ac susceptibility and weak-link-free behavior in an epitaxial film of $GdBa_2Cu_3O_{7-\delta}$

J. W. Li, R. L. Wang, H. R. Yi, H. C. Li, B. Yin, and L. Li
Institute of Physics, Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, People's Republic of China

(Received 15 July 1991; revised manuscript received 31 March 1992)

ac susceptibility $\chi'+i\chi''$ of an epitaxial superconducting $GdBa_2Cu_3O_{7-\delta}$ thin film was measured as a function of temperature and ac magnetic-field amplitude and frequency. Two separate loss peaks in the imaginary part χ'' were observed only at low ac magnetic fields and frequencies. The first peak near T_c is much higher than the second one located at lower temperature which is ascribed to the loss incurred because of flux motion in *a-b*-axis-oriented components in this film. The grain boundaries existing in the film are weak-link-free. The width of the first peak at 30% of the maximum is 0.5 K when the external ac magnetic-field amplitude is 0.04 Oe (rms) and frequency 737 Hz. The temperature at the maximum of the first peak is fairly linearly dependent on ac magnetic-field amplitudes. Temperatures at 90%, 50%, and 10% of the transition in the real part χ' give a good fit to a $\frac{2}{3}$-power law of the magnetic field: $j_c(T)=j_0(1-T/T_c)^{3/2}$ is derived from χ' at lower ac magnetic fields.

I. INTRODUCTION

High-temperature superconductors (HTSC's) are highly anisotropic compounds, which stems from the importance of the strongly superconducting Cu-O planes in their structure. This layered structure gives rise to intrinsic pinning of flux lines parallel to the planes. The weak superconducting coupling between the planes causes flux lines perpendicular to the planes to be very flexible. Therefore, this anisotropy will have an appreciable effect on the motion of flux lines in HTSC's. As the ac susceptibility is related to the motion of flux lines, it is believed if the ac susceptibility of a single crystal is measured when an ac magnetic field is parallel or perpendicular to one of its three principal axes, different results should be observed in both orientations of the field.

Some measurements of the complex susceptibility $\chi'+i\chi''$ versus temperature have been done on powder samples and single crystals of HTSC's.[1–10] Two loss peaks in the imaginary components χ'' were observed in some of the work[1–5] with or without a dc magnetic field superimposed. The first peak at the transition temperature was generally ascribed to the hysteresis loss of the matrix, and the second peak at the lower temperature to either that of grain boundaries[2] or the ac-field-induced dissipation at weak links.[3] Aside from all these different sources of the second peak, it can clearly be seen that the second peak in Refs. 1–5 is much higher and broader than the first. It means that the proportion of grain boundaries to the matrix in the samples measured is very large, and the effect of grain boundaries on the properties of the samples measured is much greater than that of the matrix. Moreover, temperature at the lower-temperature end of the second peak was much less than 60 K at 10 Oe (rms). It can also be seen that the second peak is very sensitive to the ac magnetic field when this field changes in a range from about 1 to 10 Oe (rms). This reflects the characteristics of weak lines. Based on this phenomenon, several models have been employed to explain the second peak, such as the grain boundary model,[11] surface model,[12] and Josephson-junction model.[13–15] However, all these models are connected with the weak-link behavior.

Dimos *et al.*[16,17] have reported that J_c is greatly reduced in the thin-film $YBa_2Cu_3O_{7-\delta}$ (YBCO) bicrystals at high-angle grain boundaries (misorientation angle $\geq 10°$) even in a small magnetic field. The reproducibility of this result for a wide range of misorientation relationships led them to conclude that all high-angle grain boundaries (high-angle GB's) act intrinsically as Josephson junctions with the weak-link behavior. Thus, if ac susceptibility of an oriented film with high-angle GB's is measured, the same magnetic-dependent behaviors like those in Refs. 2, 3, and 5 should be observed as the consequence of weak links.

