PHYSICAL REVIEW B VOLUME 50, NUMBER 21 1 DECEMBER 1994-I

Multiple peaks in the ac susceptibility of untwinned Y-Ba-Cu-O single crystals: A manifestation of the peak effect

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(Received 13 June 1994)

We have measured the ac susceptibility of several high quality twinned and detwinned $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystals. We see multiple peaks in the $\chi''(T)$ curves, and explain them as a manifestation of the peak effect in $J_c(T)$ near T_c and increased J_c at low temperatures. We have investigated the effect of electron irradiation on these peaks, and found that it increases J_c of the sample. We suggest that the narrow, high-temperature (just below T_c) peak in $\chi''(T)$ is an indication that the sample is clean, and therefore exhibits a first-order melting transition of the flux-line lattice. We also propose that the peak effect in J_c is a result of the softening of the lattice before it melts, in accordance with Pippard's model of this phenomenon for low-temperature superconductors. Finally, we suggest a method of determining the true irreversibility line.

INTRODUCTION

The measurement of ac susceptibility has been one of the most popular techniques of investigating flux dynamics in low- and high-temperature superconductors. In such experiments, one superimposes a small ac field on a large dc field, and measures the real and imaginary parts of the ac response. One usually finds a steplike feature in χ' accompanied by a peak in χ''. The interpretation of this peak has been a controversial subject. For example, while it has been interpreted as a phase transition by some,[1] others believe it is a skin-depth-matching effect.[2]

Observations of more than one peak in $\chi''(T)$ have also been reported. Krusin-Elbaum *et al.*[3] reported a double peak in $\chi''(T)$ in single crystals of $YBa_2Cu_3O_{7-\delta}$ (YBCO). This double peak appeared over a finite range of dc magnetic fields and intermediate field orientations. The multiple peaks were attributed to two components of J_c, arising from a "staircase vortex" pattern having portions of the flux line lying between Cu-O layers. Yazyi *et al.*[4] studied the vortex dynamics of $Bi_2Sr_2CaCu_2O_8$ crystals by ac susceptibility and high-Q mechanical oscillator techniques. They reported seeing two peaks, and associated them with two transitions in the vortex response. The one at the lower temperatures was attributed to currents flowing across the Cu-O planes, and the other to currents in the plane. In both of these studies,[3,4] a single peak in $\chi''(T)$ was observed for the case $\mathbf{H}_{dc}\|\mathbf{h}_{ac}\|c$ axis, in contrast to our results.

Recently, D'Anna, Andre, and Benoit[5] also reported seeing multiple peaks in the transverse ac losses of YBCO single crystals. They measured the losses by using a low-frequency pendulum oscillator with $\mathbf{H}_{dc}\perp\mathbf{h}_{ac}$. They suggested the peak effect as the explanation of the peak structures.

In this paper we report low-frequency ac susceptibility measurements of twinned and detwinned YBCO crystals with $\mathbf{H}_{dc}\|\mathbf{h}_{ac}\|c$ axis. We observe multiple peaks in the ac losses, and provide an explanation for them. We examine the effect of disorder on these peaks by introducing point defects; we do this by using electron irradiation to displace Cu atoms from the CuO_2 planes.[6,7] (It is already known that our unirradiated crystals have extraordinarily low flux pinning.[8]) Finally, we show how to determine the true irreversibility line.

EXPERIMENTAL METHODS

High quality single crystals of $YBa_2Cu_3O_{7-\delta}$ were grown in yttria-stabilized zirconia crucibles by using the self-flux growth technique. The method of sample preparation (including growth and oxygenation) is described in detail elsewhere.[9] The samples used in this study were cleaved to an approximately rectangular shape, and some were detwinned by applying uniaxial pressure in the direction of their short edge.[10] Each sample was then reannealed to relieve possible strains introduced by that procedure. The transition temperatures of the annealed crystals (twinned and detwinned) were in the range of 90–90.5 K, with a transition width of less than 1 K for an applied field of 1 Oe. Since the results were similar for all five of the samples we investigated (twinned and detwinned), we present that data for only two of them. Sample $Y1$, with dimensions of $1.5\times0.75\times0.025$ mm^3, was detwinned. Sample $Y2$, with dimensions of $1.00\times0.7\times0.025$ mm^3, was also detwinned, and was used in the electron irradiation study.

Both the real and the imaginary parts of the ac susceptibility of the crystals were measured, using the ac option of a 1-T Quantum Design superconducting quantum interference device (SQUID) magnetometer.[11] The advantage of using a SQUID as part of the detection system is that the sensitivity of the apparatus is enhanced, even at low frequencies, where the traditional ac susceptometer has problems. The ac susceptibility was measured as a function of temperature, dc field, ac field amplitude, and frequency. Both the dc and ac fields were oriented parallel to the c axis of the crystal. For each measurement, the sample was initially zero-field cooled to the lowest desired temperature, the dc field was then applied, and the data were taken upon slowly warming.

