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ac susceptibility and critical-current densities in sintered $YBa_2Cu_3O_{7-\delta}$ superconductors

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The field-amplitude and frequency dependent complex ac susceptibility of a well sintered $YBa_2Cu_3O_{7-\delta}$ disk has been measured at 77 K, which shows that its intra- and intergranular critical-current densities are determined by the collective flux creep inside the grains and the maximum Josephson currents across the grain boundaries, respectively. While the former is widely recognized, the latter contrasts to a popular belief of a mechanism of Josephson-vortex pinning and creep. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2336596]

It was found in the early study of sintered $YBa_2Cu_3O_{7-\delta}$ (YBCO) high-temperature superconductors (HTSs) that the temperature dependence of their low-field ac susceptibility $\chi=\chi'-j\chi''$ showed a two-stage behavior, corresponding to two contributions from the superconducting grains and the intergranular matrix.[1–3] The intergranular χ was first simulated by the critical-state model with a Kim-type internal-field dependent critical-current density $J_c(H_i)$ (Kim model[4]) after introducing an effective grain volume fraction f_{gr} to separate it from the total χ.[5,6] A similar two-stage behavior was later more carefully studied in the field-amplitude dependence, and it was found that an exponential $J_c(H_i)$ was a better choice than the Kim type for fitting the low-field intergranular data by optimizing two parameters.[7,8] On the other hand, the high-field intragranular χ could not be simulated by the critical-state model alone, but instead a successful data fit required this model to be combined with certain field-dependent surface currents to account for the contributions from the thermal equilibrium magnetization and surface barrier effects of type-II superconductors.[8,9]

When Kim *et al.* reported their $J_c(H_i)$ for type-II superconductors, its mechanism was explained by Anderson in terms of the pinning and supercurrent-driven thermally activated creep of Abrikosov vortices (AVs), assuming a fixed pinning barrier U_0 for an AV bundle, and so the activation energy $U(J)=U_0(1-|J/J_c|)$.[10] Intensive research has been carried out concerning AVs and their movements after the discovery of HTSs, and an important step was the proposition of the collective flux-creep theory for HTSs.[11] In this theory, the activation volume is not an AV bundle but increases with decreasing supercurrent density J owing to the presence of randomly distributed weak pinning centers, which leads to $U(J)=U_0 \ln|J_c/J|$.[11] This second theory has been well accepted to explain the J_c in HTS single crystals or grains. Unlike the case of grains, the situation for the intergranular J_c is controversial. A dominant view is that sintered HTSs may be modeled by a Josephson-junction (JJ) array, whose intergranular J_c arises from the pinning and creep of Josephson vortices (JVs) and follows the Kim model.[12,13] However, direct calculations of magnetic properties of resistively shunted JJ arrays show that JVs may be induced by a changing field only when the critical JJ currents are very small. Instead, if the critical JJ currents are large, a state will be induced with most JJs carrying currents close to their critical currents, i.e., the intergranular J_c comes directly from the maximum JJ currents themselves.[14]

In order to experimentally justify the above statements, we have measured the field-amplitude (H_m) and frequency (f) dependent χ of several sintered YBCO disks at 77 K. We have shown that when grain links are bad, intergranular $\chi(H_m,f)$ is consistent with a JV-creep model, but cannot be derived from the critical-state model with any type of $J_c(H_i)$.[15] In this letter, we will show that when grain links are good, low-f $\chi(H_m)$ can be derived from a certain $J_c(H_i)$ and its f dependence has a flux-flow type, directly arising from the modeled resistively shunted JJs.

An YBCO disk of diameter 12.45 mm, thickness 3.75 mm, and density 3.92 g/cm^3 was sintered at 960 °C for 12 h followed by furnace cooling. Its solid volume was mainly occupied by grains of sizes ~20 μm. Its axial χ was measured at 77 K using a well calibrated ac susceptometer.[16,17] The measured zero-field cooled χ' and χ'' as functions of μ_0H_m at $f=30$, 90, 270, 810, and 2430 Hz are plotted in Fig. 1. We observe in this figure the two-stage feature mentioned above; with increasing H_m, the first and second χ' rises and χ'' peaks correspond to the inter- and intragranular field penetrations, respectively. There is a characteristic f dependence of $\chi'(H_m)$ for its first rise; it shifts to higher H_m with a rate $d\log H_m(\chi')/d\log f$ increasing with increasing f when $\mu_0H_m>0.2$ mT, below which (and/or at low f) $\chi'(H_m)$ is f independent. Plotting Fig. 1 in a finer μ_0H_m scale (not shown), we observe that the second-stage $\chi'(H_m)$ shifts to higher H_m with increasing f from 30 to 810 Hz at a constant rate $d\log H_m(\chi')/d\log f=0.017$.