This paper reports an experimental investigation and theoretical analysis of ac susceptibility of an epitaxial $GdBa_2Cu_3O_{7-\delta}$ (GBCO) thin film at the low ac magnetic fields [between 0.04 and 1.02 Oe (rms)] and frequencies (< 1000 Hz). The work focused on the behaviors of HTSC's at the transition temperature and properties of grain boundaries, and aimed to improve our basic understanding of ac susceptibility of HTSC's as a function of temperature and ac magnetic-field amplitude and frequency. A series of samples deposited on $LaAlO_3$ and ZrO_2 substrates were studied. For simplicity and clarity of exposition, only those experimental results of one superconducting GBCO thin-film sample on a $LaAlO_3$ substrate are presented here.

II. EXPERIMENTAL DETAIL

The film of $GdBa_2Cu_3O_{7-\delta}$ was fabricated using the *in situ* dc magnetron sputtering method. It was deposited on a (100) $LaAlO_3$ substrate. Detailed information about the fabrication of the film can be found elsewhere.[18] The thickness of the film measured was about 0.7 μm.

The film was characterized using x-ray diffraction (XRD) and scanning electron microscopy (SEM). From the XRD pattern, the GBCO film on $LaAlO_3$ is found to be mainly *c*-axis oriented. The SEM micrograph of the film indicates that there are some rod-shaped particles lying on top of the film which are present in two mutually perpendicular directions. It was believed[19,20] that these particles are *a*-*b* regions in a predominantly *c*-axis-oriented film matrix. In addition, it is believed that there are also some *a*-*b*-axis-oriented components (*a*-*b* components) in the inner part of our film because of their comparably strong densities of x-ray diffraction.

The equipment[21] was specially designed for use in measuring the ac susceptibility of the films. The frequency range is from 2 Hz to 25 kHz, and the ac magnetic-field amplitude is from 0.04 to 11 Oe (rms). If frequencies employed to measure the complex susceptibility are less than 50 Hz, an appropriate ac magnetic field should be used in order to obtain a sufficiently high signal-to-noise ratio. The measurements were made with the magnetic field approximately perpendicular to the surface of the films, and null adjustment of a lock-in amplifier was made at a temperature slightly above 77.8 K. The increasing or decreasing rate of the temperature at the transition was less than 0.25 K/min. The measured results were reproducible as long as all experimental conditions were the same. Compared with commercial equipment, this system is very sensitive to the weak signal excited by thin films at the transition temperature.

III. EXPERIMENTAL RESULT

The ac susceptibility $\chi'+i\chi''$ of the film was measured in a temperature range from 77 to 100 K. The data of the real components χ' versus temperature at different ac magnetic-field amplitudes (H_0) and at 137 and 737 Hz are shown in Fig. 1. Two transitions can roughly be observed by a somewhat sudden change of the slope of $\chi' \sim T$ curves in the figure. The second transition, occurring at a lower temperature than the first, reveals only a smooth tail of the first transition. From Fig. 1, the onset temperature [the temperature at which χ' (or χ'') begins to change its values in a normal state into those in a superconducting state] holds its value no matter how the field changes.

With these data it is possible to exhibit the dependence of the temperature at 10% (T_{10}), 50% (T_{50}), and 90% (T_{90}) of the transition in the real components χ' on the ac magnetic fields. The results are shown in Fig. 2. A linear dependence of each of these three kinds of temperature on $H_0^{2/3}$ is seen from Figs. 2(a) and 2(b).

Corresponding to two transitions in the real components χ', curves of the imaginary components χ'' versus temperature reveal two separate loss peaks only at low ac magnetic fields when the measuring frequency is 137 or 737 Hz. The data are shown in Figs. 3(a) and 3(b). The temperature difference between the maxima of these two loss peaks is 1.7 K when H_0 is equal to 0.04 Oe (rms) and frequency 737 Hz. If a loss peak (the first or second peak) is divided into two parts from its maximum point, the part at the lower temperature shifts a larger distance to the low temperature than the other part at the higher temperature when the ac magnetic field increases. It results in a broadening and an asymmetry of the peak. Furthermore, like that in the real components χ', the on-