The electron irradiation of sample $Y2$ was performed at the High Voltage Electron Microscope facility of the Argonne National Laboratory. The sample was sequentially irradiated with 1-MeV electrons at room temperature to introduce flux-pinning centers. Further details about the irradiation procedure can be found elsewhere.[6]

BACKGROUND

There has been significant theoretical interest in finding the origin of the peak in $\chi''(T)$.[2,12,13] We begin by summarizing some well-known results about the ac response of normal metals and type-II superconductors. A normal metal has a current-independent resistivity, and an applied ac field induces eddy currents which reduce the size of the oscillations of the magnetic flux inside the sample (Lenz's law). This shielding creates a nonuniform distribution of the oscillating field and current inside the sample, both of which decay on moving away from the surface toward the center. This decay occurs over a characteristic length known as the skin depth of the sample δ_s, which is proportional to $(\rho_n/f)^{1/2}$, where ρ_n is the normal-state resistivity of the sample and f is the frequency of the applied ac field. The ac response of the sample is expressed as the complex ac susceptibility χ_{ac}. The imaginary part χ'' of the susceptibility measures the dissipation in the sample, and the real part χ' measures the amount of screening. Both parts are functions only of the ratio of the skin depth to the characteristic distance of the sample in the direction of the flux penetration; e.g., for a cylinder of radius R the appropriate ratio is δ_s/R. There are three cases of interest. When $\delta_s \gg R$, the ac field largely penetrates into the entire sample, and both parts of χ_{ac} are small. When $\delta_s \ll R$, the ac field is almost completely screened (mainly confined to a narrow region near the surface). In this limit, χ'' is small and χ' is approximately $-1/4\pi$. When $\delta_s \approx R$, the screening is approximately half of $-1/4\pi$ and a step appears in χ' as the temperature is varied. This step signifies the transition from nearly perfect screening to almost complete penetration of the ac field into the sample. Also for $\delta_s \approx R$, a peak appears in χ'' as losses reach a maximum value. One can explore a wide range of values of the ratio δ_s/R by sweeping temperature for fixed f and R. This measurement of χ_{ac} is then equivalent to a measurement of ρ_n. It is important to determine that the material has linear current-voltage behavior, verifying that χ_{ac} is independent of the ac field amplitude.

For type-II superconductors, we need to distinguish between two cases. For a pinning-free superconductor, an arbitrarily small current would induce a viscous flow of the flux lines, resulting in dissipation. This regime is characterized by ρ_{ff}, the flux-flow resistivity, which is ohmic in nature. Although the origin of the dissipation caused by an ac field in a normal metal is different from that in a type-II superconductor in the flux-flow regime, the response is similar. By simply replacing ρ_n with ρ_{ff} we can calculate the flux-flow skin depth δ_{ff}. Thus, as in the case of the normal metal, the measurement of χ_{ac} is equivalent to a measurement of ρ_{ff}, which must be verified to be independent of the ac field amplitude.

For a superconductor containing pinning centers, magnetic flux lines are prevented from moving until the current density has reached its critical value, $J=J_c$. The screening currents, which are established when the applied field is changed, do not decay because there is no dissipation as long as $J<J_c$. The distribution of both the magnetic flux density and the current density are described by Bean's critical-state (CS) model.[14] As the screening current responds to the ac field, there is hysteretic energy loss. The flux and the currents penetrate to a characteristic depth, called Bean's penetration depth $L_p \propto h_{\text{ac}}/J_c$. This dependence of L_p on the amplitude of the ac field is a consequence of the nonlinear relation between J and the electric field E.[15] The two parts of χ_{ac} are functions of the ratio of L_p to the characteristic length of the sample. Keep in mind two important points: (a) the measurement of χ_{ac} is equivalent to a measurement of J_c and (b) the critical-state model applies only for ac fields with amplitude higher than the threshold value. It is clear that the presence of a single peak in the $\chi''(T)$ curve would follow from a monotonic dependence of the resistivity (or the critical current density) on the temperature.

RESULTS

In Figs. 1 and 2, we display χ' and χ'' as functions of temperature for various amplitudes of the 1-kHz ac field, superimposed on a 10-kOe dc field. Note the multiple peaks and steps appearing in the $\chi''(T)$ and $\chi'(T)$ curves, respectively, indicated by arrows. We therefore suggest that ρ or J_c has a nonmonotonic T dependence. This suggestion is based on the abrupt increase of screening a few degrees below T_c (Fig. 2). This dip in $\chi'(T)$ has been associated in the past with the peak effect.[16] To confirm this suggestion we need to determine the temperature behavior of either ρ or J_c.