Compared with previous works, we have extended χ measurements of sintered YBCO samples to higher H_m and f in the present work. The f dependence is specially important for seeking the mechanism of J_c, since, while the ideal critical-state model leads to a f-independent $\chi(H_m)$, any actual mechanism of J_c will correspond to a characteristic $E(J)$

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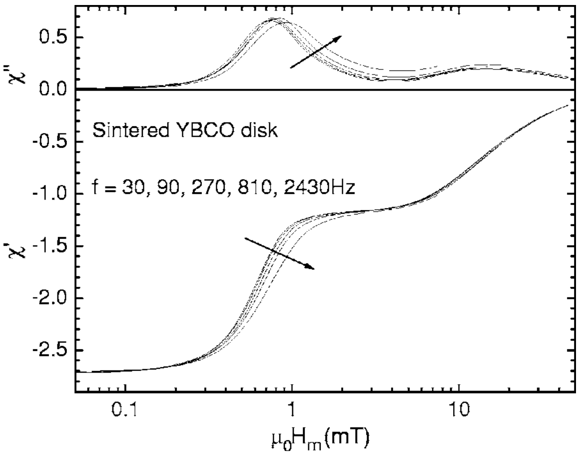

FIG. 1. χ' and χ'' of the studied YBCO disk measured at 77 K as functions of H_m and f. The arrows indicate the direction of increasing f.

function and so result in a characteristic $\chi(H_m,f)$ dependence. We have calculated, using a numerical technique proposed by Brandt,[18,19] $\chi(H_m,f)$ for an infinitely long superconducting cylinder of radius a from various $E(J)$ characteristics for the above mentioned flux-creep models and a simplified flux-flow model. We have used a power-law $E(J)=\mathrm{sgn}(J)E_c|J/J_c|^n$ and an exponential $E(J)=\mathrm{sgn}(J)E_c\exp[n(|J/J_c|-1)]$, where $n=U_0/(kT)=10$ and J_c is defined as the J when $E=E_c=10^{-4}$ V/m, for the collective and Kim-Anderson types of flux creep, respectively,[18,19] and nonzero $E(J)=[J-\mathrm{sgn}(J)J_c]\rho_f$ occurring at $|J|>J_c$, where $\rho_f=10^{-7}$ Ωm is the flux-flow resistivity, for the flux-flow case.[20] The results are plotted in Figs. 2(a)–2(c) for the collective creep, Kim-Anderson creep, and flux flow, respectively. We should note that the high-n limit of both creep models and the low-f limit of the flux-flow model are identical to the critical-state model with a constant J_c (Bean model).

We see in Fig. 2(a) that $H_m(\chi')$ increases with increasing f at a constant $d\log H_m(\chi')/d\log f$, which is consistent with the observed intragranular behavior of our YBCO disk, so that intragranular J_c is governed by the collective flux creep as expected. A similar behavior has been found for a single crystal YBCO film with $n=35$.[21] There are two differences between the polycrystalline bulk (disk) and the single crystal film; $n=1+d\log f/d\log H_m(\chi')=60$ for the bulk is much higher than 35 for the film and the maximum χ'', χ''_m, for the film is consistent with the prediction of the Bean model, but it is too low for the bulk. Although further studies are necessary for fully understanding this, one thing is clear that the magnetization is dominated by volume critical currents for the film, but it is also contributed by surface currents for the grains of the bulk.[8,9]

Comparing Figs. 2(b) and 2(c) with Fig. 1, we see that the intergranular $\chi'(H_m,f)$ of the studied disk has features very similar to those for the flux-flow model, but different from the Kim-Anderson flux-creep model. Since the Kim-Anderson model is the basis of the JV-creep mechanism of intergranular J_c proposed in Refs. 12 and 13 this should rule out this mechanism for our sample. On the contrary, the validity of the flux-flow model indicates that the intergranular J_c arises directly from the Josephson currents across weak-linked grains, since it is basically consistent with the behavior of resistively shunted JJs.[22]

