Method to extract the critical current density and the flux-creep exponent in high-T_c thin films using ac susceptibility measurements

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High-precision ac susceptibility measurements have been made on high-quality Hg-1212 thin films. A method to analyze $\chi'_1(T,H_0,f)$ and $\chi''_1(T,H_0,f)$ and extract the temperature dependence of the critical current density $J_c(T)$, as well as the temperature and field-dependent flux-creep exponent $n(T,H_0)$, is presented. With specific measurements at external ac fields H_0 in the range $7-100\ \mathrm{Oe_{rms}}$ we determine the temperature dependence of the critical current density from a *single* temperature scan. The obtained temperature dependence, $J_c(T)$, is found to be in good agreement with data obtained from measurements using the traditional ''loss-maximum'' approach. In addition we present a method to extract the temperature and ac field-dependent flux-creep exponent $n(T,H_0)$ from a set of temperature scans taken at different ac fields and driving frequencies. The observed power law describing the frequency dependence of χ' is consistent with a current-dependent effective activation energy of the form $U(J)=U_0\ln(J_c/J)$. Furthermore, the flux creep is found to increase with ac field and with temperature except at about 20–30 K below T_c, where our data suggest a slowing down of the flux creep. [S0163-1829(98)02633-2]

I. INTRODUCTION

The vortex dynamics of high-T_c superconductors (HTSC's) is a complex problem due to the extreme richness of the magnetic phase diagram.[1,2] Interesting functional properties arise due to the various comparable energy scales in these materials. The short coherence length ξ reduces the pinning energy U_{pin} and the high critical temperature T_c leads to a high thermal energy U_{th}. A direct consequence of U_{pin} and U_{th} having similar orders of magnitude is the ''giant flux creep''[3] observed in HTSC's. The penetration length λ, which can be taken as a measure of the range of vortex-vortex interactions, is larger than for conventional type-II low-T_c superconductors (LTSC's). As a consequence vortex-vortex interactions U_{int} are enhanced and the so-called collective flux-creep phenomenon[4] (CFC) is observed in a wider field and temperature range compared to LTSC's. The study of flux creep in HTSC's materials is of considerable importance both from a fundamental as well as applications point of view.

Flux creep manifests itself experimentally in many ways and there are a number of techniques to probe the vortex dynamics over a wide range of temperature, magnetic field, and current density. Three of the most common techniques are magnetic relaxation, ac susceptibility, and I-V characteristics. The interpretations of results obtained from the different techniques have matured and can now be readily compared. ac susceptibility remains a very popular choice due to its experimental simplicity: one traditionally determines the location of the loss maximum T_p and studies its dependence on driving frequency and dc field. It has been pointed out[5] that this procedure does not make use of the full available information, i.e., from an entire temperature or field scan only a single point (the loss maximum location T_p) is extracted. Using square-wave integration,[6] instead of the ordinary sine-wave integration, and defining a so-called wide-band ac susceptibility[7] it has been demonstrated that a single temperature scan is sufficient to extract the temperature dependence of the critical current density in bulk samples. A similar procedure using the imaginary part of the ordinary ac susceptibility has also been suggested[8] but to our knowledge this approach has not been exploited.

We show how recently derived expressions for $\chi'(h_0/J_c)$ and $\chi''(h_0/J_c)$ for a thin disk perpendicular to the applied field[9] can be used to extract, not only $J_c(T)$ but also the temperature and ac field-dependent flux-creep exponent $n(T,H_0)$. We test our approach on a high-quality Hg-1212 thin film and compare it with the traditional loss maximum method. A good agreement is found between the two methods and the efficiency of our method is clearly demonstrated. In the Hg-based HTSC's we find that the flux-creep effects increase (i.e., n decreases) both with temperature and ac field. Our data also indicate a slowing down of the flux creep at about 20–30 K below T_c, which might suggest a crossover to another collective flux-creep regime, possibly involving vortex bundles.

