ac susceptibility measurements and the irreversibility line of high-temperature superconductors

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(Received 22 November 1991; revised manuscript received 6 February 1992)

The irreversibility line for high-temperature superconductors has been reported in the literature both from dc magnetization measurements and from ac susceptibility measurements. The latter, determined from the temperature of the peak of the absorptive component of the susceptibility, are controversial because the magnetic-field-energy absorption can be interpreted in different ways. It is shown here that the temperature dependence of the absorptive susceptibility peak measured as a function of ac magnetic field alone for single-crystal $Bi_2Sr_2CaCu_2O_{8+y}$, $Tl_2Ba_2CaCu_2O_{8+y}$, and $YBa_2Cu_3O_{7-x}$ is not related to the irreversibility line. However, the same quantity measured using a weak ac field in the presence of a larger dc field gives good agreement with the irreversibility line determined from dc magnetization data for these materials. An explanation is given for why the absorption peaks are different in the two cases, and it is shown where the relationship to the irreversibility line arises. The case of a ceramic sample is considered, and it is demonstrated that ac measurements cannot be used for irreversibility-line measurements in this material because of internal-magnetic-field effects.

INTRODUCTION

Soon after the discovery of high-temperature superconductors (HTSC's), Müller, Takashige, and Bednorz[1] reported the existence of a magnetically reversible region above a magnetic-field–temperature line in the La-Ba-Cu-O system using dc magnetometer measurements. They called this magnetic-field–temperature relation a quasi–de Almeida–Thouless line[2] because of its similarity to properties of a spin glass.[3] More recently, Malozemoff *et al.*[4] have termed the above field-temperature relation the "irreversibility line" (IL) because it appears to separate a higher-temperature, magnetically reversible region from a lower-temperature region exhibiting magnetic hysteresis. Two methods were used to determine the IL by Malozemoff *et al.*: (1) They measured field-cooled (FC) magnetization and zero-field-cooled (ZFC) magnetization with a dc magnetometer to determine the temperature above which the FC and ZFC values coincide for a given magnetic field, and (2) they determined the temperature of the peak in the absorptive component χ'' of the ac susceptibility in a given magnetic field. The first method is straightforward in interpretation since the coinciding of the FC and ZFC data above a particular temperature for a given magnetic field implies reversible flux motion, i.e., no flux pinning. The second method, however, is controversial because a χ'' peak can be caused by any of several energy-absorptive processes, the peak may be frequency dependent, and the physical basis of χ'' in these materials is not well understood.[5] For example, some energy-absorptive sources in superconductors are the pinning of magnetic flux by structural or impurity defects, flux trapping by potential barriers in the structural lattice, or unrecognized mechanisms. As these effects increase or decrease with temperature or dc magnetic field, ac absorption (χ'') increases or decreases. Understanding of these processes is complicated by time-dependent effects, which lead to flux creep[6] or flux flow[7] in the HTSC materials.

Discussions at a recent conference[8] illustrated differences of opinion about the interpretation of χ'' and the IL. The purpose of this paper is to show under what conditions the χ'' peak can be used to define the IL and in general clarify how χ'' relates to the magnetic properties of HTSC materials.

EXPERIMENTAL DETAILS

The ac apparatus and techniques used here to measure the complex susceptibility of HTSC materials have been described earlier.[9] Briefly, a sample is mounted in the core of the secondary of a mutual-inductance coil set balanced for zero output voltage in the absence of a sample. A 0.1–20-kHz ac current in the coil set primary produces an ac magnetic field at the sample. If the sample becomes superconducting, a signal proportional to its susceptibility is developed where both the real (χ') and imaginary or absorptive (χ'') components can be measured with a phase-sensitive detector. In the present work the ac-field strength can be varied from 0.07 to 4.48 Oe rms. An axial 0–400-Oe dc magnetic field can be applied at the same time to the sample. Temperature runs were made on each sample in two ways: (1) χ' and χ'' were measured as a function of ac-field strength (zero dc field) as the sample temperature was slowly cooled from above T_c to low temperature. (2) χ' and χ'' were measured as a function of dc field and temperature for a small (0.07–0.2 Oe) constant ac field as the sample temperature was lowered from above T_c. The dc magnetization of the samples was measured with a Quantum Dynamics MPMS superconducting quantum interference device (SQUID) magnetometer. This method is well described in the literature.[10] Here a sample was measured in a constant magnetic field as it was cooled from above T_c to low temperature (field cooled). The sample was also measured by first cooling in zero field to low temperatures (5–10 K) and then applying a dc field and measuring the magnetization as the sample was warmed to above T_c (zero field cooled).