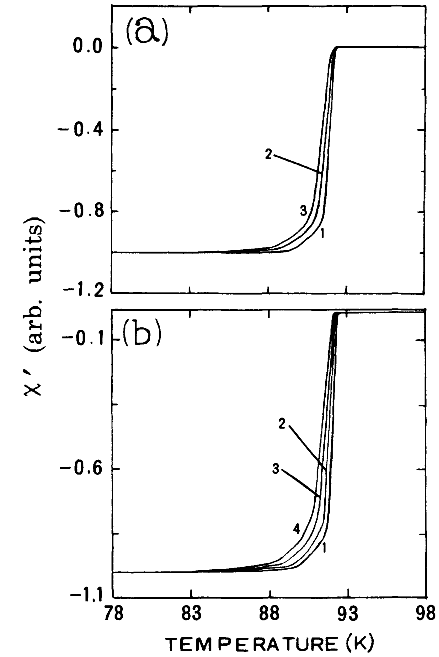

FIG. 1. χ' as a function of temperature (a) at a frequency of 137 Hz and (b) at a frequency of 737 Hz in the different ac magnetic-field amplitudes. The magnetic-field amplitudes H_0 corresponding to the curves labeled 1, 2, and 3 for 137 Hz are 0.0849, 0.399, and 1.02 Oe (rms); those labeled 1, 2, 3, and 4 for 737 Hz are 0.0425, 0.170, 0.509, and 1.02 Oe (rms), respectively. Curves at 0.299 and 0.569 Oe (rms) for 137 Hz and at 0.0849, 0.340, and 0.679 Oe (rms) for 737 Hz are not shown in Fig. 1 for clarity.

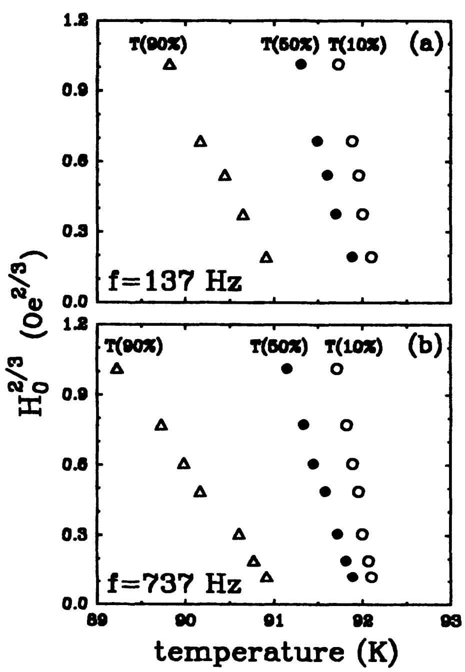

FIG. 2. Temperatures at 10%, 50%, and 90% of the transition in χ' vs $H_0^{2/3}$ (rms); the external ac magnetic-field amplitude (a) at a frequency of 137 Hz, and (b) at a frequency of 737 Hz.

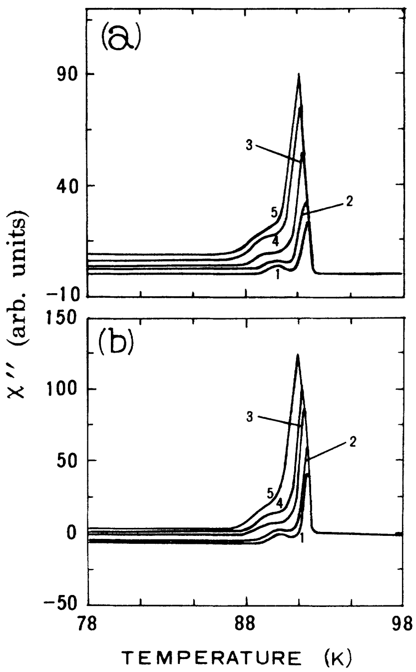

FIG. 3. χ'' as a function of temperature (a) at a frequency of 137 Hz and (b) at a frequency of 737 Hz in the different ac magnetic-field amplitudes. The magnetic-field amplitudes H_0 corresponding to curves labeled 1, 2, 3, 4, and 5 for 137 Hz are 0.0849, 0.229, 0.399, 0.569, and 1.02 Oe (rms), and for 737 Hz are 0.0425, 0.0849, 0.509, 0.679, and 1.02 Oe (rms), respectively. Curves at 0.170 and 0.340 Oe (rms) for 737 Hz are not shown in (b) for clarity.