II. COMPLEX ac SUSCEPTIBILITY

The interpretations of ac susceptibility data have matured considerably in the last years as analytical expressions have been derived for the actual sample and field geometries used in experiments. The first geometries to be considered were the infinite cylinder[10] and the infinite slab[11] in the parallel applied field where demagnetizing effects can be neglected. However, for a realistic sample with finite dimensions proper demagnetizing effects must be taken into account. This is even more so in the case of thin films in a field normal to the film plan where the demagnetizing effects dominate and the expressions derived for the parallel geometry fail to describe the response. The problem of applying the critical-state model[12] (CS) to the thin circular disk geometry was solved[13,14] in 1993 and subsequently the analytical expressions for the ac susceptibility of thin disks in perpendicular field were derived.[9] The critical state has also been analyzed for thin long strips[15] and squares.[16] It is found that due to the very large demagnetizing effects the particular in-plane shape almost does not influence the ac response and in particular the ac susceptibility differs by less than 0.2% between a square and a circular disk over the whole range of applied fields.[17] Unless there is a particular interest in the local character of the flux dynamics it is hence a very good approximation to describe the global response from a rectangular shaped film with the expressions for a circular disk. This has recently been verified by experiments on Y-Ba-Cu-O thin films of different lateral shape.[18] A recent numerical study extends the analytical expressions for the ac susceptibility of thin films in an ideal CS to thin films exhibiting flux creep.[19]

Within the framework of the CS model, the ac susceptibility of a thin circular disk with radius R and thickness d in a perpendicular time-varying field $H_a(t)=h_0\cos\omega t$ has the limiting large-field behavior[9]

$$\chi' \approx -\chi_0(1.330h^{-3/2}-0.634h^{-5/2}), \quad h \gg 1, \qquad (1a)$$

$$\chi'' \approx \chi_0(h^{-1}-1.059h^{-2}), \quad h \gg 1, \qquad (1b)$$

where h is the so-called reduced field $h=2h_0/J_c d$ and $\chi_0=8R/3\pi d$ is the value for complete shielding. In this work we make explicit use of Eqs. (1) in the limit where a single term is sufficient to describe the response,

$$\chi' \approx -\chi_0 c_1 h^{-3/2}, \quad h \gg 1, \qquad (2a)$$

$$\chi'' \approx \chi_0 c_2 h^{-1}, \quad h \gg 1, \qquad (2b)$$

and show the convenience of such simple relations when flux creep is studied. It should be noted, however, that Eqs. (2) not only apply to a thin circular disk but to any geometry, provided the applied ac field is large enough for the hysteresis loop to be approximated by a parallelogram.[20]

III. CRITICAL CURRENT FROM ac SUSCEPTIBILITY DATA

For many applications the single most important property of a superconductor is its ability to carry large currents without dissipation. The commonly used[21,22] ac susceptibility technique for measuring the critical current density makes use of the peak criterion for the imaginary part, which for a bulk sample in the parallel geometry has the general form

$$J_c(T_p)=\alpha h_0/R, \qquad (3)$$

where $\alpha \approx 1$ is a geometry-dependent factor (e.g., 3/4 for infinite slab, 1 for infinite cylinder). A similar relation[9] holds for a thin disk in a perpendicular field although due to the strong demagnetizing effects, not the radius of the sample, but instead its thickness d enters the expression

$$J_c(T_p)=1.03h_0/d. \qquad (4)$$

This relation was recently used[23] for extracting the temperature dependence of the critical current density in Y-Ba-Cu-O thin films.

Another, much less used approach makes use of the *real* part χ' of the ac susceptibility.[24] In the CS model χ' is a single-valued function of the so-called Bean penetration length $L_{CS}=h_0/J_c(T)$. It is then possible to reconstruct the temperature dependence of $J_c(T)$ by scaling data obtained for different h_0.