First we must establish the nature of the measured losses. We know that losses in the flux-flow regime would be linear (amplitude independent, although frequency dependent), and that the hysteresis regime would be nonlinear (amplitude dependent and frequency independent).[13] We can therefore determine the nature of the losses by examining the dependence of the ac response on the amplitude and frequency of the ac field. The data displayed in Figs. 1–3 show that the components of χ_{ac} depend strongly on the amplitude of the ac field but weakly on its frequency. Hence, we identify the measured losses as being hysteretic. This conclusion is confirmed by following the procedure proposed by Civale *et al.*:[15] The χ' curves are measured for several amplitudes of the ac field, varying in the ratio of 1:2:4:8:16:32. If the critical-state model applies, we should be able to inscribe rectangles, such as *ABCD* in Fig. 2, in the χ' (as well as in the χ'') curves.

In analyzing the data we must remember that if the CS model applies then (a) χ' is a function only of L_p/R, so each horizontal line corresponds to a constant value of L_p and (b) each vertical line corresponds to a constant value of $J_c(T)$. We explain how the inscription of the rectangle *ABCD* implies that the CS model applies, remembering that $L_p \propto h_{ac}/J_c$: Since $L_p(A)=L_p(B)$ (A and B are on the same horizontal line h) and $h_{ac}(A)=2h_{ac}(B)$, $J_c(A)=2J_c(B)$. Since points B and C lie on the same vertical line v, $J_c(C)=J_c(B)$, so $J_c(C)=J_c(A)/2$. Finally, since C and D belong to the same horizontal line w, $L_p(D)=L_p(C)$. This result, coupled with the fact that $h_{ac}(D)=2h_{ac}(C)$ means that $J_c(D)=2J_c(C)$, so $J_c(D)=J_c(A)$. This conclusion is consistent with the fact that A and D must have the same J_c value, since they are on the same vertical line i. Thus, *ABCD* is a rectangle, confirming that the losses are hysteretic.

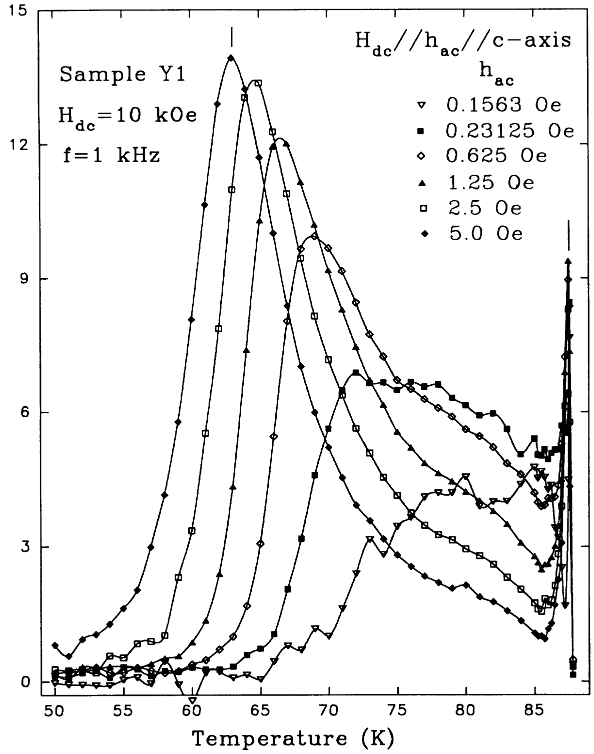

FIG. 1. χ'' vs T of sample $Y1$, measured in a dc magnetic field of 10 kOe and a 1 kHz ac field at six different amplitudes. The arrows indicate the positions of the peaks for $h_{ac}=5$ Oe.