set temperature of a loss peak in the imaginary components χ'' keeps its value at all the applied ac magnetic fields. These two kinds of onset temperature (in χ' and χ'') are equal to each other. Aside from all these mentioned above, the most important result in the data on the imaginary components is that the boundary between two loss peaks becomes indistinct as the ac magnetic field increases. These two loss peaks finally coalesce when the field is larger than 0.34 Oe (rms) for both 137 and 737 Hz, as shown in Fig. 3. This phenomenon is entirely different from the results of Shindé *et al.*[3] and Calzona *et al.*[5] In their work, the two separate peaks always stayed apart from each other as H_0 increased from about 1 to 10 Oe (rms), and the distance between the maxima of two loss peaks became larger and larger.

Because it is somewhat difficult to measure the width of a loss peak (the first or second peak) in the imaginary components χ'' in Fig. 3, the width at $(1-1/\sqrt{2})$ ($\approx 30\%$) of the maximum of the first peak was measured. Incidentally, the effect of the second peak on the width of the first peak has been simultaneously eliminated. This width versus H_0 at 137 and 737 Hz is shown in Figs. 4(a) and 4(b), respectively. The width of the first peak is 0.5 K when $H_0=0.04$ Oe (rms) and frequency is 737 Hz. Moreover, because these two peaks begin to coalesce at 0.34 Oe (rms), it is difficult to quantitatively determine how the width of the second peak changes with the field. Nevertheless, it can be seen that the second peak is

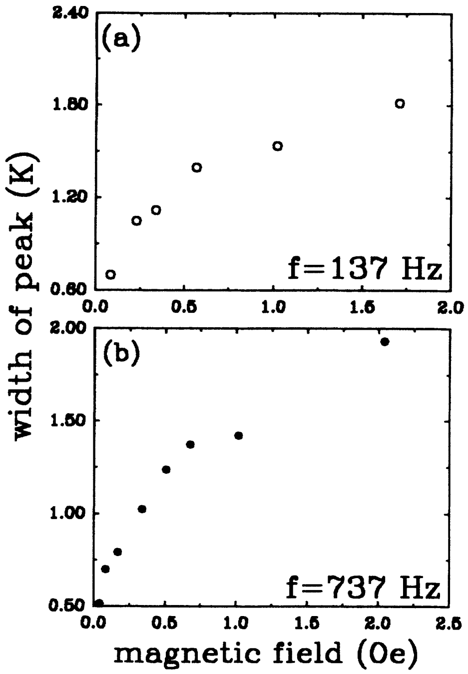

FIG. 4. The width of the first peak in χ'' vs H_0 (rms); the external ac magnetic-field amplitude (a) at a frequency of 137 Hz and (b) at a frequency of 737 Hz.

broadened with increasing H_0. In addition, Fig. 4 shows that the width of the first peak is quadratically broadened as the ac magnetic field increases, but the broadened magnitude is less than 1.5 K when the field increases from 0.04 to 1.02 Oe (rms). As the first loss peak is caused by the matrix, this small broadened magnitude is therefore an indication of insensitivity of the first peak to the ac magnetic fields.

Figure 5 shows the linear dependence of the temperature at the maximum of the first peak (T_P) in χ'' on ac magnetic fields for the film at frequency of 137 and 737 Hz. Within experimental uncertainty, the data of T_P at 137 and 737 Hz fall onto the same straight line, without any detectable difference. Especially, T_P moves to low

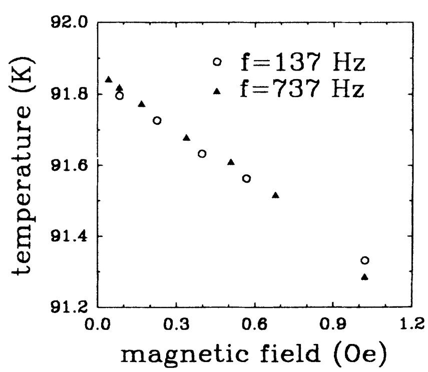

FIG. 5. Temperature at the maximum of the first peak in χ'' vs H_0 (rms); the external ac magnetic-field amplitude at frequencies of 137 and 737 Hz.

temperatures when the ac magnetic field increases, but the total displacement is only 0.56 K when the field changes between 1.02 and 0.04 Oe (rms). Therefore, it is another evidence of insensitivity of the first peak to the ac magnetic field.