A more direct approach with simpler functional dependence on L_{CS} is obtained by defining a so-called wide-band susceptibility[7] from the value of the sample magnetization when the external ac field reaches its maximum (χ_a) and is zero (χ_r), respectively. In the linear regime the wide-band susceptibility coincides with the usual complex ac susceptibility but they differ when higher harmonics are present in the pick-up signal. Due to the simpler functional dependence of $\chi_a(h_0/J_c)$ and $\chi_r(h_0/J_c)$, data from the whole temperature scan can be directly used[5] to extract $J_c(T)$ instead of only retaining a single point (from the loss maximum at T_p) per scan as in the traditional approaches using Eqs. (3) and (4).

We show in this work that it is not necessary to change the traditional definition of the complex ac susceptibility and consequently have to change the experimental detection scheme.[6] At high enough ac field amplitudes (in the following denoted by the root-mean-square value $H_0=h_0/2^{1/2}$), it is instead possible to use the limiting expressions Eqs. (2) for χ' and χ'' to extract the temperature dependence of the critical current density in superconducting thin films. No scaling procedure is needed and the full temperature dependence of $J_c(T)$ is obtained from a single temperature scan.

IV. FLUX CREEP FROM ac SUSCEPTIBILITY DATA

It is experimentally known[25] that the complex ac susceptibility of high-T_c materials can be strongly frequency dependent due to certain time scales in the vortex response. Experimentally this means that the loss maximum position T_p will not only depend on H_0 through Eq. (3) but also on the frequency. The HTSC's response to an external ac field can be described by characteristics of one of the following three different regimes:[26] (i) the CS regime with no detectable frequency dependence but large ac field dependence ($U_{pin} \gg U_{th}$), (ii) the linear Ohmic regime with eddy current frequency dependence but no ac field dependence ($U_{pin} \ll U_{th}$), and (iii) the intermediate flux-creep regime with both frequency and ac field-dependent ac susceptibility of varying importance ($U_{pin} > \approx U_{th}$). The first two regimes are relatively well understood and can, in principle, be regarded as limiting cases of the third more complex intermediate regime. A common phenomenological way to describe the electrodynamics in (iii) is to assume a nonlinear current-voltage relation (nonlinear resistivity), which in the logarithmic approximation described below takes the form[4]

$$\rho=\mathbf{E}/\mathbf{J}=(E_c/J_c)|J/J_c|^{n-1}. \qquad (5)$$

Here n is the so-called flux-creep exponent that determines the degree of nonlinearity, or equivalently the importance of flux-creep effects. Small n means large creep effects and the limiting cases (i) and (ii) can be described as $n\rightarrow\infty$ and $n=1$, respectively. For a device exploiting the large critical current density of superconducting materials it is essential that n be as large as possible and in the following we will limit ourselves to the regime with $n>1$ and therefore do not further discuss the linear Ohmic regime.

The frequency-dependent loss maximum can be analyzed if one defines a frequency-dependent critical current

$$J_c(T,H_0,f)=J_c(T,H_0)g[kT\ln(f_0/f)/U(T,H_0)] \quad (6)$$

and applies it to the critical-state formalism.[3,24] Here, the function $0<g(y)<1$ describes the effective reduction of the critical current density during one period of the applied ac field. The functional form of $g(y)$ depends on the detailed nature of the thermally activated flux motion and in particular in what way the activation energy U depends on the momentary current density. In the so-called logarithmic or Zeldov approximation,[27] U depends on current as $U(J)=U_0^*\ln(J_c/J)$. Within the collective flux-creep theory[28] (CFC) such a dependence will turn $g(y)$ into an exponential

$$g(y)=\exp(-y). \quad (7)$$

Substitution of Eq. (7) in Eq. (6) then implies that the critical current density should depend on frequency following a power law

$$J_c(T,H_0,f)=J_c(T,H_0)(f/f_0)^{1/n}, \quad (8)$$

where $n=U(T,H_0)/kT$ is the same flux-creep exponent as in Eq. (5). It is thus possible to determine $n(T,H_0)$ by measuring $J_c(T,H_0,f)$ at different ac fields and frequencies, in a single temperature scan run.