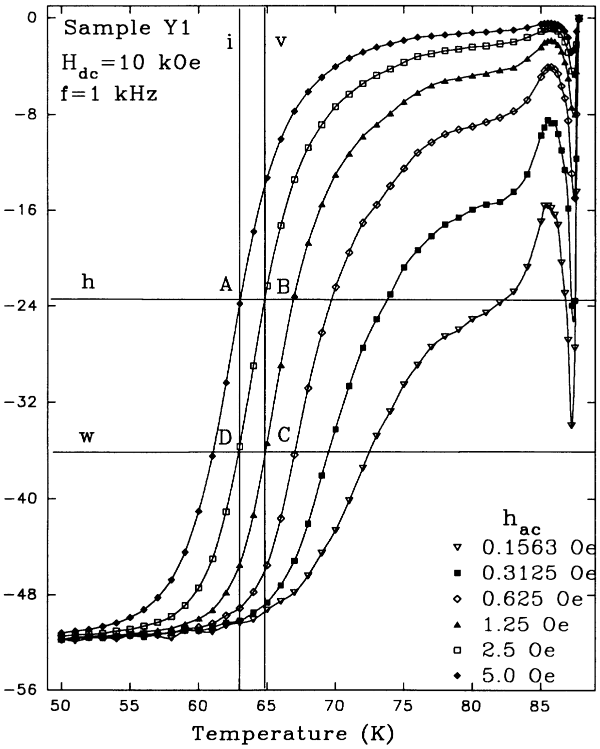

FIG. 2. χ' vs T of sample $Y1$, measured in a dc magnetic field of 10 kOe and a 1-kHz ac field at six different amplitudes, varying in the ratios 1:2:4:8:16:32. The results indicate a strong dependence of χ_{ac} on amplitude. Note the sharp increase of the screening at high temperatures followed by its abrupt decrease to zero. The rectangle construction shows that the critical-state model applies (see text).

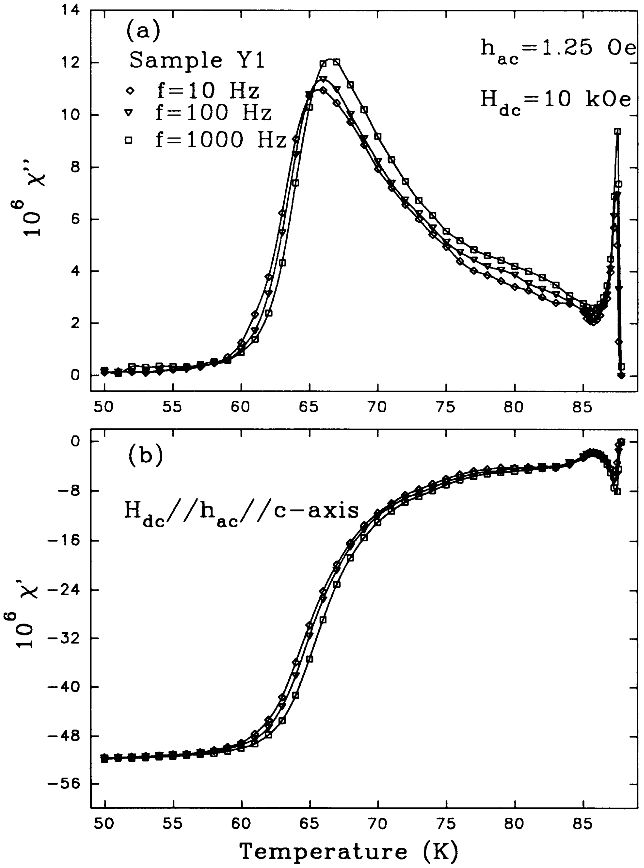

FIG. 3. (a) χ'' vs T of sample $Y1$, measured in a dc magnetic field of 10 kOe and a 1.25-Oe ac field at three different frequencies. (b) The corresponding χ' vs T curves. The results indicate a weak dependence of χ_{ac} on frequency.

We now proceed to investigate the dependence of J_c on temperature. In Fig. 4, we have plotted χ'' as a function of the amplitude of the ac field for several temperatures. According to the CS model[14] the amplitude of the ac field corresponding to the maximum χ'' value ($h_{\mathrm{ac}}^{\max}$) is proportional to J_c for that particular temperature and dc field. Therefore, by plotting $h_{\mathrm{ac}}^{\max}$ as a function of temperature for a given dc field, we determine the temperature dependence of J_c. This is shown in Fig. 5. Notice the usual behavior of J_c. Upon warming, J_c decreases rapidly until it reaches a plateau, and then increases sharply before finally dropping to zero. This is the well known peak effect, previously observed in classical superconductors.[16] It has recently been offered by others[5] as the explanation for the appearance of multiple peaks in the $\chi''(T)$ curves of YBCO crystals. In that work, however, the authors implicitly assumed that the critical-state model applies, whereas we show explicitly that it applies to our data. Also Kwok *et al.*[17] directly observed the peak effect by resistivity measurements, but they attributed it to pinning of vortices by the twin boundaries. Our ac susceptibility measurements indicate that the peak effect is present even without twin boundaries.

