbino disk geometry, and the complex resistivity  $\rho = \rho_1 + i\rho_2$  is measured over the continuous frequency range from 45 MHz to 45 GHz using a novel nonresonant technique, with care taken to account for the effects of the substrate [14]. The microwave power is carefully adjusted to be within the range of +5 to –30 dBm to minimize the rf current in the sample without losing a clear reflected signal [13]. In this work we measure several laser ablated  $t_0 = 150$  nm thick  $c$ -axis-oriented YBCO films (on  $\text{LaAlO}_3$  substrates) of varying quality, as determined by high resolution x-ray diffraction [15] and by high frequency characterization. Since we find very consistent behavior from all the samples studied, initially we focus on the results obtained from a single thin film (thin film No. 1) with  $\delta T_c \sim 0.8$  K (10–90% transition width measured at 45 MHz). The frequency-dependent complex resistivity  $\rho(\omega)$  for this film is shown in Figs. 1(a) and 1(b). Well above  $T_c$ ,  $\rho(\omega)$  is predominantly real and is nearly frequency independent. As the temperature is decreased to the immediate vicinity of the resistive transition [see the inset of Fig. 1(b)], both  $\rho_1(\omega)$  and  $\rho_2(\omega)$  are nonzero and exhibit considerable frequency dependence.

To extract the frequency-dependent fluctuation conductivity  $\sigma^{\text{fl}}(T, \omega)$  from the total measured conductivity  $\sigma^{\text{tot}}(T, \omega) = 1/\rho(T, \omega)$ , we write the conductivity as a sum of mean-field and fluctuation contributions,  $\sigma^{\text{tot}}(T, \omega) = \sigma^{\text{mf}}(T) + \sigma^{\text{fl}}(T, \omega)$ , with the assumption that  $\sigma^{\text{mf}}$  is independent of frequency:  $\sigma^{\text{mf}}(T) =$

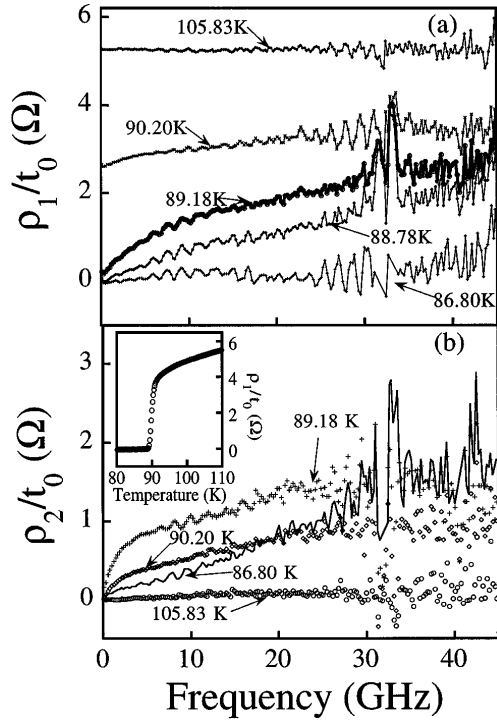


FIG. 1. The frequency dependence of (a)  $\rho_1/t_0$  and (b)  $\rho_2/t_0$  at various temperatures in the vicinity of  $T_c$ . Inset:  $\rho_1(T)$  measured at 270 MHz.

$1/\rho^{\text{mf}}(T) = 1/(r_0 + r_1 T)$ . We obtain a measure of the mean field contribution  $\sigma^{\text{mf}}(T)$  by fitting the temperature dependence of the dc resistivity, which is measured separately from 290 K down to  $T_c$ . We fit the dc data for  $T > T_c$  to the form  $\rho_{\text{dc}}(T) = 1/[\sigma^{\text{mf}}(T) + \sigma^{\text{fl}}(T, \omega = 0)]$ , using simple theoretical models for  $\sigma^{\text{fl}}(T, \omega = 0)$  in order to describe the data as close to  $T_c$  as possible. We obtain the best fit using the Lawrence-Doniach [16] model for the dc fluctuation conductivity with  $\rho^{\text{mf}}(T) = 10.49 + 0.70 T \mu\Omega \text{ cm}$  and  $T_c^G = 89.83 \text{ K}$ . (As we will discuss later, the Gaussian fluctuation models are inconsistent with our data very near  $T_c$ , so the  $T_c^G$  extracted here is not an accurate determination of  $T_c$ .) Other fits, using the Aslamasov-Larkin 2D and 3D forms for  $\sigma^{\text{fl}}(T, \omega = 0)$  [17] and a simple fit with  $\sigma^{\text{fl}} = 0$  also render very similar values for  $\sigma^{\text{mf}}(T)$ , differing from the above value at temperatures near  $T_c$  by at most 5%. The results which follow do not depend significantly on the choice of  $\sigma^{\text{mf}}(T)$  within this range.