In this work we demonstrate how to extract the flux-creep exponent $n(T,H_0)$ for superconducting thin films from a small set of temperature scans of χ' taken at different ac fields and frequencies. In a typical temperature scan for the determination of $n(T,H_0)$ a single ac field is used and 4–5 different frequencies. The analysis is particularly straightforward due to the simple relation [Eq. (2a)] between J_c and χ'. There are in the literature examples of using the frequency dependence of χ' to extract n for single-crystal samples.[24] However in order to get J_c from χ' those studies use the rather tedious scaling procedure mentioned above, which limits its usefulness. We believe that our simpler approach represents an efficient way to extract the maximum information of the field- and temperature-dependent parameters that govern the vortex response in high-T_c materials.

V. EXPERIMENTAL DETAILS

A conventional two-step method was used for the sample preparation: deposition of Hg-free precursor films on $SrTiO_3$ substrates followed by annealing at 820 °C for 30 min in a controlled Hg-vapor atmosphere. Samples were rectangular in shape with typical area of 2×4 mm^2. Film thickness was estimated to 4000 Å. The x-ray-diffraction pattern collected within $5°<2\theta<70°$ using a Siemens D5000 Diffractometer shows mainly lines belonging to the c-axis-oriented Hg-1212 phase with minute traces of c-axis Hg-1223. Rocking curves on the (006) line indicate good orientation with full width at half maximum of around 0.3°. ac susceptibility measurements were carried out in a home-built high sensitivity susceptometer with a three-coil mutual inductance bridge and a two-position background subtraction scheme. Sine-wave integration on the fundamental frequency was used, i.e., the susceptometer measures $\chi_1'V$ and $\chi_1''V$ (in units of emu/Oe), which in the following will be denoted by χ' and χ'' for brevity. Root-mean-square ac fields ranged from $H_0=0.1$ mOe to 100 Oe and frequencies from 1.81 to 891 Hz. No dc field was applied and the Earth's field was not shielded. The films were always measured perpendicular to the applied ac field.

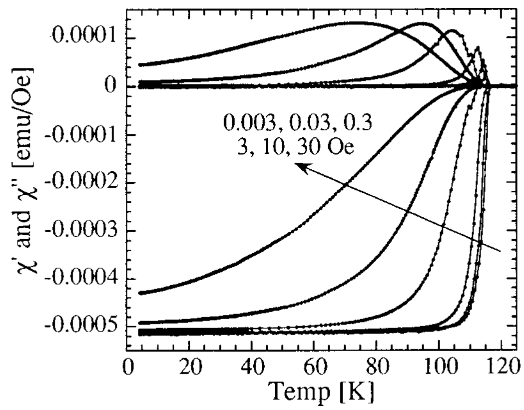

FIG. 1. χ' and χ'' for six different ac fields in the range $H_0=3$ mOe–30 Oe.

VI. RESULTS AND DISCUSSION

A. ac field amplitude dependence—determination of $J_c(T)$

The temperature dependence of the components of the ac susceptibility $\chi'-i\chi''$ of a rectangular (2×4 mm^2) Hg-1212 thin film is shown in Fig. 1 for increasing applied ac fields H_0. At all fields, a single transition is observed as is expected for a high-quality thin film. The transition is broadened as H_0 increases and the loss maximum is suppressed to lower temperatures. This general behavior is typically expected within a Bean critical-state model. However, we also observe a weak H_0 dependence of the maximum value of χ'' (in particular for $H_0<3$ Oe) which is not consistent with a CS description. In this work we do not further analyze this discrepancy and argue that in order to interpret the experimental results in a CS context it is useful to work at values of $H_0>3$ Oe where a true critical state seems to be established. In the following we always keep $H_0>7$ Oe.