If the peak effect is present, one would naively expect either one or three peaks in $\chi''(T)$, depending on the amplitude of the ac field: one peak for ac field amplitudes greater than 1 Oe or lower than 0.3 Oe and three peaks for $0.3 < h_{\mathrm{ac}} < 1$ Oe (see Fig. 5). Two peaks would be present only at $h_{\mathrm{ac}} = 1$ Oe. Our data show, however, that two peaks are sometimes present at values of h_{ac} near, but not, at 0.3 or 1 Oe. The second peak is caused by the variation of J_c. It occurs when the amplitude of the ac fields is either near but below 0.3 Oe (the plateau region) or near but above 1 Oe (the maximum of the peak), so that the matching of δ_s to the sample size can be approximately, although not exactly, satisfied near the temperature of the local maximum or plateau of $h_{\mathrm{ac}}^{\max}$. Note that the amplitude of the peak induced by the variation of J_c becomes smaller as the amplitude of the ac field becomes either increasingly larger than 1 Oe or increasingly smaller than 0.3 Oe. This behavior is easily visualized by examining Fig. 4. For example follow the values of χ'' at which the vertical line crosses the curves measured at different temperatures. The value of χ'' varies as a function of temperature in such a way as to result in two peaks.

It is known that the occurrence of the peak effect depends strongly on the amount and strength of the pinning defects present in the sample.[16] It does not appear in samples which contain either no defects or a high density of defects. Our samples, which have been shown to be of very high quality[18] (exhibiting the first-order melting transition of the flux-line lattice), show a peak effect, indi-

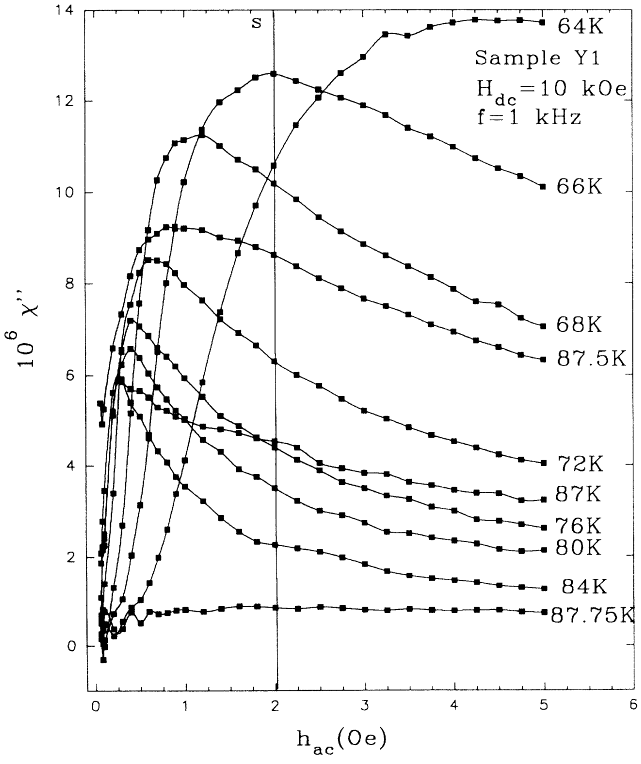

FIG. 4. χ'' vs h_{ac} of sample $Y1$, measured in a dc field of 10 kOe and 1-kHz ac fields at several temperatures. Note the nonmonotonic behavior. The peak of each curve is used to construct the J_c vs T curve displayed in Fig. 5. The vertical line is discussed in the text.

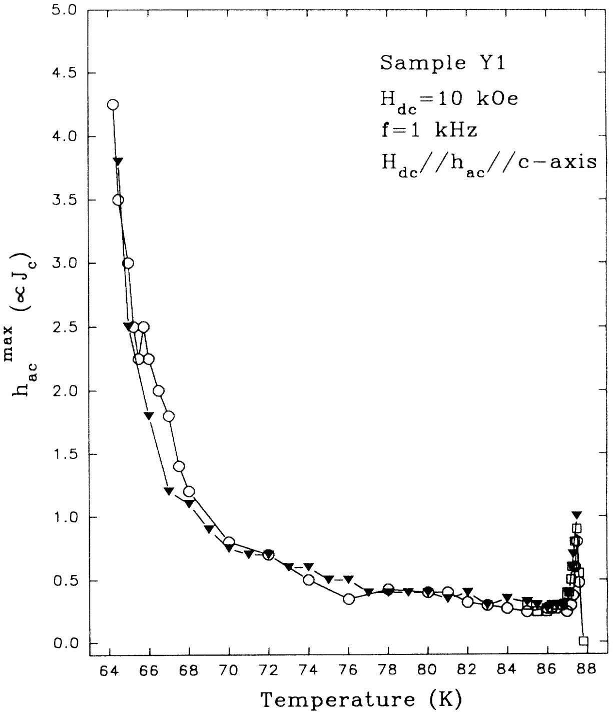

FIG. 5. $h_{\mathrm{ac}}^{\max}$ (in Oe) vs T of sample $Y1$ for a dc field of 10 kOe and 1-kHz ac fields. The different symbols correspond to different experimental runs. The reproducibility is good. The J_c values corresponding to these ac fields can be estimated by assuming that the characteristic dimension of the sample is the geometric mean of its three dimensions ($d_{\mathrm{eff}} = 0.3$ mm) and taking $d_{\mathrm{eff}} = L_p = (c/4\pi)(\mathbf{h}_{\mathrm{ac}}/J_c)$. Thus, J_c is proportional to h_{ac}. The peak effect in J_c is seen near 88 K.

