Thickness dependence of the irreversibility line in $YBa_2Cu_3O_{7-x}$ thin films

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We report a determination of the irreversibility line in the field-temperature plane of $YBa_2Cu_3O_{7-x}$ films measured by ac susceptibility. For films thicker than ~ 1000 Å, H_{irr} is similar to that observed in single crystals even though the critical current in the films is orders of magnitude larger. However, for films thinner than 1000 Å, the irreversibility line is observed to shift to lower temperatures. This suppression is discussed in terms of various models for the irreversibility line.

INTRODUCTION

One of the most intriguing properties of the high-T_c superconductors is the existence of a large region in the field-temperature (H-T) plane below the mean-field line $H_{c2}^{0}(T)$, where a linear diamagnetic response is observed,[1-3] and of a line $H_{irr}(T)$, below which the magnetic properties become irreversible and the I-V curves develop nonlinear character at low currents.[4] This phenomenon was first observed by Muller *et al.*[1] in ceramic samples of La-Ba-Cu-O. Making dc magnetization measurements, they found that the zero-field-cooling (ZFC) and field-cooling (FC) curves coincided in the region between T_c and a certain lower temperature T_{irr}. They also found that $(T_c - T_{irr}) \sim H^{2/3}$, where H is the applied magnetic field, and proposed the first theoretical description of this behavior based on an analogy with the spin glasses.[1,5] In that model the material was described as a disordered network of weak links connecting superconducting nodes. Although it was tempting to associate this network with the granular characteristics of the ceramic samples, simple scaling arguments[1] showed that, if the irreversibility line was caused by a granular behavior, then granularity occurs in a scale much smaller than the typical ceramic grains. The irreversibility line was later observed in $YBa_2Cu_3O_{7-x}$ (Ref. 6) and Bi-Sr-Ca-Cu-O (Ref. 7) crystals, in untwinned $YBa_2Cu_3O_{7-x}$ crystals,[8] and $YBa_2Cu_3O_{7-x}$ films,[9] showing that it is an intrinsic property[10] not directly related to any particular preparation technique or high-T_c system.

Even more puzzling than the universal presence of the irreversibility line is the fact that its position in the H-T plane is almost independent of the structure of defects existing in a sample. It was shown,[11] for instance, that this transition coincides in ceramics and single crystals of Bi-Sr-Ca-Cu-O. We have recently found[12] that the position of $H_{irr}(T)$ in $YBa_2Cu_3O_{7-x}$ single crystals is almost unaffected by proton irradiation, although the critical current J_c is increased by up to 2 orders of magnitude by that treatment. The irreversibility line represents the line of the H-T plane where the critical current drops to zero, and so it is surprising that its position is not affected by the large change in J_c. At a first glance, these results suggest that a melting model[13,14] is the most natural to explain the origin of the irreversibility line. According to this model, the location of the phase boundary is governed by the competition between the thermal energy and the elastic energy of the vortex lattice, the pinning energy playing no role in this picture, although it is necessary to have some pinning to account for the existence of a critical current in the frozen phase.[4] A more careful analysis shows,[12] nevertheless, that the melting model also has difficulties in explaining those results. Moreover, the lack of dependence of the location of H_{irr} on J_c does not appear to be a universal; recent results[15] show a large shift of the irreversibility line of Bi-Sr-Ca-Cu-O single crystals after neutron irradiation.

Although the existence of an irreversibility line is a well-established fact, its underlying physics is far from being understood. A wide variety of models have been proposed including the Josephson coupled glass model,[1,5] giant flux creep,[16] vortex lattice melting,[13,14] vortex glass melting,[17] and transitions within an entangled flux liquid state.[18] None of these models is yet developed well enough to provide clear quantitative predictions for the various experimental observations, and all have problems explaining some of the existing results.

This paper presents a new experimental observation of the reversible-irreversible transition—namely, the thickness dependence of the location of $H_{irr}(T)$ in $YBa_2Cu_3O_{7-x}$ thin films. We first show that the irreversibility line of $YBa_2Cu_3O_{7-x}$ thin films in the $\mathbf{H}\|\mathbf{c}$ axis configuration lays close to that of single crystals, provided that the films are thicker than ~ 1000 Å. This is by itself a notable result, considering that below $H_{irr}(T)$ the critical currents of films and crystals typically differ by 2 orders of magnitude, and again indicates a pinning-independent location of the onset of irreversibility. We also show that films thinner than 1000 Å show a progressively depressed H_{irr} in the $\mathbf{H}\|\mathbf{c}$ geometry. This result is discussed in the context of the various proposed models for the $H_{irr}(T)$ line.