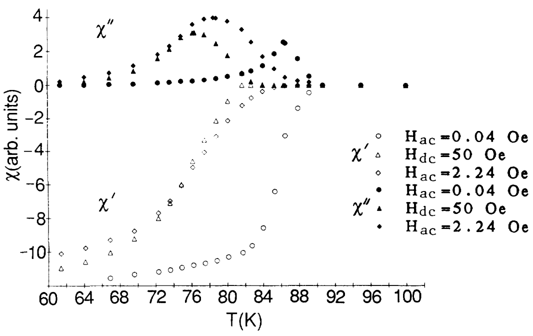

FIG. 1. Magnetic susceptibility vs temperature at various magnetic-field conditions for single-crystal $Bi_2Sr_2CaCu_2O_{8+y}$.

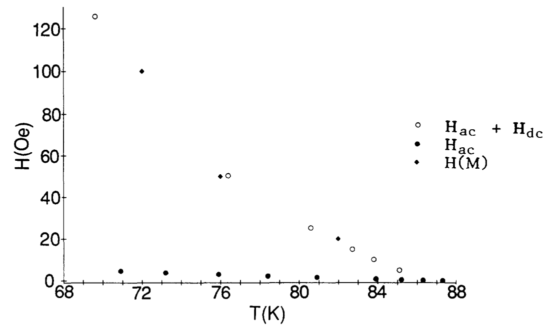

FIG. 3. Irreversibility line for single-crystal $Bi_2Sr_2CaCu_2O_{8+y}$. Also plotted are χ'' peak temperatures vs ac and ac + dc magnetic fields.

RESULTS

Typical ac χ' and χ'' measurements at a frequency of 500 Hz are shown in Fig. 1 for a single crystal of $Bi_2Sr_2CaCu_2O_{8+y}$. Three cases are shown: (1) low ac field, zero dc field, (2) low ac field, larger dc field, and (3) relatively large ac field, zero dc field. In (1) the field is not large enough to penetrate the sample except very close to T_c; χ' shows a relatively sharp transition and full flux exclusion at low temperature, and χ'' shows a relatively sharp peak just below T_c. In (2) the dc field is large enough to penetrate the sample at higher temperatures, so that χ' is reduced and χ'' shows a broader peak at a lower temperature than in (1). In (3) the same effects on χ' and χ'' are seen as in (2), although the ac field is much smaller in size than the dc field used in (2). In the case of single-crystal $YBa_2Cu_3O_{7-x}$, the curves for (1) and (2) are the same and similar to the case of (1) for $Bi_2Sr_2CaCu_2O_{8+y}$ because the available dc field is not large enough to affect the sample except very close to T_c. However, in case (3) for $YBa_2Cu_3O_{7-x}$, the relatively weak ac field does produce a measurable shift in the χ'' peak temperature, as shown in Fig. 2. Results for the FC- and/or ZFC-determined IL for $Bi_2Sr_2CaCu_2O_{8+y}$ are shown in Fig. 3, where the lowest temperature of the coinciding values of the two magnetization curves for a given magnetic field is plotted versus dc field. Also plotted in Fig. 3 is the χ'' peak temperature versus magnetic field for the case of an ac field only and the case of a small ac field with larger dc field. Similar plots are shown in Fig. 4 for a single crystal of $Tl_2Ba_2CaCu_2O_{8+y}$ and in Fig. 5 for a ceramic rod of $YBa_2Cu_3O_{7-x}$.

DISCUSSION

Figures 3 and 4 show that, similar to the results obtained by Malozemoff *et al.*[4] for $YBa_2Cu_3O_{7-x}$, the data for the χ'' peak temperature T_p versus magnetic field for the case of a small ac field and larger dc field coincide closely with the IL obtained with SQUID measurements for both $Bi_2Sr_2CaCu_2O_{8+y}$ and $Tl_2Ba_2CaCu_2O_{8+y}$.

An example of how this data can be used is shown in Fig. 5. The thermally assisted flux-flow (TAFF) theory proposed by Kes *et al.*[7] and others[11] predicts that the quantity

$$1-T_p/T_c \propto H^{2/3} ,$$

where T_p is the temperature of the χ'' peak, T_c is the superconducting transition temperature, and H the applied dc magnetic field. A plot of $1-T_p/T_c$ versus $H^{2/3}$ is shown to be linear for both $Bi_2Sr_2CaCu_2O_{8+y}$ and $Tl_2Ba_2CaCu_2O_{8+y}$ in Fig. 5.

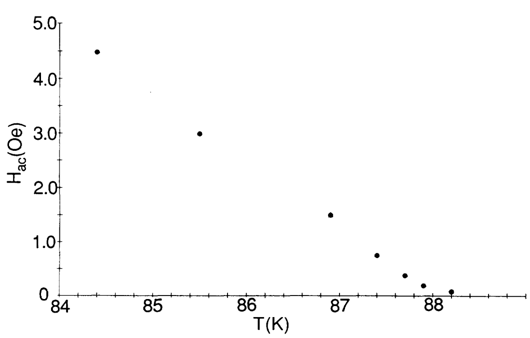

FIG. 2. χ'' peak temperature vs ac magnetic field for single-crystal $YBa_2Cu_3O_{7-x}$.