A central issue for the following flux-creep analysis is the confirmation of the simple relation between J_c and χ [Eqs. (2)]. We measured χ' and χ'' vs T at different ac fields $H_0=7-100$ Oe and studied the ac field dependence of data taken at same temperatures. For $H_0<35$ Oe we indeed found a power-law dependence of χ' and χ'' on H_0 with exponents -1.5 and -1.0, respectively. The validity of these exponents can be seen by plotting $\chi' H_0^{3/2}$ [Figs. 2(a) and 2(b)] and $\chi'' H_0$ [Figs. 3(a) and 3(b)] vs T, which confirms that over a large region the different curves overlap. Each set of data

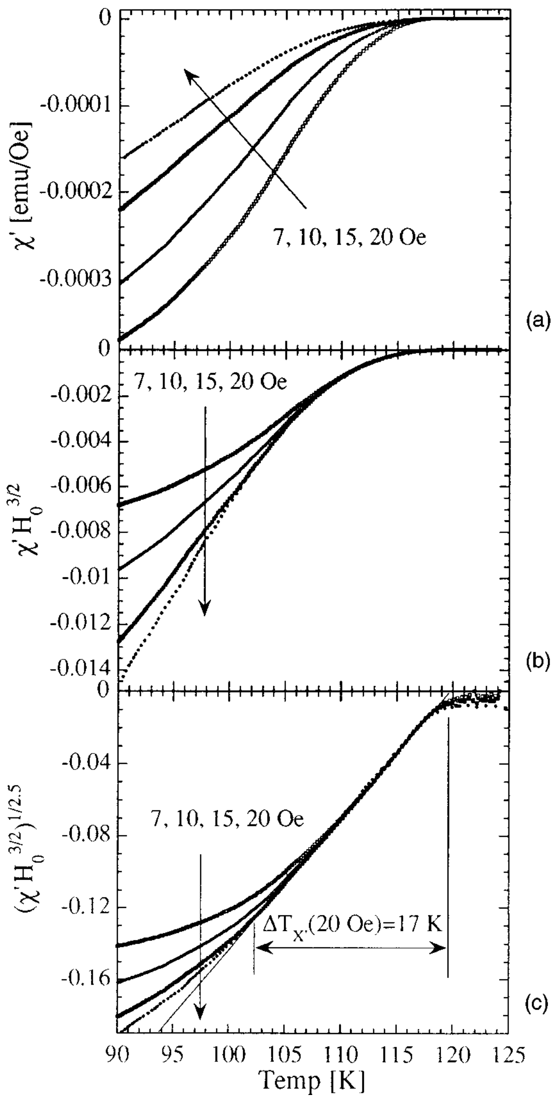

FIG. 2. (a) Initial part of χ' for $H_0 = 7$, 10, 15, and 20 Oe. (b) Plotting $\chi' H_0^{3/2}$ vs T makes all data fall onto a single curve in a certain temperature range $\Delta T_{\chi'}(H_0)$ that increases with H_0. (c) Plotting $(\chi' H_0^{3/2})^{1/2.5}$ vs T yields a straight line.