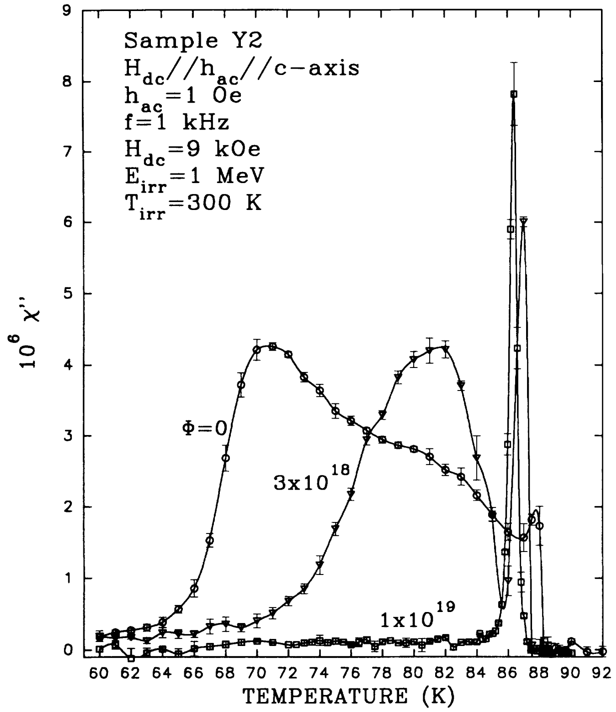

FIG. 6. χ'' vs T of sample $Y2$, measured in a dc magnetic field of 9 kOe and a 1-kHz ac field of amplitude 1 Oe before and after irradiation with 1-MeV electrons to two doses, 3×10^{18} and 1×10^{19} e/cm^2. Note that the number of peaks decreases from three to two to one.

cating the presence of some pinning defects. We have investigated the effect of point defect disorder on the dissipation peaks of $\chi''(T)$: In Fig. 6, we show the evolution of these peaks with increasing doses of 1-MeV electrons. Following the low-dose irradiation (3×10^{18} e/cm^2) the number of peaks is reduced from three to two, and the peak effect is still present. Further irradiation to 1×10^{19} e/cm^2 removes one of these two peaks. Unfortunately, we could not determine whether the peak effect is still present following the high-dose electron irradiation because we are limited to a maximum h_{ac} of 5 Oe. The presence of only a single peak at high temperature indicates that the pinning centers introduced by electron irradiation enhanced J_c of the sample.

The dependence of the high-dose peak on the ac field amplitude and frequency is shown in Figs. 7 and 8, respectively. Note that the dependence of the peak on both quantities is similar in strength, in contrast to the behavior seen in Figs. 1–3. The response is still nonlinear (amplitude dependent), but not as strongly as in the unirradiated case. Thermally activated processes are now more important, generating a frequency dependence, as one would see in the pure flux-flow regime. There is another distinct difference between the unirradiated and irradiated cases. The amplitudes of the $\chi''(T)$ peaks decrease upon reducing the amplitude or the frequency of the h_{ac} in the unirradiated case, while they remain approximately unchanged in the irradiated case. Whereas this behavior observed for the irradiated case is expected, that for the unirradiated case remains a puzzle.