EXPERIMENTAL RESULTS

Irreversibility line of crystals and thicker films

We have studied laser-ablated thin films of $YBa_2Cu_3O_{7-x}$ with thickness between 100 and 10 000 Å, epitaxially grown on (100)$SrTiO_3$ substrates, with the Cu-O planes parallel to the substrate surface. The details of the fabrication have been reported previously.[19] Films of different thickness were prepared by varying the number of laser pulses used for ablation ($\sim$0.5 Å/pulse). The thickness of the films was measured with the aid of a mechanical profilometer with a submicron stylus (Sloan Dektak 3030). Rutherford-backscattering spectroscopy (RBS) was used to estimate the thickness of the thinner films. The accuracy of the measurement, particularly for the thinnest films, is ±50 Å. X-ray analysis shows no evidence of secondary phases or regions with different crystallographic orientation, within the experimental resolution (about 5%).

We have measured the irreversibility line using an ac susceptibility technique.[20] The films were fixed on a small copper coil which is driven with a small ac current. A dc field up to 9 T was applied parallel to the c axis of the film. The inductance and the resistance of the coil were recorded as a function of temperature and dc field. The inductance change ΔL is proportional to the real part of the susceptibility of the sample, χ', and the resistance change ΔR is proportional to the imaginary part χ''. During the first stage of our measurements, we followed the usual approach of taking the maximum in χ'' as the definition of the irreversibility line. It is known[6] that $H_{irr}(T)$ defined in this way shows a small frequency dependence in $YBa_2Cu_3O_{7-x}$ crystals. All the results presented here were obtained using an ac field of 1 MHz.

Figure 1 shows the location of the irreversibility line of several laser-ablated $YBa_2Cu_3O_{7-x}$ films in the H-T plane. The corresponding results for a typical $YBa_2Cu_3O_{7-x}$ single crystal[12] are shown in the same figure for the purpose of comparison. It has been found that $H_{irr}(T)$ is very similar for a large number of different crystals. Although the critical temperatures of the films are typically a few degrees lower than those of the crystals, this minor difference does not affect the main and surprising fact—namely, that $H_{irr}(T)$ is very similar for films and crystals, although it is known that the J_c of the films is typically 2 orders of magnitude larger than for the crystals.[21,22] This result reinforces our previous conclusion[12] that the location of $H_{irr}(T)$ is largely independent of the density of defects in the sample.

Thickness dependence

Figure 2 shows $H_{irr}(t)$ (where $t = T/T_c$) for films ranging between 100 and 10 000 Å. T_c is defined as the position of the maximum of χ'' at $H=0$. It is clear that the thinner films show a depressed irreversibility line. For clarity, only some of the samples are included in this plot. The data plotted with the solid squares in Fig. 3 show the reduced irreversibility temperature t_{irr} at a particular value of the applied field, $H=7$ T, for 12 films of different thickness. It is clear from this plot that the depression of H_{irr} starts at $\sim$1000 Å. The corresponding results for typical single crystals are also included in the same figure. The same analysis performed at different fields shows similar results.

It is important to emphasize that this depression of $H_{irr}(T)$ is not a consequence of a general degradation of the superconducting properties in the thinner films. In fact, there is no evidence of degradation in any other of the superconducting parameters as the thickness is reduced. Although there is some dispersion in the values of T_c, all of them are in the range between 85 and 90 K and exhibit no obvious correlation with the thickness. In particular, our thinnest film (100 Å) has a very respectable T_c of 87.2 K. The T_c of all the films shown in Fig. 3 is listed in Table I. Also, the critical current of similarly grown films has been shown[21] to be almost independent of the thickness. The presence of second phases cannot explain the observed behavior either. It is known, for instance, that epitaxially grown Y-Ba-Cu-O films some-

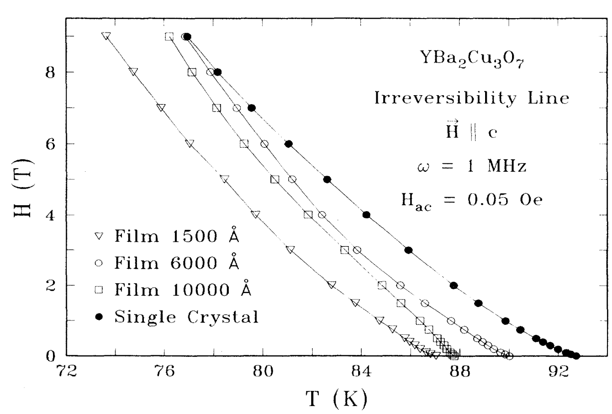

FIG. 1. Irreversibility line defined by the maximum in χ'' for various Y-Ba-Cu-O laser-ablated films of different thickness and for a typical Y-Ba-Cu-O single crystal.