To understand the nature of dynamic fluctuations in our YBCO samples, we examine both the frequency and the temperature dependence of  $\sigma^{\text{fl}}(T, \omega)$  in terms of a general theory of fluctuation conductivity at a second order phase transition [18,19]. The theory contains as a special case the Gaussian results, and with this theory we can identify the universality class to which the YBCO samples belong. The theory predicts that in the vicinity of a second order phase transition the fluctuation con-

ductivity should scale with the appropriate power of the (temperature-dependent) correlation length  $\xi$  and the fluctuation lifetime  $\tau^{\text{fl}}$ :  $\sigma(\omega) \approx \xi^{2-d+z} S_{\pm}(\omega \tau^{\text{fl}})$ . Near the phase transition the correlation length diverges with temperature as  $\xi = \xi_0 \varepsilon^{-\nu}$ , where  $\varepsilon = \ln(T/T_c)$ , while the fluctuation lifetime diverges as  $\tau^{\text{fl}} = \tau_0 \varepsilon^{-\nu z}$ . The phase of the fluctuation conductivity  $\phi_{\sigma} (= \tan[\sigma_2/\sigma_1])$  also scales with the argument  $\omega \tau^{\text{fl}}$  as  $\phi_{\sigma}(T, \omega) = \Phi_{\pm}(\omega \tau^{\text{fl}})$ . The functions  $S_+$  ( $S_-$ ) and  $\Phi_+$  ( $\Phi_-$ ) are universal scaling functions above (below)  $T_c$ , which should be the same for all members of a given universality class, as should be the critical exponents  $\nu$  and  $z$  (the quantities  $\tau_0$  and  $\xi_0$ , along with  $T_c$ , are by contrast nonuniversal, and differ from sample to sample). The scaling functions can be calculated in the Gaussian limit [19,20], but are in general unknown otherwise.

In the limit of  $T \rightarrow T_c$ , both  $\xi$  and  $\tau^{\text{fl}}$  diverge, but the quantities  $|\sigma^{\text{fl}}(\omega)|$  and  $\phi_{\sigma}(\omega)$  remain finite and reduce to simple forms that depend only on the dimensionality  $d$  of the system and the exponent  $z$ . In other words, the theory predicts at  $T = T_c$  a power law dependence of the magnitude of the conductivity  $|\sigma^{\text{fl}}(T_c, \omega)| = c|\omega|^{-(2-d+z)/z}$ , where  $c$  is a constant, and a constant phase angle  $\phi_{\sigma}(T_c, \omega) = (\pi/2)(2-d+z)/z$ . Examination of our experimental data [shown in the insets of Figs. 2(a) and 2(b)] shows that the conductivity magnitude is best fit to a power law at  $T = 89.18 \pm 0.1 \text{ K}$ , where  $|\sigma^{\text{fl}}(\omega)| \approx \omega^{-0.62 \pm 0.04}$ , while  $\phi_{\sigma}$  is most constant (i.e., the smallest standard deviation from a constant value) also at  $T = 89.18 \pm 0.1 \text{ K}$ , with a mean value  $\phi_{\sigma} \approx (\pi/2) \cdot (0.64 \pm 0.1)$ . Note that the conductivity phase is an increasing (decreasing) function of frequency for temperatures above (below)  $T_c$ . These two independent results allow us to directly determine the thermodynamic critical temperature to be  $T_c = 89.18 \pm 0.1 \text{ K}$  and the dynamical critical exponent (assuming  $d = 3$ ) to be  $z = 2.65 \pm 0.3$  for this sample (the accuracy of this determination of  $T_c$  and  $z$  is limited by the separation in temperature of the isotherms). This value of  $z$  is substantially smaller than that found earlier at the vortex glass-liquid transition [13], suggesting that our results reflect mainly fluctuation effects. Other samples give consistent values for  $z$ . In all cases our results for the value of the dynamical critical exponent are significantly larger than the Gaussian (or relaxational) prediction of  $z = 2$ . Previous determinations of the dynamical critical exponent  $z$  in YBCO films from the scaling of the nonlinear dc resistivity in zero magnetic field give widely varying results, from  $z = 2.2 \pm 0.4$  [6] up to  $z = 8.3 \pm 0.3$  [7].