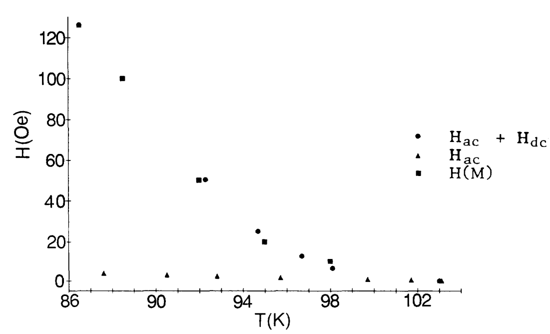

FIG. 4. Irreversibility line for single-crystal $Tl_2Ba_2CaCu_2O_{8+y}$. Also plotted are χ'' peak temperature vs ac and ac + dc magnetic fields.

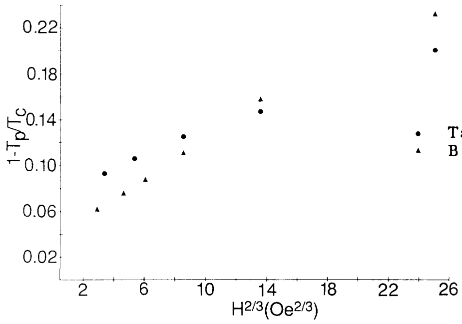

FIG. 5. $1-T_p/T_c$ vs $H^{2/3}$ for Bi_2Sr_2, $CaCu_2O_{8+y}$, and $Tl_2Ba_2CaCu_2O_{8+y}$.

However, the plots of χ'' T_p versus ac magnetic field alone for these two materials do not agree at all with the SQUID IL, nor does the plot for $YBa_2Cu_3O_{7-x}$ shown in Fig. 2, where the SQUID IL would be vertical on the scale shown. The decrease in T_p for only an ac magnetic field appears to be linear with magnetic field for all three materials.

The agreement of one χ'' peak and magnetic-field plot with the FC and ZFC IL but not with the other plot described above can be understood from the following arguments. With only an ac magnetic field present, the peak in χ'' versus temperature occurs in the critical-state model[12] at the point where the ac magnetic-field flux just penetrates to the center of a sample.[13] The explanation for this occurrence is that if one first considers a superconductor far below T_c with full flux exclusion and $H_{ac} < H_{c1}$, the screening currents generated by the alternating field are confined near the sample surface and little or no magnetic loss takes place: χ'' is small or zero. As the temperature is allowed to increase and both J_c and H_{c1} decrease, the field begins to penetrate the sample and χ'' increases as the magnetic-energy absorption or loss increases. This process and thus χ'' continue to increase until the field penetrates the sample completely (reaches the center of the sample). As the temperature continues to increase, less and less of the sample volume remains superconducting as the magnetic flux penetrates more and more of the sample, so that less and less magnetic loss takes place and χ'' decreases, eventually reaching zero at T_c. Thus, in this process, χ'' peaks when the ac field reaches the center of the sample, whether other loss mechanisms are occurring or not, so that this measurement does not directly relate to, for example, reversibility or irreversibility loss mechanisms. In the data shown, the χ'' peak temperatures for only an ac field present are at field values much below those of the SQUID IL measurements and the curve slopes of the two plots are quite different.

In the case of a weak ac field superimposed upon a larger dc field, the change in magnetization of the sample due to the ac field represents a (very) minor hysteresis loop on the overall magnetization of the sample; in effect, the ac measurement in this case is sampling the magnetic state of the material. If there is an IL in the sample being measured, then as the temperature is lowered from T_c, in the magnetically reversible region, the initially zero χ'' will increase with decreasing temperature as the flux motion, in response to the alternating field, becomes more and more viscous for lowering temperature. When the temperature decreases below the IL value at the measurement dc field, the flux motion begins to be impeded by now active pinning sites, less magnetic energy is absorbed, and χ'' decreases, approaching zero at low temperatures where most of the flux is pinned. Thus, in this experimental situation, χ'' should peak at the temperature of the IL line. The data of Fig. 3 and 4 show this to be the case, where there is good agreement with the SQUID IL data.

A complication with ac measurements of HTSC materials is that the ac results are frequency dependent. This factor has been explained as the result of thermally activated flux-flow processes[7] or by a vortex-glass theory.[1] Experimentally, it has been shown[14] that below about 1 kHz the loss mechanisms are essentially independent of frequency, so that for comparison with dc results measurements below 1 kHz should be used, as in the present work.