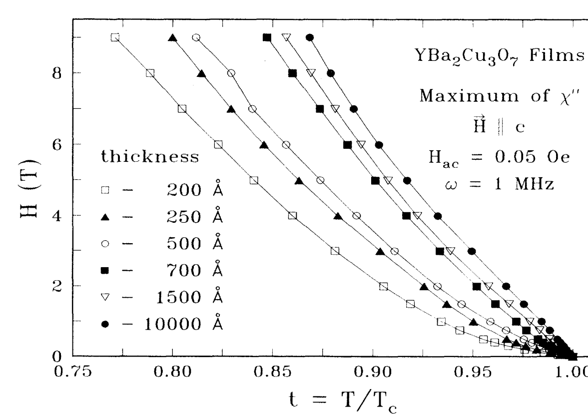

FIG. 2. Irreversibility line $H_{irr}(t)$ defined by the maximum in χ'' for Y-Ba-Cu-O laser-ablated films of different thickness. Here t is the reduced temperature, $t = T/T_c$.

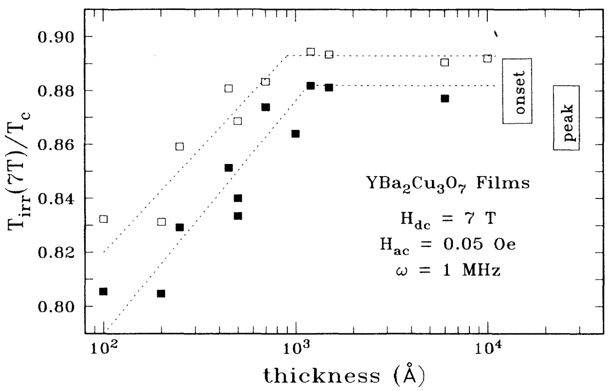

FIG. 3. Irreversibility-reduced temperature $t_{irr} = T/T_c$, at $H_{dc} = 7$ T, for Y-Ba-Cu-O laser-ablated films of different thickness. Solid symbols are defined by the maximum in χ'', as in Fig. 2. Open symbols are defined by the onset in χ'', as in Fig. 5. The rectangular boxes indicate the range of results for numerous Y-Ba-Cu-O single crystals. The dotted lines are guides to the eye.

times contain *a*-axis misoriented grains. The signal produced by those regions would correspond to the irreversibility line as observed when the applied field is parallel to the Cu-O planes. We have measured films in that configuration and found that the irreversibility line is approximately five times steeper, as will be reported elsewhere. If a significant amount of the *a*-axis oriented phase were present in any of the films, that would produce a clearly resolved peak at higher temperature, which is not observed. X-ray analysis indicates that secondary phases, if any, represent less than 5% of the films. As the sensitivity of our technique is proportional to the area of the screening current loops, the signal corresponding to those small grains would be undetectably small.

Amplitude dependence

The determination of the irreversibility line as the position of the peak in χ'' has recently been criticized[23] on the basis that the observed behavior of χ' and χ'' does indicate the presence of dissipation but not necessarily the existence of irreversibility. In fact, a similar response (namely, a drop in χ' and a peak in χ'') is observed in a normal metal due to purely electrodynamical properties. In that case, the maximum in χ'' occurs when (ignoring a geometrical factor of the order of unity), the skin depth δ equals the thickness of the sample. As δ depends on the resistivity of the material and the frequency of the measurement, it was argued[23] that the temperature dependence of the ac susceptibility of high-T_c superconductors could originate in the temperature dependence of the flux-flow resistivity ρ_{ff} and have no relation with the onset of the irreversible behavior. Such a phenomenon has indeed been observed and analyzed[24] in NbTa alloy samples which were prepared to minimize the critical current and thus the irreversibility of the magnetic response.