Now we examine the scaling behavior of the data for temperatures  $T \geq T_c$ , which provides further information about the dynamics and can be used to deduce the static critical exponent  $\nu$ . From the scaling form of the conductivity given above, and assuming  $d = 3$ , we expect the conductivity scaled as  $|\sigma^{\text{fl}}| \varepsilon^{\nu(z-1)}$  plotted against the scaled frequency  $\omega/\varepsilon^{\nu z}$  to yield the scaling function  $|S_+|$ , independent of temperature. Figure 2(a)

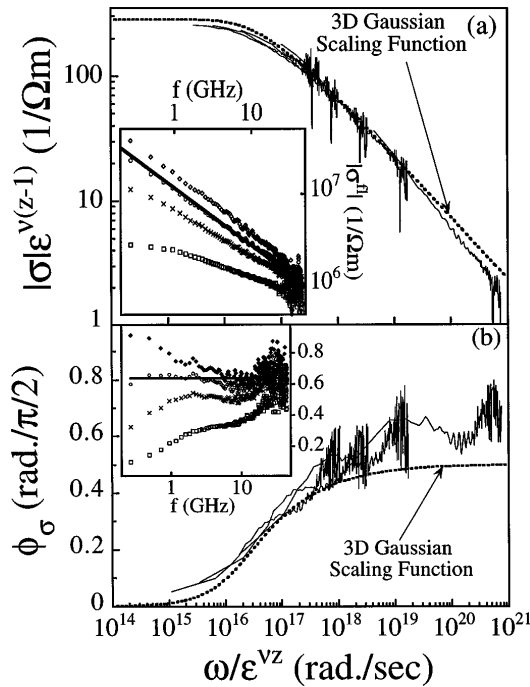


FIG. 2. Scaling behavior of the frequency-dependent fluctuation conductivity for  $T > T_c$  of (a) the magnitude and (b) the reduced phase of the fluctuation conductivity, with  $T_c^{\text{scaling}} = 89.10$  K,  $\nu = 1.2$  and  $z = 2.6$ . Each data set spans the frequency range 45 MHz–45 GHz at temperatures  $T = 89.18, 89.38, 89.58, 89.80$ , and, for (a) only, 90.00 K. The dashed lines are the 3D Gaussian scaling functions. Insets: the unscaled  $|\sigma_1^{\text{fl}}(\omega)|$  and  $\phi_\sigma(\omega)$  at 88.99 K ( $\diamond$ ), 89.18 K ( $\circ$ ), 89.38 K ( $\times$ ), and 89.80 K ( $\square$ ). The solid lines in the insets: the best power-law fit in (a) and the constant reduced phase  $\phi_\sigma(\omega) \approx 0.6$  in (b).

shows such a plot for five different temperatures within 1 K above  $T_c$ , which all collapse onto a single curve with the choice of  $T_c^{\text{scaling}} = 89.10$  K,  $z = 2.65$ , and  $\nu = 1.0$  [21]. Similarly, as shown in Fig. 2(b), the phase of the conductivity  $\phi_\sigma$  scales with the same values of  $\nu$ ,  $z$ , and  $T_c^{\text{scaling}}$ . In general, there exists a range of values for  $\nu$ ,  $z$ , and  $T_c^{\text{scaling}}$  which cause the data to collapse in this manner, but these values are constrained somewhat by the results for  $T_c$  and  $z$  derived from the critical isotherm above. Overall we find that  $\sigma_1^{\text{fl}}$  of YBCO is consistent with the scaling theory, and obtain  $T_c^{\text{scaling}} = 89.10 \pm 0.1$  K and the critical exponents in the range of  $\nu = 1.0 \pm 0.2$  and  $z = 2.65 \pm 0.3$  [21]. Note the values for  $T_c$  and  $z$  are consistent with those obtained from the frequency dependence of the unscaled quantities  $|\sigma_1^{\text{fl}}(\omega)|$  and  $\phi_\sigma(\omega)$  within our experimental error.