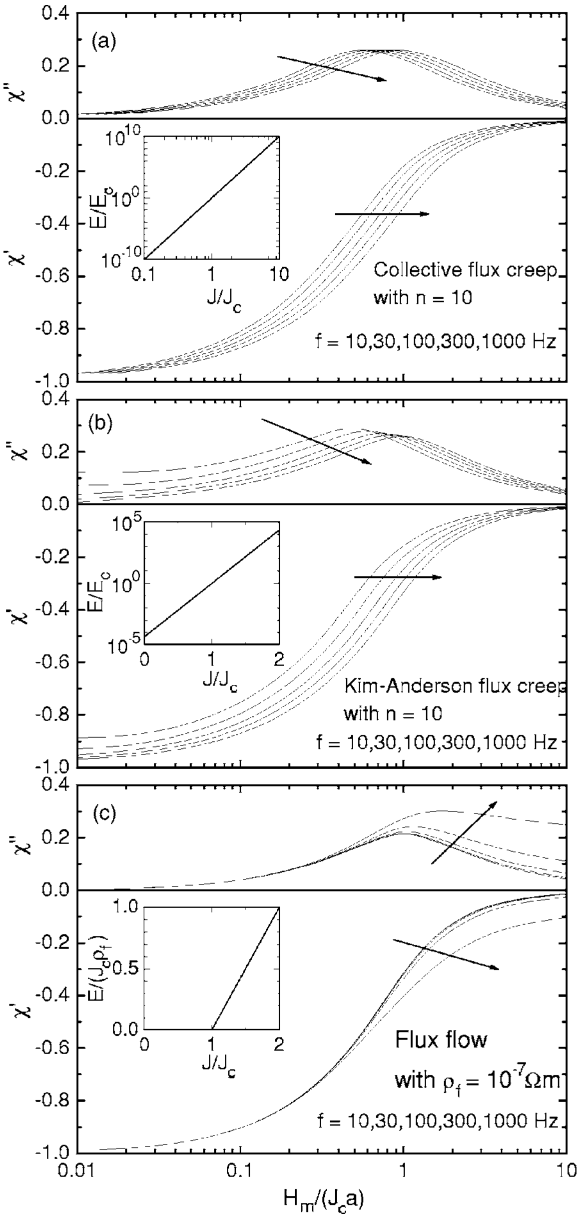

FIG. 2. χ' and χ'' of a long superconducting cylinder with radius a and critical-current density J_c as functions of $H_m/(J_c a)$ and f calculated for (a) collective flux-creep, (b) Kim-Anderson flux-creep, and (c) flux-flow models. Insets show the $E(J)$ characteristics and arrows indicate the direction of increasing f.

The low sensitivity of intergranular $\chi(H_m)$ to f at low f not only shows its flux-flow feature, but also indicates the validity of the critical-state model to it. It will be interesting to compare the experimental data with existing results calculated from the critical-state model. For this, we define the low-H_m limit of $-\chi'$ as $\chi_0(=2.71)$ and obtain $f_{\mathrm{gr}}=0.435$, following the procedure described in Ref. 7 so that the partial χ_0 for the grains and the intergranular matrix are $\chi_{0,\mathrm{gr}}=\chi_0 f_{\mathrm{gr}}=1.18$ and $\chi_{0,\mathrm{mtx}}=\chi_0(1-f_{\mathrm{gr}})=1.53$, respectively. The partial intergranular χ is thus $\chi_{\mathrm{mtx}}=\chi+\chi_{0,\mathrm{gr}}$ for the matrix in the first stage. The $\chi''_{\mathrm{mtx}}/\chi_{0,\mathrm{mtx}}$ vs $\chi'_{\mathrm{mtx}}/\chi_{0,\mathrm{mtx}}$ function is plotted in Fig. 3 for two cases of $f=30$ and 270 Hz. We see that both curves are similar with the maximum $\chi''_{\mathrm{mtx}}/\chi_{0,\mathrm{mtx}}\approx 0.44$ occurring at $\chi'_{\mathrm{mtx}}/\chi_{0,\mathrm{mtx}}\approx -0.26$, which can be compared with 0.19 and −0.35, recently calculated from the Bean model for finite cylinders.[23] The difference should be due to the field dependence of J_c in the matrix.

