Magnetic-field dependence of the ac susceptibility in granular $YBa_2Cu_3O_7$: Data and models

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For a sintered rod of $YBa_2Cu_3O_7$ at 78.3 K, detailed data are reported for the complicated dependence of the ac susceptibility $\tilde{\chi}=\chi'-i\chi''$ on applied ac and dc fields, ascribed to the inter- and intragranular components of the material. The intergranular data are understood in detail by a modified critical-state model, assuming a critical current density $J_c(H)\sim H^{-2}$. $\tilde{\chi}(H_{dc})$ also displays hysteresis which is not readily explicable by this model.

Owing to its granular nature the superconductor $YBa_2Cu_3O_7$ displays an ac susceptibility $\tilde{\chi}=\chi'-i\chi''$ with elaborate dependence on temperature T and ac and dc applied fields H_a.[1] Bulk specimens are generally assumed to consist of superconducting grains connected by weak links, referred to as intra- and intergranular components, respectively; both contribute to $\tilde{\chi}$ at high and low fields, respectively. A semiquantitative interpretation of the data, particularly the temperature dependence, has been obtained by calculations[2,3] based on critical-state models.[4–6] The key to a more detailed understanding of $\tilde{\chi}(H_a)$ in the low-field region, our concern in this report, is the dependence of the critical current density J_c on field. We present detailed data as well as a modified critical-state model, finding very good agreement.[7,8] The model also predicts well the observed harmonic generation.[9]

Experimental results. The data were obtained for a cylinder (radius $R=0.85$ mm, length $=10.5$ mm) fabricated from sintered polycrystalline $YBa_2Cu_3O_7$.[10] This was encased in a thin-wall quartz tube wound with a 380-turn coil and connected to a digital impedance analyzer (HP 4192A), which provided a controlled ac current and thus an ac field $H_1\cos(2\pi ft)$ on the sample. The analyzer also measured the effective inductance L and resistance R of the coil. The sample was also subject to a quasi-dc field H_{dc} from a coaxial copper solenoid driven by a symmetric-triangle ramp current at $f_{scan}\approx 0.0025$ Hz from a synthesizer (HP 3325A). All instruments and data acquisition were computer controlled. For a given set of controlled parameters (T,H_{dc},H_1,f), three were held constant and the other one was varied over its range, both with and without the sample in the coil; the data sets were subtracted to give ΔL and ΔR, from which χ' and χ'' were computed, including small corrections for demagnetization factor and coil resonance.[8] All the data shown, except for Fig. 2(h), were taken with the same sample at $T=78.3$ K and $f=10$ kHz. Figure 1(a) shows the dispersive component of the susceptibility, χ' vs H_{dc}, for selected values of H_1: 0.03–7.91 Oe; these data were taken by cooling the sample in zero field and slowly ramping H_{dc} from 0 to 40 Oe, down to 0, down to -40 Oe, etc. Our model predictions, in Fig. 1(b), are seen to be in unusually good and detailed agreement with the data. Similarly, the data, Fig. 1(f), for the absorption component, χ'' vs H_{dc}, are well predicted by the model, Fig. 1(g), except for a small hysteresis between increasing and decreasing fields scans. For frequencies in the range $10^3<f<10^6$ Hz both χ' and χ'' show a very small dependence ($<1\%$) on f, implying that the loss mechanism is hysteresis, predicted by the critical-state model. Data for $\chi'(H_1)$ are shown in Fig. 2(a) for selected values of H_{dc} from 0 to 15 Oe; these are also well predicted by the model, Fig. 2(b). Data for $\chi''(H_1)$ are presented in Fig. 2(c), with the model results in Fig. 2(d). The hysteresis found in $\tilde{\chi}(H_{dc})$ is shown more clearly in Fig. 2(e) for successive scans 1,2,3, . . . , for $0\le H_{dc}\le 100$ Oe; a larger hysteresis is found in scans to 800 Oe, shown in Figs. 2(f) and 2(g).