Since  $\tau^{\text{fl}} \sim 1/\varepsilon^{\nu z}$ , our results indicate that  $\tau^{\text{fl}}$  diverges with exponent  $\nu z = 2.35\text{--}3.55$ , which is significantly faster than the Gaussian prediction of  $\nu z = 1$ , suggesting a strong slowing down of the fluctuation relaxation rate as  $T \rightarrow T_c^+$ . The effect of the divergence

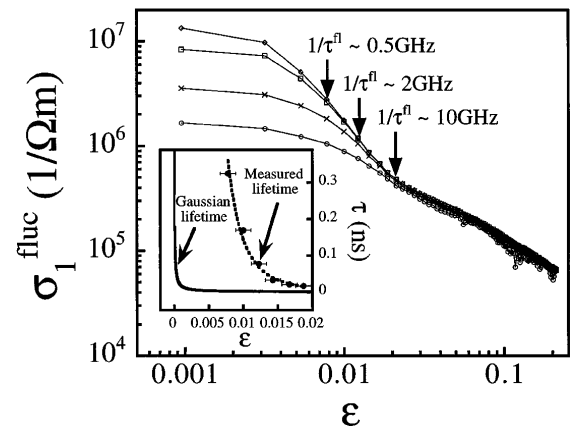


FIG. 3. The fluctuation conductivity  $\sigma_1^{\text{fl}}(\varepsilon)$  (with  $T_c^{\text{scaling}} = 89.10$  K) at 10 GHz ( $\circ$ ), 2 GHz ( $\times$ ), 0.5 GHz ( $\square$ ), and 0.27 GHz ( $\diamond$ ). The arrows indicate approximately where  $\omega\tau^{\text{fl}} \sim 1$ . Inset: the temperature dependence of the fluctuation lifetime  $\tau^{\text{fl}} \sim \varepsilon^{-3}$  (dashed line), along with  $\tau^{\text{fl}} \sim \varepsilon^{-1}$  (solid line) of the Gaussian model.

of the fluctuation lifetime can also be seen directly (without any scaling analysis) in the temperature dependence of the fluctuation conductivity measured at several fixed frequencies as shown in Fig. 3. In contrast to the behavior of  $\sigma_1^{\text{fl}}(T)$  measured at dc, which diverges as  $\varepsilon \rightarrow 0$ , it has been observed that the divergence of  $\sigma_1^{\text{fl}}(T)$  at finite frequency  $\omega$  is cut off when  $\omega\tau^{\text{fl}} \sim 1$ , [13] and  $\sigma_1^{\text{fl}}$  approaches a finite value as  $\varepsilon \rightarrow 0$ . The arrows in Fig. 3 roughly indicate the points where  $\omega\tau^{\text{fl}} \sim 1$  for measurement frequencies of 10, 2, and 0.5 GHz. The measured  $\tau^{\text{fl}}$  is plotted against  $\varepsilon$  in the inset of Fig. 3, which suggests