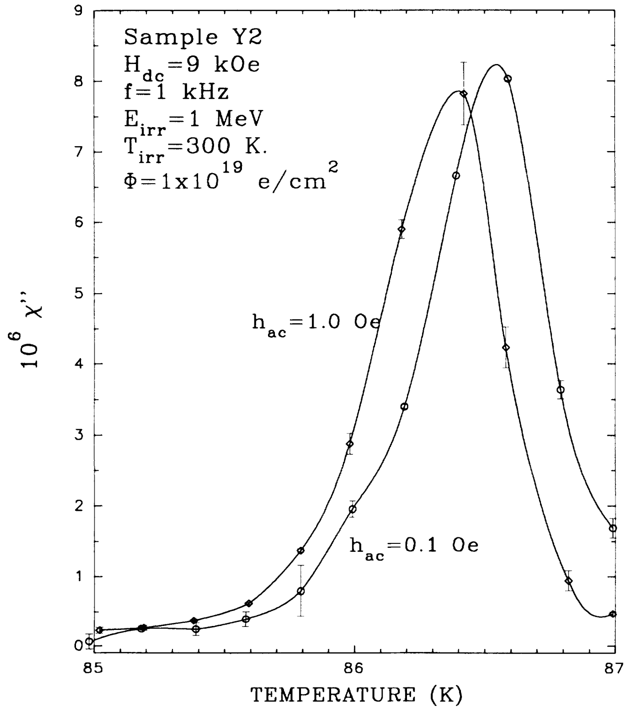

FIG. 7. χ'' vs T of sample $Y2$, measured in a dc field of 9 kOe and a 1-kHz ac field at several ac field amplitudes following electron irradiation to a dose of 1×10^{19} e/cm^2.

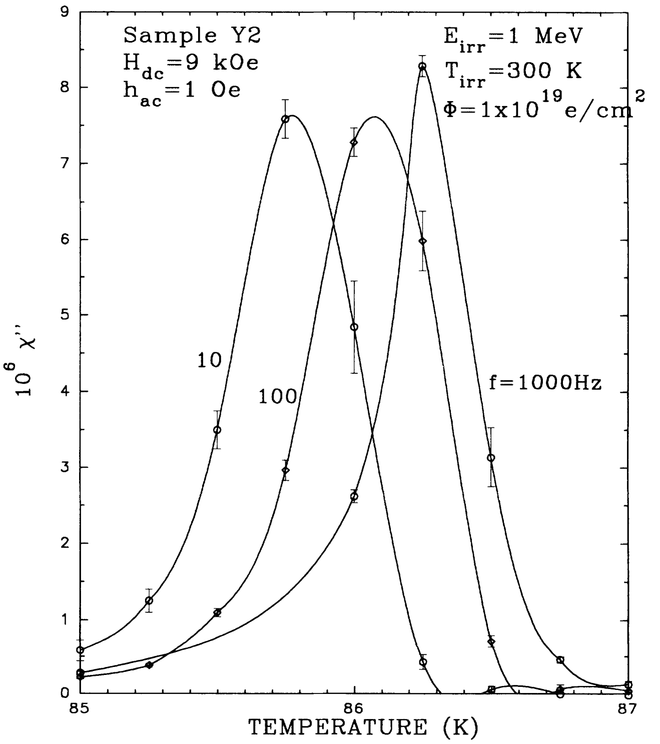

FIG. 8. χ'' vs T of sample $Y2$, measured in a dc field of 9 kOe and a 1-Oe ac field at various frequencies, after electron irradiation to a dose of 1×10^{19} e/cm^2.

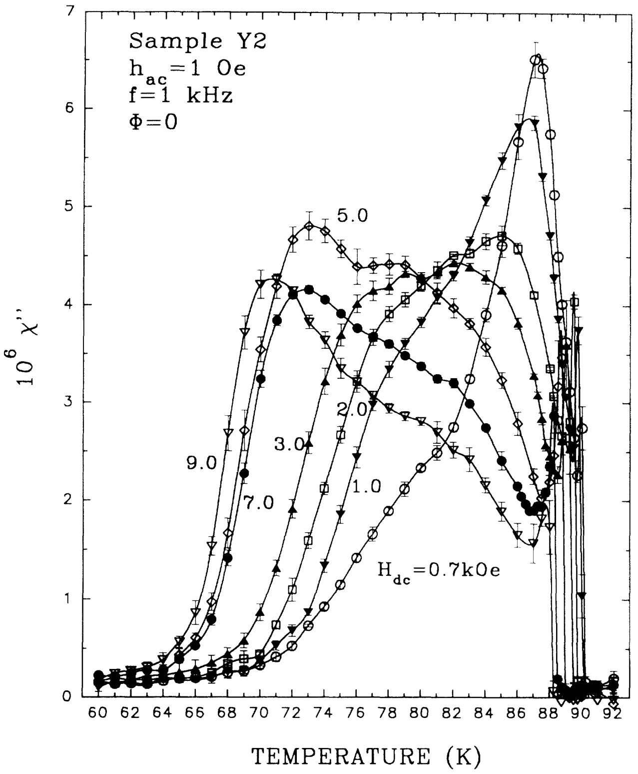

FIG. 9. χ'' vs T of sample $Y2$, measured in various dc magnetic fields ranging from 0.7 to 9.0 Oe, using a 1-kHz ac field of 1 Oe.