Modified critical-state model. To understand the above rather complicated data we start with a two-dimensional model of Clem[11] for a long cylindrical sample of radius R containing long, superconducting "grains" of radius R_g and penetration depth λ_g, the remaining volume being assumed to be the so-called intergranular region of weak links, e.g., Josephson-junction barriers, with penetration depth λ_J; we assume $R\gg R_g\gg\lambda_g$ and $\lambda_J\gg\lambda_g$, and an applied field $H_a(t,r=R)=H_{dc}+H_1\cos(2\pi ft)$ parallel to the cylinder axis. After zero field cooling let the applied field be increased to some value $H_a>H_{c_{1J}}$, the lower critical field for the intergranular region; flux lines of B, in units of fluxons $hc/2e$, enter through the cylinder walls giving rise to self-induced circular shielding currents of density J, which pull the fluxons inward with a Lorentz force density[12] $F_L=|\mathbf{J}\times\mathbf{B}|/c\approx-(dH/dr)B/4\pi$, where $H(r)$ is the local magnetic field in the cylinder created by the shielding current $J(r)$; F_L is essentially the gradient of the magnetic pressure. The fluxons quickly move in until $|F_L|\le\alpha_c$ at all points in the sample, where α_c is the fluxon pinning force density arising from lattice defects, impurities, etc., and is sample dependent. This state is called the critical state[4–6] in which there is no net force on the fluxons and $J\rightarrow J_c$, the critical value of the current density, experimentally known to depend on the local field H. Except for slow thermally activated flux creep the critical state is a (barely) stable state and will be assumed to be maintained for sufficiently small and slow changes in H_a. If H_a is now reduced, fluxons will be forced outward through the cylinder walls by a reversed shielding current density, leaving, however, some fluxons trapped in the sample. The above process is concisely stated by the critical-state equation of Bean:[4]

$$dH/dr = (\pm)4\pi J_c(H)/c \tag{1}$$

for the local field H, where $(\pm)$ is determined by the sign of the electromotive force. The equation predicts the magnetization loop $M(H)$ vs H, with hysteresis, as observed in type-II superconductors. Equation (1), with a suitable equation for $J_c(H)$, allows a calculation of the local field $H(r)$; and with $H_a(t)$, the time-dependent local field $H(r,t)$ in the sample, and its spatial average value $\overline{H}(t)$.[2,7–9,13] From $\overline{H}(t)$ we calculate the real and imaginary parts of $\tilde{\chi}$ (Gaussian units) defined by

$$1+4\pi\chi' = \frac{\omega}{\pi H_1}\mu_{\rm eff}\int_0^{2\pi/\omega}\overline{H}(t)\cos(\omega t)dt \ , \tag{2}$$

$$4\pi\chi'' = \frac{\omega}{\pi H_1}\mu_{\rm eff}\int_0^{2\pi/\omega}\overline{H}(t)\sin(\omega t)dt \ , \tag{3}$$

where $\mu_{\rm eff}$ is a geometrical filling factor. These equations are used, with suitable parameters, to separately compute the contributions of the inter- and intragranular components to the total $\tilde{\chi}=\tilde{\chi}_J+\tilde{\chi}_g$. Here we report model predictions only for $\tilde{\chi}_J$, observed in the low-field region, $H_a \lesssim 50$ Oe.

The key to a good model of $\tilde{\chi}(H_{\rm dc},H_1)$ is a realistic expression for $J_c(H)$ based on experimental data rather than simple assumptions. In our modified critical-state model for the intergranular region we use

$$J_c(H) = \alpha' c/(|H|+H_0)^\beta \tag{4}$$

where α' is a fluxon pinning parameter, assumed field independent, and H_0 and β are parameters that determine the form of $J_c(H)$ and are to be determined by a fit to our data. The parameter α' is related to the fluxon pinning force density $\alpha_c = \alpha'/(|H|+H_0)^{\beta-1}$. Bean's simplified model assumed $\beta=0$; Anderson and Kim assumed $\beta=1$, as did Müller[2] and Ishida and Goldfarb[3] in models and measurements of $\tilde{\chi}$ in $YBa_2Cu_3O_7$, finding good agreement for the temperature dependence.