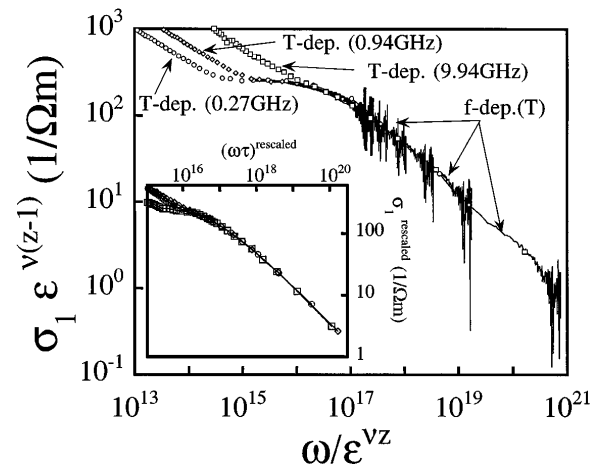


FIG. 4. The scaled  $\sigma_1^{\text{fl}}(T)$  measured at various fixed frequencies 0.27 ( $\circ$ ), 0.94 ( $\diamond$ ), 9.94 GHz ( $\square$ ), along with the scaled  $\sigma_1^{\text{fl}}(\omega)$  measured at different fixed temperatures  $T = 89.18, 89.38, 89.58, 89.80, 90.00$  K, and 90.20 K (solid lines), all with  $T_c^{\text{scaling}} = 89.10$  K,  $\nu = 1.2$ , and  $z = 2.6$ . Inset: the scaled  $\sigma_1^{\text{fl}}(T)$  for thin film No. 1 ( $\diamond$ ), thin film No. 2 ( $\circ$ ), and a single crystal ( $\square$ ), with  $\nu = 1.2$ , and  $z = 2.6$ . For the inset, the  $\sigma_1^{\text{fl}}(T)$  and  $\omega\tau$  axes were rescaled for the different samples to take into account differences in the nonuniversal quantities  $\tau_0$  and  $\xi_0$  (see Table I).

TABLE I. Values of  $T_c$ ,  $\xi_0$ , and  $\tau_0$  for the samples shown in the inset to Fig. 4. Also given is the  $c$ -axis lattice parameter in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (and % abundance) determined by high resolution x-ray diffraction, which measures the oxygen disorder for the different samples.

| Sample     | $T_c$<br>(K) | $\xi_0/\xi_0^{\text{film No. 1}}$ | $\tau_0/\tau_0^{\text{film No. 1}}$ | $c$ -axis lattice parameter <sup>a</sup><br>(nm) |
|------------|--------------|-----------------------------------|-------------------------------------|--|
| Film No. 1 | 89.18        | 1                                 | 1                                   | 1.1688 (97%), 1.1752 (3%)                        |
| Film No. 2 | 88.60        | 0.625                             | 2                                   | 1.1698 (94%), 1.1674 (6%)                        |
| Crystal    | 90.11        | 1                                 | 0.09                                | 1.1696 (70%), 1.1730 (30%)                       |

<sup>a</sup>See Ref. [15].

$\tau^{\text{fl}}$  (dashed line), consistent with the temperature dependence of  $\tau^{\text{fl}}(\sim 1/\varepsilon^{(2.35-3.55)})$  determined from the scaling of the frequency dependent data above. This is in contrast to the fluctuation lifetime in the Gaussian model ( $\tau^{\text{fl}} = \pi\hbar/16k_B T_c \varepsilon$ ) [20], which predicts  $\tau^{\text{fl}} \sim 1/\varepsilon$  (solid line in the inset of Fig. 3) and that all the  $\sigma_1^{\text{fl}}(\varepsilon)$  would remain together until  $\varepsilon \sim 1 \times 10^{-3}$  for the frequencies shown in Fig. 3.

For further examination of the scaling models, which suggests the general scaling behavior for all independent parameters, we scale both the temperature (the same data shown in Fig. 3) and the frequency dependence of  $\sigma_1^{\text{fl}}(T, \omega)$ , using the same values for the critical exponents determined earlier. In Fig. 4, we plot the scaled temperature dependence measured at three discrete frequencies along with the scaled frequency dependence of  $\sigma_1^{\text{fl}}$  measured at five different temperatures, all with the same values of  $T_c^{\text{scaling}}$ ,  $\nu$ , and  $z$ . The figure shows dramatically how both the temperature and frequency dependence of  $\sigma_1^{\text{fl}}$  are both described by the single universal scaling function  $S_+(\omega\tau^{\text{fl}})$  within the scaling regime, thus confirming the validity of the scaling model for  $\sigma^{\text{fl}}(T, \omega)$ . The width of the scaling regime is given by the point at which the temperature-dependent data deviate from the universal curve (at  $T \approx T_c + 2$  K for the sample in Fig. 4).