IV. DISCUSSION

In our experiment it can be seen from Fig. 3 that there are two separate loss peaks only when the ac magnetic field is less than 0.34 Oe (rms). They will finally coalesce if the field continuously increases. Furthermore, the height of the second peak is much smaller than that of the first. When the ac magnetic field is 10 Oe (rms), the temperature at the lower end of the combined peak (two loss peaks have coalesced at such a high field) was equal to 78.5 K. By a comparison between the $\chi'' \sim T$ curves in Figs. 3(a) and 3(b) and by examining the changes in the second peak with varying magnitudes of the ac magnetic fields, we found that the behaviors of the second peak were nearly the same as those of the first peak which have been obtained in the discussion of Figs. 4 and 5. It is weak-link-free.

It has already been mentioned that the width and T_P of the second peak could not be determined through curves in Figs. 3(a) or 3(b). However, the real components χ' can give some information about the second peak in the imaginary components χ''. From Figs. 2(a) or 2(b), we can see that T_{90} is involved in the second transition which is corresponding to the second peak in χ''. Therefore, its magnetic dependence should reflect the characteristics of the second peak.

As with the other two temperatures T_{10} and T_{50}, in Figs. 2(a) or 2(b), T_{90} also gives a good fit to a $\frac{2}{3}$-power law of the magnetic field. That is,

$$H_0 = H(0)(1 - T_{90}/T_c)^{3/2} , \tag{1}$$

where $H(0)$ is H_0 at $T_{90}=0$ and T_c is the transition temperature. If a type-II superconductor changes from its normal state into a superconducting state in an ac magnetic field $[H(t)=H_0 \sin(\omega t)]$, two prerequisites must be satisfied: First, H_0 must be less than $H_{c2}(T_1)$, which is the upper critical field at temperature T_1. Second, the induced-current densities j due to the field must be less than $j_c(T_2)$, which is the critical current densities at temperature T_2. We select the smaller one between T_1 and T_2 as T_c^*. Therefore, when $T \le T_c^*$ the superconductor is in a superconducting state. For a type-II superconductor, there exists a mixed state when $H_{c1}(T) < H_0 < H_{c2}(T)$, where $H_{c1}(T)$ is the lower critical field. Using the equation $E = -d\phi/dt$, where ϕ is the flux variable and t is the time variable, we have

$$j = [(1-N)H_0/R] \cos(\omega t) , \tag{2}$$

where N is the demagnetizing factor and R is the resistance per unit area of the superconductor in the normal state. Thus, when T approaches T_c^* from the higher-temperature side, j must be equal or less than $j_c(T)$ for a superconductor going into the superconducting state from the normal state. That is,

$$j_c(T) \ge [(1-N)H_0/R] \cos(\omega t) . \tag{3}$$

Inserting Eq. (1) into (3) at $T \to T_c$, we have

$$j_c(T) \ge j_0(1 - T_{90}/T_c)^{3/2} , \tag{4}$$

where j_0 is equal to $[(1-N)H(0)/R]\cos(\omega t)$. The exponent is $\frac{3}{2}$, which is equal to the theoretic value for a thin film. For a weak-link junction, the critical current densities at temperature T are given by[22]

$$j_c(T) = [\pi\Delta(T)/2eR_n] \tanh[\Delta(T)/2kT] , \tag{5}$$

where R_n is the tunneling resistance per unit area of the junction existing in a superconductor in a normal state and $2\Delta(T)$ is the energy gap. When T is near T_c, $j_c(T)$ is reduced to

$$j_c(T) \propto (1 - T/T_c) . \tag{6}$$

Here, the exponent is 1.