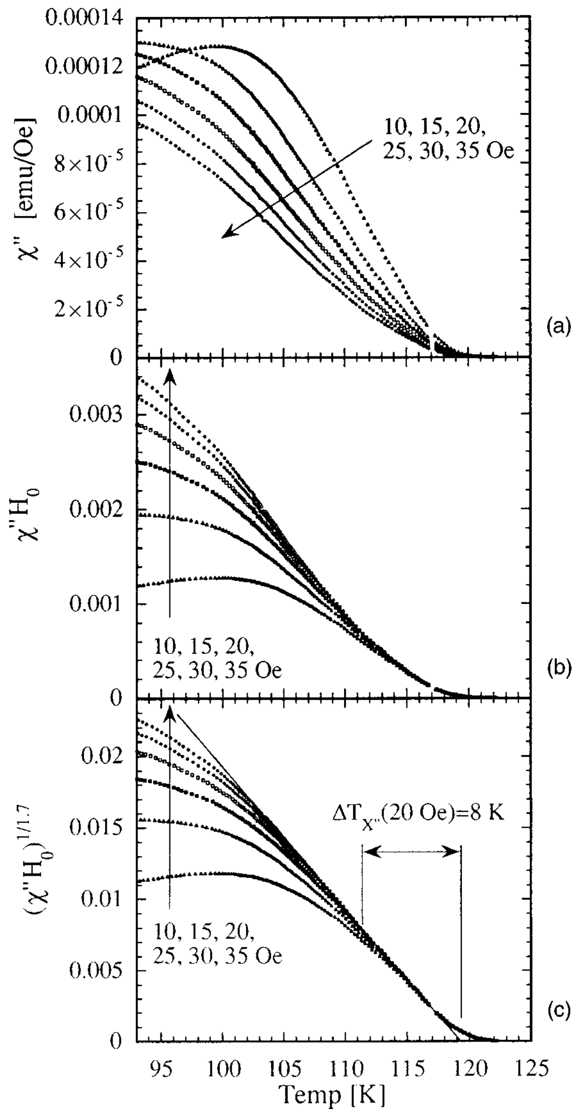

FIG. 3. (a) Initial part of χ'' for $H_0 = 10$, 15, 20, 25, 30, and 35 Oe. (b) All data points fall onto a single curve when plotting $\chi'' H_0$ vs T. (c) A straight line is obtained when plotting $(\chi'' H_0)^{1/1.7}$ vs T.

points starts to deviate at an H_0-dependent temperature as the second term in Eqs. (1) becomes comparable to the first. At ac fields $H_0 > 35$ Oe we observed a slight increase of both exponents with H_0. We attribute these stronger field dependences as coming from a field dependence of the critical current density. Below we will see that this is indeed consistent with a field-dependent flux-creep exponent, $n(T,H_0)$.

From the confirmation of Eq. (2a) we conclude that, in a certain temperature region, a plot of $J_c(T) = 2^{3/2} H_0 (-\chi'(T)/c_1\chi_0)^{2/3}/d$ vs T will directly give the temperature dependence of the critical current density. Assuming a temperature dependence of the form

$$J_c(T) = J_{c0}(1 - T/T_c)^{\beta}, \tag{9}$$

we expect a straight line if we plot $J_c(T)^{1/\beta}$ vs T. In practice this means raising χ' to some power value $1/\nu$ and accepting the value that yields the best straight line [Fig. 2(c)]. We find $\nu = 2.5$ and hence get $\beta = 1.7$ from Eq. (2a). Note that $T_c = 119.5$ K is conveniently found from the intersection of the straight line with the temperature axis. According to Eq. (2b) it should be equally valid to extract $J_c(T)$ from a single temperature scan $\chi''(T)$ by finding the exponent $1/\nu$ that transforms $\chi''^{(1/\nu)}$ into a linear fit. This is indeed demonstrated by using the same exponent $\beta = 1.7$ and noting that over a substantial temperature region all the curves fall onto the same straight line [Fig. 3(c)]. A closer comparison between the two methods of extracting $J_c(T)$ reveals that using χ' is the better choice since the temperature region ΔT where the data fall on the straight line, i.e., where a single term in Eqs. (1) ($h^{-3/2}$ or h^{-1}) dominates the response, is wider for χ' than for χ''. At $H_0 = 20$ Oe this range is $\Delta T_{\chi'} = 17$ K for χ' and only $\Delta T_{\chi''} = 8$ K for χ''. For the analysis in the following section it is important that the data be obtained in the region $\Delta T_{\chi'}$ where we thus can rewrite Eq. (2a) as

$$\chi'(T,H_0,f) = \text{const} J_c(T,H_0,f)^{3/2}, \tag{10}$$

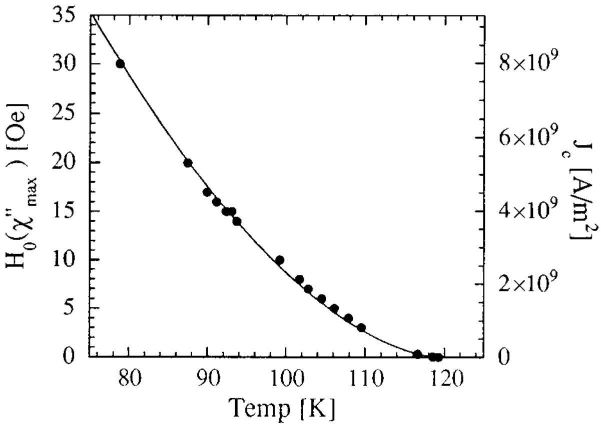

FIG. 4. Temperature dependence of J_c obtained using the traditional loss-maximum method. The line is a power-law fit with exponent $\beta=1.7$.

and not further consider the prefactor const$=-\chi_0 c_1(d/2H_0)^{3/2}$.