In spite of the similarities described above, the ac response arising from a purely ohmic character (either normal resistivity or flux flow) and that arising from an hysteretic behavior have an important difference. Since the first case represents a linear response, χ' and χ'' are independent[24] of the amplitude of the ac field used to determine them. On the other hand, the ac susceptibility of a superconductor having a finite J_c is amplitude dependent.[25] In this last case, according to the critical-state Bean model, the maximum in χ'' approximately occurs when the ac field reaches the center of the sample.[25] Disregarding again geometrical factors of the order of unity, this condition is fulfilled when $H_{ac} = J_c \times d/2$, where H_{ac} is the amplitude of the ac field and d is the relevant dimension of the sample (the thickness of the film in our case). If χ'' is measured using two different values of H_{ac}, the maximum observed for the larger amplitude corresponds to a larger value of J_c and thus will be shifted to lower temperature. The determination of the amplitude dependence of the ac response thus provides important information about the mechanisms generating dissipation in a given sample. Figure 4 shows the dissipation χ'' as a function of temperature of the 700-Å film for a dc applied field of 1 T. Each curve corresponds to a different value of the ac field. It is seen that increasing ac amplitude induces a broadening of the dissipation peak and a shift of the maximum in χ'' to lower temperatures, as predicted by the critical-state model and in qualitative disagreement with the behavior expected in a purely ohmic regime.

The previous result demonstrates that the peak in χ'' in these samples is indeed associated with irreversibility. Nevertheless, the correlation between the position of the maximum in χ'' and that of $H_{irr}(T)$ requires further analysis. As mentioned before, the irreversibility line is defined as the locus of the points in the H-T plane where J_c drops to zero. However, the maximum in χ'' corresponds to a finite value of J_c—namely, $2\,H_{ac}/d$, thus introducing[26] a systematic underestimation of T_{irr}. This fact is relevant to our analysis because, if we measure films of different thickness with the same ac amplitude, we are in fact determining the position of points with higher J_c in thinner films. This experimental artifact will

TABLE I. Critical temperature of the measured films, as determined by ac susceptibility.

Thickness (Å)	T_c (K)
100	87.16
200	87.51
250	88.49
450	88.56
500	85.90
500	86.98
700	85.22
1000	89.10
1200	89.49
1500	87.29
6000	90.01
10 000	87.77

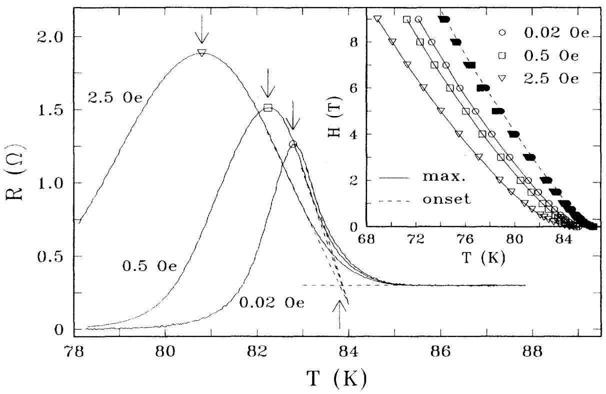

FIG. 4. Dissipation peak $\Delta R \sim \chi''$ as a function of temperature of the 700-Å film for a dc applied field of 1 T. Each curve corresponds to a different value of the ac field, as indicated in the figure. The upper arrows indicate the position of the maximum for each amplitude. The lower arrow indicates the amplitude-independent position of the onset. The inset shows the irreversibility line $H_{irr}(T)$ of the same film as defined using either the maximum or the onset of χ'', for the three values H_{ac}.

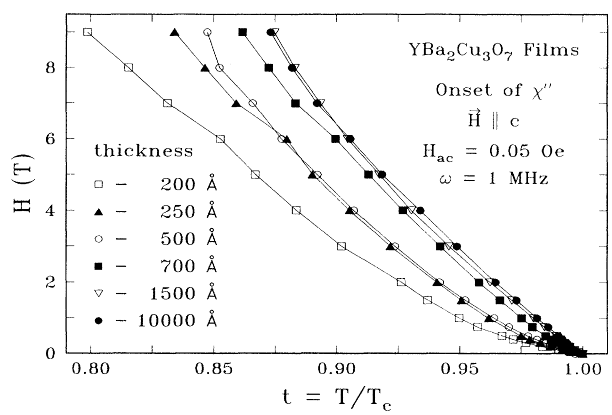

FIG. 5. Irreversibility line $H_{irr}(t)$ defined by the onset in χ'' for the same Y-Ba-Cu-O laser-ablated films of Fig. 2.

also generate a fictitious downward shift of the measured $H_{irr}(T)$ of the thinner films. There is a simple way to eliminate this problem: We can measure films of different thickness using different ac amplitudes such that the ratio H_{ac}/d remains constant. We have applied this "scaling rule" to analyze our data, and the conclusion is that the shift in the thinner films is much larger than what is expected according to this argument, thus confirming that the depression of $H_{irr}(T)$ represents an intrinsic behavior of the thinner films.