Based on the magnetic-field-dependent behaviors of the film mentioned above, which do not correspond to weak links, and through a comparison between Eqs. (4) and (6), we concluded that the second peak is free from weak links. Its magnetic-field-dependent behaviors are similar to those of the first peak. Therefore, it is not the same mechanism as that introduced in Refs. 2, 3, and 5 that causes the second peak.

As there are some a-b components in our c-axis-oriented film, high-angle GB's inevitably exist in the film. If the conclusions of Dimos *et al.*[16,17] imply inclusion of these GB's in the high-angle Josephson-junction regime, these GB's may be the cause of the second peak whose magnetic behaviors are like those of the coupling peak introduced in Refs. 2, 3, and 5. However, no weak-link behaviors in the second peak were observed on the basis of the experimental results. It implies that these kinds of GB are like those discovered by Babcock *et al.*[23] These authors have found that J_c was not greatly reduced at some high-angle GB in a bicrystal when a magnetic field was applied. They concluded that at least some high-angle GB's can carry significant supercurrents at 77 K in high magnetic fields and they are weak-link-free. Therefore, we believed that high-angle GB's existing in our film are in this weak-link-free regime.

It is known that the imaginary components χ'' in ac susceptibility $\chi' + i\chi''$ are related to hysteresis loss at low frequencies, and the hysteresis loss to the flux pinning. As there are two kinds of oriented component in the film, which are mainly c-axis-oriented components (because the film is c-axis-oriented film) and a-b components, we ascribed two peaks observed in χ'' to hysteresis loss, respectively, occurring in both. But it only means that the two peaks are caused by anisotropy of the pinning rather than the respective pinning mechanisms existing in these two components. In view of the fact that HTSC's are layered compounds, we believe that the intrinsic pinning[24,25] plays an important role in this pinning anisotropy. The original model for intrinsic pinning assumed the flux lines to be situated between the layers for magnetic fields in the a-b plane. When the coherence length ξ_c perpendicular to the layer is of the order of the Cu-O plane, spacing

s at the low temperature[26] or larger than s near T_c,[27,28] a strong modulation of the order parameters arises and the free energy of the superconductor is minimized. In other words, the strong modulation of the parameters acts as pinning centers when the flux lines are situated in between the layers. Clearly, it costs superconducting condensation energy for a flux line to cross a layer. Consequently, they prevent the motion of flux lines across the layers. For this reason, the pinning strength along the a-b planes is stronger than that which is parallel to the c axis. Therefore, for a c-axis-oriented film with a-b components, this fact determines the loss in a-b components occurring at a lower temperature than that in the c-axis-oriented components as the field is parallel to the c axis of the film. This is in accord with the experimental results.

In conclusion, performing ac susceptibility measurements, we have observed that there were two loss peaks in χ'' at different magnetic fields and frequencies in a superconducting $GdBa_2Cu_3O_{7-\delta}$ thin film. The first peak is caused by the matrix and the second peak is due to the a-b components in our c-axis-oriented thin film. It is a direct consequence of the strong anisotropy of HTSC's. From the real components χ' we have deduced that $j_c(T)=j_0(1-T/T_c)^{3/2}$ at T_c when the ac magnetic field was less than 1.02 Oe (rms). Combining this information with an analysis of the imaginary components χ'', we have concluded that the GB's in our c-axis-oriented film were weak-link-free.

ACKNOWLEDGMENTS

We are grateful to Changan Wang and Yuhe Wang for helpful discussions. This work was supported by the National Science Foundation of China, the National Center for Research and Development on Superconductivity of China, the Beijing Zhongguancun Associated Center of Analysis and Measurement, and the Structure Research Laboratory of Chinese University of Science and Technology.