In order to check the validity of this procedure we carried out a traditional loss maximum determination of the critical current density vs T and found that a power law with $\beta=1.7$ represents a good fit to the data (Fig. 4). We have thus shown that a single temperature scan of χ' at sufficiently high ac field is enough to determine $J_c(T)$. To get the absolute value of $J_c(77\text{ K})$ we use Eq. (4) (with $h_0=2^{1/2}H_0$), approximating our sample with a disk, and find $J_c(77\text{ K})=8.7\times10^9$ A/m^2. Extrapolation to lower temperatures using Eq. (9) yields $J_c(4.2\text{ K})=4.8\times10^{10}$ A/m^2.

B. Frequency dependence—determination of the flux-creep exponent

In Fig. 5 is shown how χ' increases with frequency, which indicates the presence of a certain time scale for the vortex motion. We make isothermal cuts in Fig. 5 and plot $\chi'(T,H_0,f)$ vs f in a log-log diagram (Fig. 6). By combining Eqs. (8) and (10) we get a power-law expression for $\chi'(T,H_0,f)$,

$$\chi'(T,H_0,f)=P(T,H_0)f^{m(T,H_0)}, \tag{11}$$

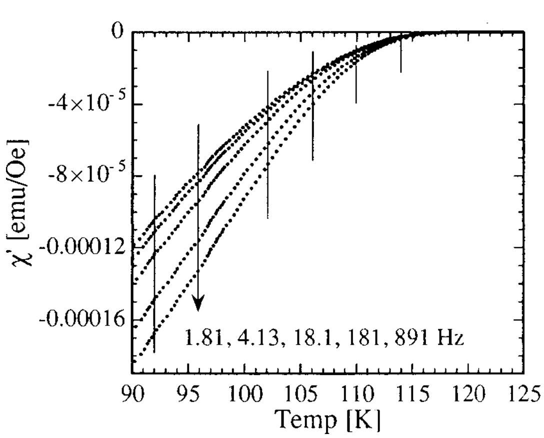

FIG. 5. Initial part of χ' for $H_0=20$ Oe and $f=1.81$, 4.13, 18.1, 181, 891 Hz. The straight lines indicate the isothermal cuts plotted in Fig. 6(b) below.

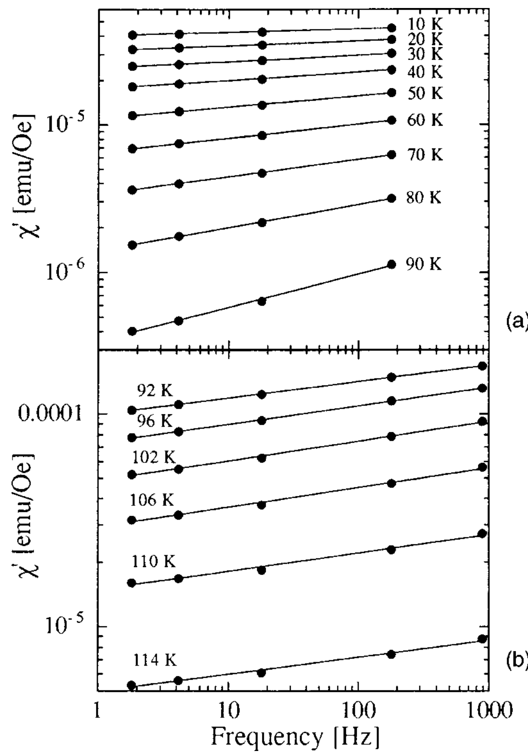

FIG. 6. (a) log-log plot of χ' vs f at $H_0=100$ Oe. Lines are power-law fits $\chi'(T,H_0,f)=P(T,H_0)f^{m(T,H_0)}$. (b) same analysis for $H_0=20$ Oe with data from the isothermal cuts in Fig. 5.