Although we have shown that the "amplitude dependence" does not modify the qualitative conclusions about the thickness dependence, it certainly affects the quantitative analysis of the data. The most convincing way to measure the temperature where $J_c=0$ is to carefully examine the *I-V* curves.[4] However, this procedure requires making good contact to the samples and in the case of films requires lithography to define a small enough sample to achieve the required sensitivity. ac susceptibility does not require contacts, is much faster to run, and is not as susceptible to artifacts due to inhomogeneities. So we decided to search for a better experimental determination of $H_{irr}(T)$ that avoids amplitude-dependent complication. A possible approach is to measure χ'' for several H_{ac} values and extrapolate the position of the maximum to $H_{ac}=0$. This procedure is slow and has obvious experimental difficulties. Another possibility is suggested by inspection of Fig. 4. It is seen there that, while the position of the maximum is amplitude dependent, the onset of dissipation at high temperature is not. We can define an $H_{irr}(T)$ curve using an "onset" criterion. The experimental procedure used to define this point is shown in Fig. 4. The inset of Fig. 4 shows the irreversibility line of the 700-Å film measured with three different ac amplitudes and defined using either the maximum or the onset of χ''. It is apparent that the last definition gives results independent of the amplitude of the ac field.

Figure 5 shows the irreversibility line of the same samples of Fig. 2, but now determined through the onset of the dissipation. In this case T_c is defined as the position of the onset of χ'' at $H=0$. It is clear that the depression of H_{irr} in the thinner films is also observed here. Finally, the open squares of Fig. 3 correspond to t_{irr} at $H=7$ T as obtained with this definition. Again, the shift to lower temperatures starts when the thickness of the film is about 1000 Å.

DISCUSSION

Many of the proposed models for the irreversibility line are consistent with the observed suppression of the irreversibility line in thinner films. The flux-creep models describe the irreversibility line in terms of a crossover from fast to slow flux dynamics determined by a pinning potential.[16] The pinning potential is typically modeled as the condensation energy times the volume of the flux bundle, $H_c^2 V_b$. As the sample thickness is reduced enough to begin to reduce V_b, the reversibility line would be suppressed, as we observe. However, recent transport measurements in films support a true phase transition to a vortex glass state at the irreversibility line, not a flux-creep crossover.[27] In the vortex glass model,[17] the three-dimensional nature of the sample is essential for the phase transition to occur. The theory suggests that as the films become thinner, a crossover from three- to two-dimensional behavior would suppress the irreversibility line. However, this model predicts a current-dependent crossover, in contradiction with our observation of a current-independent onset of irreversibility at all thicknesses. A third class of models[18] suggest that an entangled flux liquid could support critical current due to an exponentially increasing viscosity below a temperature where a barrier to flux line cutting develops. In this case, when the films become thin enough to limit entanglement, the irreversibility line would be suppressed. Although all of the mentioned models appear to be consistent with our observation of suppression, none of them is sufficiently developed to make quantitative comparisons with our data.

In summary, we have shown that the irreversibility line $H_{irr}(T)$ of $YBa_2Cu_3O_{7-x}$ films thicker than 1000 Å coincides with that of single crystals, in spite of the large difference in their critical current density. Thinner films have a progressively depressed reversible-irreversible transition. We have analyzed the influence of the amplitude of the measuring ac field in this phenomenon and proposed an alternative experimental determination of $H_{irr}(T)$ that gives amplitude-independent results. Theoretical explanation of the results could involve the response of the flux dynamics governed by pinning potentials, freezing of a flux liquid into a vortex glass state, or a viscosity transition in an entangled flux liquid regime, but theories involving these phenomena need further development to make quantitative comparison possible.