A further complication with determining the IL is that the internal magnetic field of the material is the relevant field, so that the demagnetizing factor for the sample shape must be considered. Thus the internal magnetic field $H_{in} = H_{ex} - DM$, where H_{ex} is the applied field, D the demagnetizing factor, and M the effective sample magnetization. Since for a superconductor M is negative and often large, a sample with an appreciable demagnetizing coefficient will have an internal field greater than the applied field. For accurate comparison of different measurements, then, either the demagnetizing field must be calculated for each situation or the same sample or field orientation must be used for both the magnetization and ac measurements, as is the case for the present work.

For a ceramic $YBa_2Cu_3O_{7-x}$ sample, as shown in Fig. 6, there are other considerations, since in this case the weak ac-field or larger dc-field measurement of the IL differs appreciably from the SQUID-determined IL. In this porous structure individual grains are joined together by small contact areas. In the latter regions the magnetic-field lines are concentrated by the repulsion of the surrounding bulk grains. This concentration in-

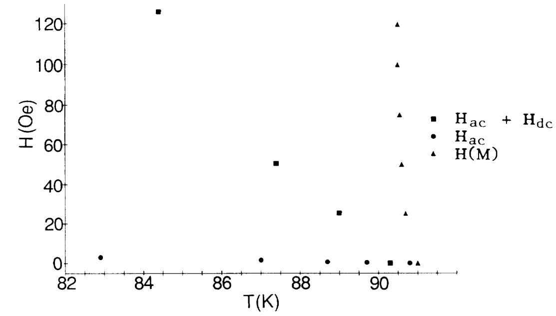

FIG. 6. Irreversibility line for ceramic $YBa_2Cu_3O_{7-x}$. Also plotted are χ'' peak temperatures vs ac and ac + dc magnetic fields.

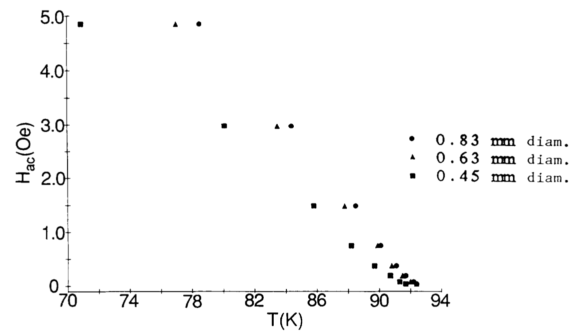

FIG. 7. χ'' peak temperature vs ac magnetic field for different sample diameters of ceramic $YBa_2Cu_3O_{7-x}$.

creases the strength of the local field in the small cross-section areas to above that of the external applied field, producing an apparent downward shift in the IL.[14] This explanation has been shown by Askew *et al.*[15] to explain irreversible critical-current behavior in $YBa_2Cu_3O_{7-x}$.

For the case of an ac field alone for ceramic material, the χ'' peak temperature position should be at the point where the field H^* penetrates to the center of the sample; thus it should be a function of the size of the sample. To test this assumption measurements were first made for the original rod-shaped sample, with an initial diameter of 0.83 mm. The sample was next reduced in diameter by mechanical scraping to 0.63 mm and the measurements repeated. Finally, the diameter was further reduced to 0.45 mm and again measured. The results of this experiment are show in Fig. 7. The above theory predicts that at a given temperature the smaller the sample diameter, the smaller is the field required to reach the sample center. The data of Fig. 7 agree with this prediction. Although the functional sample-size dependence of H^* appears nonlinear, the sample size could not be adjusted accurately enough to determine an analytical expression for the H^*-versus-size relation.

CONCLUSIONS

We have shown in this paper how peaks in the absorptive component of the susceptibility χ'' are related as a function of temperature to the penetration of magnetic fields into a high-temperature superconductor. In the case of an ac magnetic field alone, the peak position is determined by the field reaching the center of the sample and, thus, by the geometrical size of the latter. This assumption was shown to be valid for a ceramic sample of $YBa_2Cu_3O_{7-x}$. For a small ac field superimposed upon a larger dc field, the χ'' peak position is a function of magnetic-field-energy absorption, which peaks at the boundary between reversible and irreversible regions in the material. This χ'' peak position was shown to coincide with the irreversibility line defined by zero-field-cooled and field-cooled dc magnetization measurements for $Bi_2Sr_2CaCu_2O_{8+y}$ and for $Tl_2Ba_2CaCu_2O_{8+y}$, as was earlier shown for $YBa_2Cu_3O_{7-x}$. Finally, the ac or dc field procedure to determine the IL line does not work for ceramic materials because of internal-field effects of the granular structure.

ACKNOWLEDGMENTS

We acknowledge helpful discussions with T. R. Askew. The single-crystal samples were provided by M. A. Subramanian and the ceramic $YBa_2Cu_3O_{7-x}$ by N. McN. Alford.

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