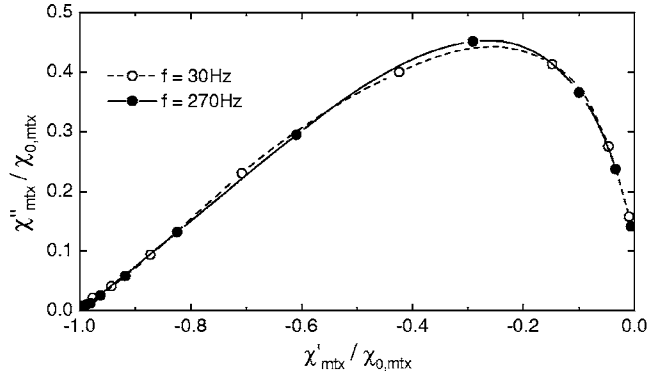

FIG. 3. Normalized partial $\chi''(\chi')$ curves for the intergranular matrix at $f=30$ and 270 Hz.

Investigating the critical-state $\chi(H_m)$ calculated with different types of $J_c(H_i)$,[24] we find that our situation is consistent with an exponential $J_c(H_i)$ with $J_c(H_i=0)$ to be about 10 times $J_c(H_i=H_p)$, where H_p is the full penetration field, equal to about $1.6H_m(\chi''_m)$ for the disk.[23] Such a $J_c(H_i)$ is consistent with the diffraction patterns of the intergranular JJs. There will be about $5\Phi_0$ flux in an effective JJ area of 10^{-11} m^2 (for grain size of 20 μm) at $\mu_0 H_p=1$ mT, and if considering the flux expelled from the weak-linked shielded grains to the JJ region, $5\Phi_0$ may be increased to $10\Phi_0$ so that $J_c(0)\approx 10J_c(H_p)$ can be reached.

We should mention that there is a difference in $\chi''(H_m,f)$ between the first stage in Fig. 1 and the flux-flow model in Fig. 2(c); χ'_m increases with f for the latter, but it is somewhat insensitive to f for the former. This may be understood as follows. With a constant ρ_f, the high-f limit of χ'_m for the calculated long cylinder should be the normal eddy-current $\chi''\approx 0.377$, which is greater than the low-f Bean limit $\chi'_m=0.212$, so that χ'_m increases with increasing f. For our disk, however, the low-f maximum $\chi'_{\rm mtx}/\chi_{0,\rm mtx}\approx 0.44$ is greater than the eddy-current value of ~ 0.35 as calculated for the eddy-current case in Refs. 17 and 23 so that χ'_m should decrease with increasing f after a possible initial rise as shown in Fig. 1. A similar phenomenon has been reported for a weak-linked Bi-2223/Ag sample.[20]

In conclusion, we have measured the field-amplitude H_m and frequency f dependent ac susceptibility $\chi=\chi'-j\chi''$ of a well sintered YBCO disk up to high H_m and f values. Compared with the $\chi(H_m,f)$ calculated from the flux-creep model of collective or Kim-Anderson type and a flux-flow model, the results show that the intragranular J_c is determined by the collective flux creep, whereas the intergranular J_c follows a different mechanism, coming from the maximum Josephson currents of weak links themselves. Although the theoretical justification of such a mechanism for the intergranular J_c based on the dc and ac Josephson equations with the gauge-invariant phase difference requires lengthy calculations and arguments,[25] its practical impact is straightforward; the only way to increase weak-link intergranular J_c is to improve the superconducting connections between grains. When the weak links are improved to strong links by grain alignments in Bi-2223/Ag tapes, melt-textured YBCO superconductors, or YBCO coated conductors, the intergranular J_c is significantly increased and the depinning of AVs or Abrikosov-Josephson vortices becomes again the governing dissipation mechanism. The presence of Josephson vortices is harmful, as already demonstrated in Ref. 15. In this work, we have conceptually or qualitatively questioned the JV-creep mechanism of intergranular J_c; a recent quantitative investigation on several HTS samples using formulas for the JV-creep model proposed in Ref. 13 has led to a huge unphysical temperature-dependent correction to the sample thickness.[26]

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