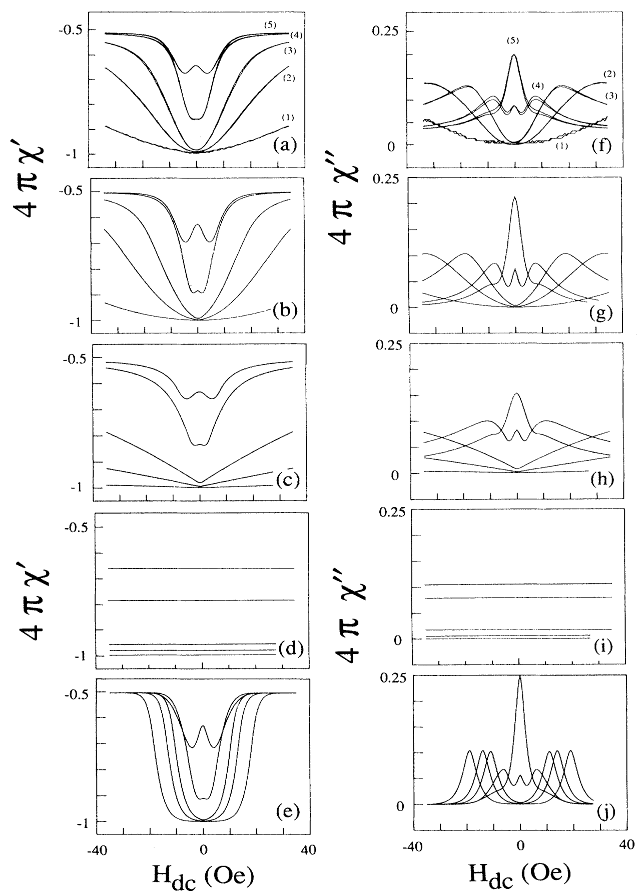

FIG. 1. (a) Measured ac susceptibility $\chi'(H_{\rm dc})$ (Gaussian units) for a $YBa_2Cu_3O_7$ sample at $T=78.3$ K and $f=10$ kHz for values of ac field H_1 (Oe): (1) 0.03, (2) 0.22, (3) 0.71, (4) 4.03, (5) 7.91. (b) Modified critical-state model prediction for the data of (a) using parameters $\beta=2.25$ and $H_0=3.0$ Oe in Eq. (4). (c) Same as (b) but with $\beta=1.0$. (d) Same as (b) but with $\beta=0$. (e) Critical-state model prediction for exponential dependence of $J_c(H)$. (f)–(j) same as (a)–(e), but $\chi''(H_{\rm dc})$ plotted.