which we fit to the data. From these fits we extract the exponent $m(T,H_0)$ and then get, via Eq. (10), the flux-creep exponent at all temperatures and different ac fields as $n=3/(2m)$. It is seen in Fig. 7 that $m(T,H_0)$ increases $[n(T,H_0)$ decreases$]$ both with temperature and with the applied ac field. The increase of flux creep with temperature is expected for thermally activated vortex motion. The increase of flux creep with magnetic field indicates a possible corresponding decrease of the activation energy barrier $U(T,H_0)$. There is also a clear maximum in $m(T,H_0)$ [minimum in $n(T,H_0)$] at about 20–30 K below T_c and the relaxation seems to slow down again. It might be that the logarithmic approximation is no longer valid in this region and that a new creep regime is entered. Such a slowing down may be

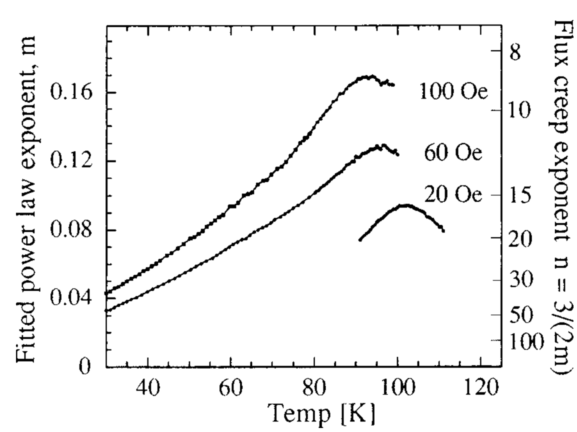

FIG. 7. The fitted parameter $m(T,H_0)$ as a function of temperature at different ac fields $H_0=20$, 60, and 100 Oe. On the second scale is shown the flux-creep exponent $n=(3/2)/m$.

attributed to flux creep of vortex bundles[29] instead of single-vortex creep.

The strong dependence of n and m on H_0 qualitatively explains the slight deviation from the limiting expressions [Eq. (2)] in Sec. VI at the highest ac fields $H_0 > 35$ Oe. At these ac field values $\chi'(T,H_0,f)$ goes as $H_0^{-\gamma}$, $\gamma = 1.5-2$ and not exactly as $H_0^{-3/2}$ since the reduction with field of $J_c(T,H_0,f)$ will contribute. From the analysis above we conclude that this reduction of $J_c(T,H_0,f)$ with field can be related to the increase in flux creep for large H_0.

VII. SUMMARY AND CONCLUSION

We present an ac susceptibility method for the study of the temperature-dependent critical current density and the temperature and field-dependent flux-creep exponent. The main advantage vis-a-vis traditional methods is the amount of information that can be extracted from a small number of temperature scans. We show that the results on the critical current density agree with results obtained using the traditional ''loss-maximum'' approach. The critical current density of a high-quality Hg-1212 thin film is well described by $J_c(T) = J_{c0}(1 - T/T_c)^{\beta}$ with $\beta = 1.7$ in the range 77–120 K. At 77 K we find $J_c(77\,\mathrm{K}) = 8.7 \times 10^9$ A/m^2 and an extrapolation to lower temperatures yields $J_c(4.2\,\mathrm{K}) = 4.8 \times 10^{10}$ A/m^2. Furthermore we show how to extract the temperature and field-dependent flux-creep exponent $n(T,H_0)$ from the frequency dependence of the real part, χ'. Flux creep is found to increase both with temperature and with the ac field. However, at about 20–30 K below T_c a slowing down of the flux creep is observed, which might indicate a crossover to a regime of flux bundle creep.

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