Because our experimental results can be influenced by the existence of material disorder and inhomogeneity, we compare the scaling behavior of  $\sigma_1^{\text{fl}}$  obtained from samples with varying amounts of oxygen disorder, as measured by high resolution x-ray diffraction [15]. In the inset to Fig. 4 we plot the scaled temperature dependence (with  $\nu = 1.2$  and  $z = 2.6$ ) of the fluctuation conductivity  $\sigma^{\text{fl}}$  for thin film No. 1, along with the scaled temperature dependence of a more disordered thin film (thin film No. 2) and also a twinned single crystal, all measured at approximately 10 GHz. We find that in order to have the data from these three very different samples collapse onto the same curve, we need only to adjust the values of nonuniversal quantities  $\tau_0$  and  $\xi_0$ , along with  $T_c$ , in the quantities  $\xi = \xi_0/\varepsilon^\nu$  and  $\tau = \tau_0/\varepsilon^{\nu z}$ . This suggests that the scaling behavior holds over the range of oxygen disorder (quantified in Table I) present in these samples (this is consistent with the Harris criterion [22] for a system with heat capacity exponent  $\alpha < 0$  [2]), and that our

experimental results reflect the intrinsic behavior of  $\sigma^{\text{fl}}$  rather than extrinsic sample properties.

In summary, we have measured the fluctuation conductivity over three decades of frequency in the vicinity of  $T_c$  for YBCO thin films. From the frequency dependence of  $\sigma^{\text{fl}}(T, \omega)$ , we have determined the thermodynamic critical temperature and the dynamical critical exponent  $z = 2.35-3.0$ , extracted  $\tau^{\text{fl}}(\varepsilon)$ , and shown that the divergence of the fluctuation lifetime  $\tau^{\text{fl}}$  is much faster than what the Gaussian theory predicts (all corroborated by scaling analysis), consistent with critical slowing down of the fluctuation relaxation rate for  $T - T_c \lesssim 1-2$  K in YBCO thin films.

This work was supported by NSF NYI DMR-9258183 and NSF DMR-9624021.

- [1] S. Kamal *et al.*, Phys. Rev. Lett. **73**, 1845 (1994).
- [2] N. Overend *et al.*, Phys. Rev. Lett. **72**, 3238 (1994).
- [3] S. J. Hagen *et al.*, Phys. Rev. B **38**, 7137 (1988).
- [4] T. A. Friedmann *et al.*, Phys. Rev. B **39**, 4258 (1989).
- [5] Z.-H. Lin *et al.*, Europhys. Lett. **32**, 573 (1995).
- [6] C. Dekker *et al.*, Physica (Amsterdam) **185-189C**, 1799 (1991).
- [7] J. M. Roberts *et al.*, Phys. Rev. B **51**, 15281 (1995).
- [8] S. L. Lehoczky and C. V. Briscoe, Phys. Rev. Lett. **24**, 880 (1970).
- [9] D. B. Tanner, Phys. Rev. B **8**, 5045 (1973).
- [10] H. K. Olsson *et al.*, Phys. Rev. Lett. **66**, 2661 (1991).
- [11] H. Wu *et al.*, Phys. Rev. Lett. **71**, 2462 (1993).
- [12] N.-C. Yeh *et al.*, Phys. Rev. Lett. **71**, 4043 (1993).
- [13] D. H. Wu *et al.*, Phys. Rev. Lett. **75**, 525 (1995).
- [14] J. C. Booth *et al.*, Rev. Sci. Instrum. **65**, 2082 (1994); J. C. Booth, Ph.D. thesis, University of Maryland, 1996.
- [15] E. F. Skelton *et al.*, SPIE Proc. **1516**, 160 (1995).
- [16] W. E. Lawrence and S. Doniach, LT-12 Proc., 361 (1970).
- [17] L. G. Aslamazov and A. I. Larkin, Fiz. Tverd. Tela **10**, 1104 (1968) [Sov. Phys. Solid State **10**, 875 (1968)].
- [18] D. S. Fisher *et al.*, Phys. Rev. B **43**, 130 (1991); P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).
- [19] A. T. Dorsey, Phys. Rev. B **43**, 7575 (1991).
- [20] H. Schmidt, Z. Phys. **216**, 336 (1968).
- [21] The large value of  $\nu$  may be due to our ignorance of the correction terms to scaling of the conductivity: see G. Ahlers, in *Quantum Liquids*, edited by J. Ruvalds and T. Regge (North-Holland, Amsterdam, 1978), p. 1.
- [22] A. B. Harris, J. Phys. C **7**, 1671 (1974).