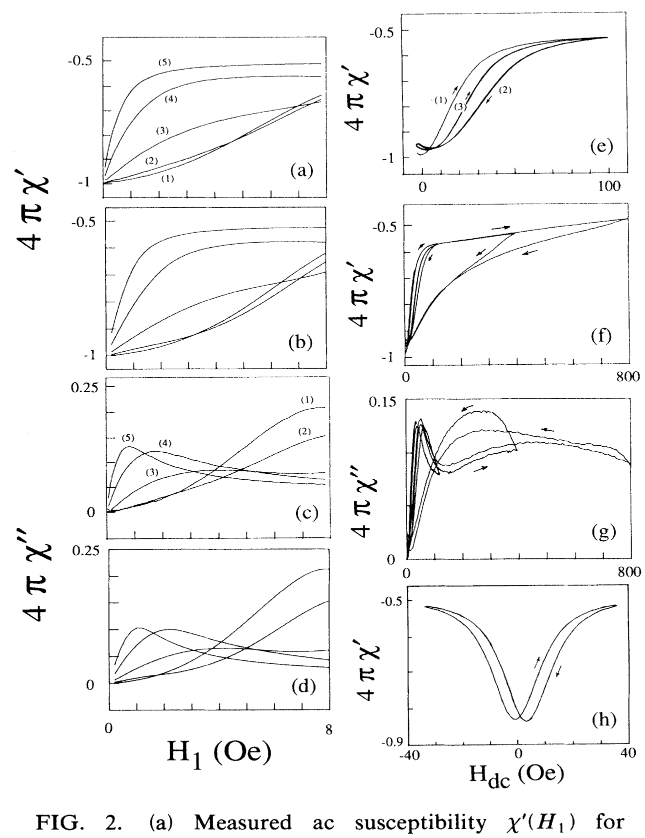

FIG. 2. (a) Measured ac susceptibility $\chi'(H_1)$ for a $YBa_2Cu_3O_7$ sample at $T=78.3$ K and $f=10$ KHz for values of dc field $H_{\rm dc}$ (Oe): (1) 0, (2) 2, (3) 5, (4) 10, (5) 15. (b) Modified critical-state model prediction for the data of (a) using parameters $\beta=2.25$ and $H_0=3.0$ Oe in Eq. (4). (c),(d) Same as (a),(b) but $\chi''(H_1)$ plotted. (e)–(h) measured hysteresis of ac susceptibility $\tilde{\chi}(H_{\rm dc})$ at $H_1=0.22$ Oe, $T=78.3$ K.

We now discuss transport measurements of $J_c(H)$ for $YBa_2Cu_3O_7$ bars at $T=77$ K, particularly those of Ekin *et al.*,[14] Peterson and Ekin,[15,16] and Male *et al.*[17] Although different samples have different maximum J_c, the dependence of $J_c(H)$ is somewhat invariant and may be described as follows in terms of three regions: (i) $J_c \approx 300$ A/cm^2 remains constant for $0.03 \lesssim H \lesssim 3$ Oe and then falls as $J_c \propto H^{-\gamma}$ with $1.5 \lesssim \gamma \lesssim 2$ to a (ii) plateau at $J_c \approx 10$ A/cm^2 in the region $100 \lesssim H \lesssim 3000$ Oe, where it again (iii) falls rapidly to $J_c \approx 0.1$ A/cm^2 as $H \rightarrow 10^5$ Oe. These regions are identified[15] as follows. Region (i): Current limited by Josephson weak links or junctions with field dependence best described by an Airy diffraction pattern, averaged over a distribution of junction lengths and orientations[16]; this model is in good agreement with their transport $J_c(H)$ data and predicts a flat region at very low fields followed by a falloff, $J_c \propto (H/H_0)^{-3/2}$, where $H_0=[(hc/2e)/\text{grain area}] \approx 3-5$ Oe. The data of Male *et al.*[17] show similar behavior with $J_c \propto H^{-2}$ in the same region. Equation (4) can be seen to be a simple algebraic approximation for region (i). Region (ii): Current limited by remanent percolation paths in a small volume fraction of the sample. Region (iii): Flux flow region as $H \rightarrow H_{c_2}$ for the weakest link. Except for Figs. 2(f) and 2(g), our data are taken in region (i), hence the form of Eq. (4), with the expectation that $\beta \approx 2$.

Model parameters. At some constant temperature the model [Eqs. (1)–(4)] predicts χ' and χ'' as functions of the experimentally known parameters (H_{dc}, H_1, R) and those characterizing sample properties (α', H_0, β, and μ_{eff}). However, there is an additional relationship $\alpha'=[(H^*+H_0)^{\beta+1}-H_0^{1+\beta}]/[4\pi(\beta+1)R]$, where H^* is the value of H_a for which the flux front just reaches $r=0$, corresponding to a maximum of $\chi''(H_1)$ at $H_1=H^*$, readily measured[8] to be ≈ 8.0 Oe. The saturated value $4\pi\chi' \rightarrow \mu_{eff}-1$ for $H_a \gtrsim 20$ Oe, yielding $\mu_{eff}=0.49$. Thus there are only two parameters, β and H_0, to be fit by the experimental data $\tilde{\chi}(H_{dc}, H_1)$; from these one can find α' from the relationship $H^*=[4\pi\alpha'(1+\beta)R+H_0^{1+\beta}]^{1/1+\beta}-H_0$, derived by the model. The best fit to the $\tilde{\chi}$ data of Figs. 1(a) and 1(f) was found for the parameters $\beta=2.25$ and $H_0=3.0$ Oe, yielding the modified critical-state predictions shown in Figs. 1(b) and 1(g), which closely agree with data both as a function of H_{dc} and of H_1. Those parameters correspond to a pinning parameter $\alpha'=688$ Oe$^{3.25}$/cm and a pinning force density $\alpha_c=174$ Oe^2/cm. Similar data at $T=82.5$ K also showed good agreement with the model using $\beta=2.22$ and $H_0=2.5$ Oe. Data at $T=85.7$ K are well explained by the model with $\beta=2.20$ and $H_0=2.0$ Oe, except for the hysteresis.