[1] D.-X. Chen, R. B. Goldfarb, J. Nogués, and K. V. Rao, J. Appl. Phys. **63**, 980 (1988).
[2] M. Nikolo and R. B. Goldfarb, Phys. Rev. B **39**, 6615 (1989).
[3] S. L. Shindé, J. Morrill, D. Goland, D. A. Chance, and T. McGuire, Phys. Rev. B **41**, 8838 (1990).
[4] H. Mazaki, M. Takano, Y. Ikeda, Y. Bando, R. Kanno, Y. Takeda, and O. Yamamoto, Jpn. J. Appl. Phys. **26**, L1749 (1987).
[5] V. Calzona, M. R. Cimberle, C. Ferdeghini, M. Putti, and A. S. Siri, Physica C **157**, 425 (1989).
[6] A. P. Malozemoff, T. K. Worthington, Y. Yeshurun, F. Holtzberg, and P. H. Kes, Phys. Rev. B **38**, 7203 (1988).
[7] F. Gömöry and P. Lobotka, Solid State Commun. **66**, 645 (1988).
[8] Yu Mei, H. L. Luo, D. X. Chen, J. Nogues, and K. V. Rao, J. Appl. Phys. **64**, 2533 (1988).
[9] L. T. Sagdahl, S. Gjolmesli, T. Laegreid, K. Fossheim, and W. Assmus, Phys. Rev. B **42**, 6797 (1990).
[10] Hiromasa Mazaki, Mikio Takano, Ryoji Kanno, and Yasuo Takeda, Jpn. J. Appl. Phys. **26**, L780 (1987).
[11] D. S. Ginley, E. L. Venturini, J. F. Kwak, R. J. Baughman, B. Morosin, and J. E. Schirber, Phys. Rev. B **36**, 829 (1987).
[12] R. B. Goldfarb, A. F. Clark, A. I. Braginski, and A. J. Panson, Cryogenics **27**, 475 (1987).
[13] H. Küpfer, I. Apfelstedt, W. Schauer, R. Flükiger, R. Meier-Hirmer, and H. Wühl, Z. Phys. B **69**, 159 (1987).
[14] J. F. Kwak, E. L. Venturini, D. S. Ginley, and W. Fu, in *Novel Superconductivity,* edited by S. A. Wolf and V. Z. Kresin (Plenum, New York, 1987), pp. 983–991.
[15] D. K. Finnemore, R. N. Shelton, J. R. Clem, R. W. McCallum, H. C. Ku, R. E. McCarley, S. C. Chen, P. Klavins, and V. Kogan, Phys. Rev. B **35**, 5319 (1987).
[16] D. Dimos, P. Chaudhari, J. Mannhart, and F. K. LeGoues, Phys. Rev. Lett. **61**, 219 (1988).
[17] D. Dimos, P. Chaudhari, and J. Mannhart, Phys. Rev. B **41**, 4038 (1990).
[18] H. C. Li, H. R. Yi, R. L. Wang, Y. Chen, B. Yin, X. S. Rong, and L. Li, Appl. Phys. Lett. **56**, 2454 (1990); Huai-ren Yi, Rui-Lan Wang, Hong-cheng Li, Y. Chen, Bo Yin, Xi-sheng Rong, and Lin Li, *ibid.* **56**, 2231 (1990).
[19] T. S. Ravi, D. M. Hwang, R. Ramesh, Siu Wai Chan, L. Nazar, C. Y. Chen, A. Inam, and T. Venkatesan, Phys. Rev. B **42**, 10 141 (1990).
[20] C. W. Nieh, L. Anthony, J. Y. Josefowicz, and F. G. Krajenbrink, Appl. Phys. Lett. **56**, 2138 (1990).
[21] J. W. Li, H. C. Li, and L. Li (unpublished).
[22] V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963).
[23] S. E. Babcock, X. Y. Cai, D. L. Kaiser, and D. C. Larbalestier, Nature **347**, 167 (1990).
[24] M. Tachiki and S. Takahashi, Solid State Commun. **70**, 291 (1989).
[25] B. I. Ivlev and N. B. Kopnin, Phys. Rev. B **42**, 10 052 (1990).
[26] B. I. Ivlev, N. B. Kopnin, and V. L. Pokrovsky, J. Low Temp. Phys. **80**, 187 (1990).
[27] B. I. Ivlev and N. B. Kopnin, J. Low Temp. Phys. **80**, 161 (1990).
[28] W. K. Kwok, U. Welp, V. M. Vinokur, S. Fleshler, J. Downey, and G. W. Crabtree, Phys. Rev. Lett. **67**, 390 (1991).