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[1]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 408 (1987).
[2]U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).
[3]Z. Hao, J. R. Clem, M. W. McElfresh, L. Civale, A. P. Malozemoff, and F. Holtzberg, Phys. Rev. B **43**, 2844 (1991).
[4]T. K. Worthington, F. Holtzberg, and C. A. Feild, Cryogenics **30**, 417 (1990).
[5]I. Morgenstern, K. A. Müller, and J. G. Bednorz, Z. Phys. B **69**, 33 (1987).
[6]A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg, and P. H. Kes, Phys. Rev. B **38**, 7203 (1988).
[7]Y. Yeshurun, A. P. Malozemoff, T. K. Worthington, R. M. Yandrofski, I. Krusin-Elbaum, F. Holtzberg, T. R. Dinger, and G. V. Chandrashekhar, Cryogenics **29**, 258 (1989).
[8]T. R. Dinger, G. J. Dolan, D. Keane, T. R. McGuire, T. K. Worthington, R. M. Yandrofski, and Y. Yeshurun, in *High-Temperature Superconducting Compounds: Processing and Related Properties,* Proceedings of the 1989 Symposium on High Temperature Oxides: Processing and Related Properties, 118th Annual Meeting of the Minerals, Metals, and Materials Society—American Institute of Metallurgy, Las Vegas, 1989, edited by S. H. Whang and A. Das Gupta (Minerals, Metals, and Materials Society Publications, Warendale, PA, 1989), pp. 23–40.
[9]J. H. P. M. Emmen, G. M. Stollman, and W. J. M. deJonge (unpublished).
[10]A. P. Malozemoff, Mater. Res. Bull. **15**, 50 (1990).
[11]H. Safar, C. Duran, J. Guimpel, L. Civale, J. Luzuriaga, E. Rodriguez, F. de la Cruz, C. Fainstein, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. B **40**, 7380 (1989).
[12]L. Civale, A. Marwick, M. W. McElfresh, T. K. Worthington, A. P. Malozemoff, F. Holtzberg, J. R. Thompson, and M. A. Kirk, Phys. Rev. Lett. **65**, 1164 (1990).
[13]P. L. Gammel, D. J. Bishop, G. J. Dolan, J. R. Kwo, C. A. Murray, L. F. Schneemyer, and J. V. Waszczak, Phys. Rev. Lett. **59**, 2592 (1987).
[14]A. Houghton, R. A. Pelcovitz, and A. Sudbo, Phys. Rev. B **40**, 6763 (1989).
[15]W. Kritscha, F. M. Sauerzopf, H. W. Weber, G. W. Crabtree, Y. C. Chang, and P. Z. Jiang (unpublished).
[16]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988); Y. Yeshurun, A. P. Malozemoff, F. Holtzberg, and T. R. Dinger, Phys. Rev. B **38**, 11 828 (1988).
[17]M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).
[18]D. R. Nelson and H. S. Seung, Phys. Rev. B **39**, 9153 (1989); M. C. Marchetti and D. R. Nelson, *ibid.* **41**, 1919 (1990); S. P. Obukhov and M. Rubinstein, Phys. Rev. Lett. **65**, 1279 (1990).
[19]G. Koren, A. Gupta, E. A. Giess, A. Segmuller, and R. B. Laibowitz, Appl. Phys. Lett. **54**, 1054 (1989).
[20]T. K. Worthington, W. J. Gallagher, D. L. Kaiser, F. H. Holtzberg, and T. R. Dinger, Physica C **153-155**, 32 (1987).
[21]T. R. McGuire, D. Dimos, A. Gupta, G. Koren, and R. B. Laibowitz, J. Appl. Phys. **67**, 5070 (1990).
[22]A. P. Malozemoff, in *High-Temperature Superconductivity II,* edited by S. H. Whang, A. Das Gupta, and R. B. Laibowitz (Minerals, Metals, and Materials Society Publications, Warendale, PA, 1990, in press).
[23]V. B. Geshkenbein, V. M. Vinokur, and R. Fehrenbacher (unpublished).
[24]J. R. Clem, H. R. Kerchner, and S. T. Sekula, Phys. Rev. B **14**, 1893 (1976).
[25]J. R. Clem, J. Appl. Phys. **50**, 3518 (1979).
[26]A. Shaulov and D. Dorman, Appl. Phys. Lett. **53**, 2680 (1988).
[27]R. H. Koch, V. Foglietti, W. J. Gallagher, G. Koren, A. Gupta, and M. P. A. Fisher, Phys. Rev. Lett. **63**, 1511 (1989).