Figures 1(c) and 1(h) show the predictions if β is changed to 1.0, and are seen to fail to fit at small H_1. If we use $\beta=0$, the predictions as seen in Figs. 1(d) and 1(i) do not agree at all, since this assumes no field dependence of J_c. For an assumed exponential field dependence,[18] $J_c(H) \propto \exp(-|H|/H_e)$, the best fit, obtained for $H_e \approx 2.5$ Oe, still fails noticeably at large H, as shown in Figs. 1(e) and 1(j). We conclude that detailed $\tilde{\chi}(H_{dc}, H_1)$ data are very sensitive to $J_c(H)$ and can in fact be used to determine $J_c(H)$.

Hysteresis of $\tilde{\chi}(H_{dc})$. The small hysteresis in Fig. 1(f) is revealed more fully in Fig. 2(h), in sample No. 2, from a different batch of $YBa_2Cu_3O_7$. The central feature is that $\chi'(H_{dc})$ for symmetric scanning between $\pm H_{dc}|_{max}$ shows two minima near $H_{dc}=0$, shifted to a higher (lower) field for field decreasing (increasing). Both χ' and χ'' have the property $\chi(H_{dc})|_{s=1}=\chi(-H_{dc})|_{s=-1}$, with $s=\text{sgn}(dH/dt)$. If $|H_{max}| \neq |H_{min}|$, this symmetry breaks down. The splitting ΔH between the minima is a measure of the hysteresis, and increases with increasing temperature and decreasing H_1 and is somewhat sample dependent. This behavior of $\tilde{\chi}$ has not been previously reported, to our knowledge, but appears to be related both to the coherent-detected second-harmonic voltage and to modulated microwave power absorption in $YBa_2Cu_3O_7$.[19] Figures 2(f) and 2(g) with separate scans up to $H_{max}=40$, 100, 400, and 800 Oe show a clear distinction between the intergranular contribution to $\tilde{\chi}$ for $H_{dc} \lesssim 50$ Oe [with small hysteresis, weak pinning, saturation of $\chi'(H_{dc})$, and a peak in $\chi''(H_{dc})$ at ≈ 40 Oe] and the intragranular contribution for $H_{dc} \gtrsim 80$ Oe (with very large hysteresis, strong pinning, minimal structure of χ', and a very broad peak in χ'' at ≈ 400 Oe). If the sample is powdered, the features below $H_{dc} \approx 50$ Oe disappear. We point out that this hysteresis in $\tilde{\chi}$ cannot be formally predicted by the above critical-state models for either component, owing to the assumption that the critical current density J_c depends only on the magnitude of the local field H, according to Eq. (4). Most transport measurements report $J_c(H)$ measured after zero field cooling and then monotonic increase in H. However, Male *et al.*[17] report J_c for a well-characterized $YBa_2Cu_3O_7$ bar at 77 K for a perpendicular applied field, first increasing to a field H_{max} and then decreasing to zero (for $H_{max}=100$, 300, and 500 Oe), showing a hysteresis in $J_c(H_{dc})$ quite similar to $\chi'(H_{dc})$ in Fig. 2(f). The fact that J_c depends not only on H but also the magnetic history is a likely source of the hysteresis in Fig. 2(f), with strong intragranular pinning, as well as in Fig. 2(h), with much weaker intergranular pinning. Evetts and Glowacki[20] have proposed a mechanism for hysteresis in $J_c(H_{dc})$.

To summarize, for the $YBa_2Cu_3O_7$ at 78.3 K we report detailed data on $\tilde{\chi}$ as a function of small applied dc and ac fields, finding close agreement with a modified critical-state model assuming a critical current density $J_c(H) \sim H^{-2}$, a steeper field dependence than the original Bean, Anderson, Kim model, but in reasonable agreement with transport measurements. We find that $\tilde{\chi}(H_{dc})$ shows a small hysteresis for the intergranular component and much larger hysteresis for the intragranular component, neither being formally explicable by existing critical-state models, requiring further investigation.

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