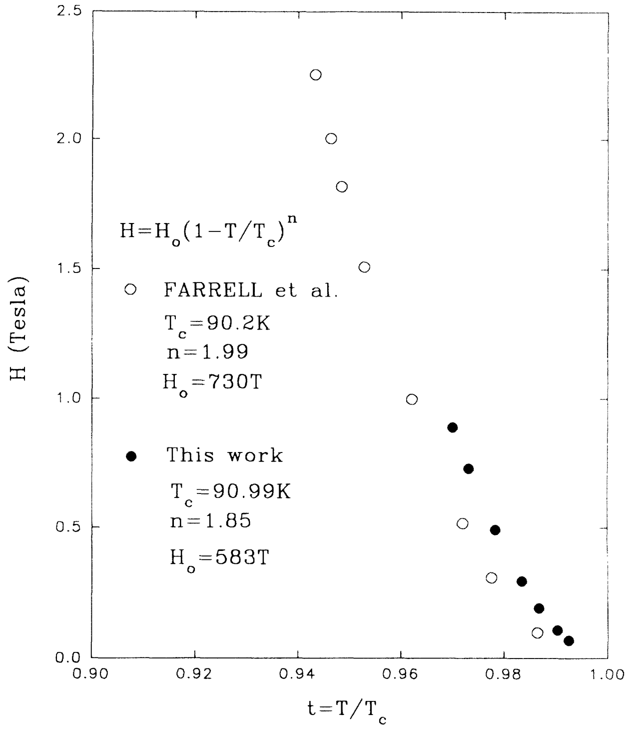

FIG. 10. Relationship between the temperature where χ' becomes zero (near T_c) and the applied dc field. For comparison, we also plot the melting line of Ref. 8 (hollow circles). Both sets of data can be fitted to an equation of the form $H=H_0(1-T/T_c)^n$, where H_0, T_c, and the exponent n are treated as fitting parameters. The values of these parameters are shown in the figure.

Another interesting feature of the data presented in Figs. 1 and 2 is the narrowness of the high-temperature $\chi''(T)$ peak and the associated abrupt decrease of the screening, shown in $\chi'(T)$. Figure 9 exhibits the dependence of this feature on the applied dc field for an undoped sample. By plotting the temperature where χ' becomes zero as a function of the dc magnetic field, we have constructed a line in the H-T phase diagram which nearly coincides with the melting line determined by Farrell, Rice, and Ginsberg[8] by using torque magnetometry on crystals of similar quality (see Fig. 10). We therefore suggest that the narrow high-temperature peak in $\chi'(T)$ indicates a first-order melting transition. We therefore propose that the enhancement of J_c at high temperatures (the peak effect) occurs because of the adjustment of the flux lines to the pinning centers. This process is assisted by the softening of the flux-line lattice prior to the melting as in classic (low-T_c) superconductors.[19]

The temperature dependence of J_c, displayed in Fig. 5, raises some questions about the accuracy of the common method of determining the irreversibility line (IRL). This method of tracking the position of the peak in the $\chi''(T)$ curve in different dc fields for an arbitrary, but fixed, value of the ac field amplitude, selects a nonzero value of J_c. The true IRL should be determined at $J_c=0$. This observation also raises questions about using dc magnetization to determine the IRL. In this case, we are limited by the sensitivity of the SQUID magnetometer which, in the case of Quantum Design, is about 1×10^{-6} emu. Using Bean's formula to calculate the minimum J_c that can be detected, we find that this value ranges between 100–1000 A/cm^2, with the lower values corresponding to large dirty samples and the higher values corresponding to small clean samples.

CONCLUSIONS

We have shown that Bean's critical-state model applies for the frequency regime investigated by us. By using this model, we have mapped out the temperature dependence of J_c. We have found that the multiple peaks in $\chi''(T)$ curves are a manifestation of a peak effect in $J_c(T)$.

We used electron irradiations to study the evolution of the multiple peaks as a function of disorder. The number of peaks (for the h_{ac} fields investigated by us) was reduced, and their positions shifted to higher temperatures as we introduced more flux-pinning defects. This indicates that J_c of the sample was enhanced. The position of the peak in $\chi''(T)$ following the high-dose irradiation exhibited a stronger frequency dependence than that observed in the unirradiated case.

We propose that the narrow high-temperature peak in $\chi''(T)$ can be associated with first-order melting of the flux-line lattice. Moreover, we suggest that the peak effect is a result of the softening of the flux-line lattice that occurs before it melts. Finally, we have shown how to determine the true irreversibility line.

ACKNOWLEDGMENTS

We are grateful for stimulating discussions with Kees van der Beek, Valerii Vinokur, and John Clem. This work was supported by the National Science Foundation Grant No. DMR 91-20000 through the Science and Technology Center for Superconductivity (J.G., D.M.G.), by NSF Grant No. DMR 89-20538 (R.L.N.), and by the U.S. Department of Energy BES-Materials Sciences under Contract No. W-31-109-ENG-38 (M.A.K.).

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