Thermal scanning ac susceptometry method for high-T_c superconductor studies

A. C. Bódi
Institute of Experimental Physics, Kossuth University, P.O. Box 105, Debrecen, 4001 Hungary

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Conventional ac susceptibility measurements and the conditions used in thermal analysis methods have been combined for normal-superconductor transition studies. Accordingly, during the ac susceptibility measurements, the Y-based sample is exposed to a temperature gradient. By controlling this gradient, the boundary between normal and superconducting region can be swept slowly at will across the specimen. The extended thermal scanning assures a high resolution to the new method. The observed fine details (even discontinuities) of the susceptibility spectra demonstrate its performance and basis.

I. INTRODUCTION

During different studies on high-T_c superconductor materials, various anomalies[1–3] and fine structures[4,5] have been observed. Unfortunately, conventional ac magnetic field susceptometry (one of the most commonly used method for normal-superconductor transition studies), avoiding temperature gradients in samples by fast heat removal, can detect—among the creep and jump type motions of fluxes[6]—only the flux creep. For this reason it provides only continuous spectra.[7] In order to fit the conventional ac susceptometry for observing structured susceptibility curves, both at superconducting-normal (SN) and normal-superconducting (NS) transitions, the susceptibility measurements have been combined with the conditions used in thermal analysis methods.[3] In the new thermal scanning susceptometry (TSS) method, created by analogy with differential scanning calorimetry, the sample is exposed, by slow heat removal, to a (variable) temperature gradient. One of the possibilities—to realize an easily controllable temperature gradient along the sample—is to keep one end (top) of the sample at a constant (T_t), while the other end (bottom) is at a variable temperature (T_b). With adequate variation of T_b, the boundary between normal and superconducting regions can be swept, fulfilling the requirements, slowly across the specimen. So the extension of the transition can be varied by controlling the sweeping time and temperature interval. Because the local inhomogeneities (flux jumps) appearing in these thermal conditions are not immediately compensated, they became observable.

II. EXPERIMENT

For susceptibility measurements, the single coil method has been used (Fig. 1). The dimensions of the pick-up coil were adapted to the dimensions of the samples. First, the variation of the empty coil's (base line) and after that of the filled coil's inductance and dissipation factor have been determined as a function of temperature. A computer-controlled precision LCR meter (Hewlett–Packard type 4284 A) has been used for the measurements (made at 10 kHz, 100 kHz, and 1 MHz frequencies). In all the experiments, the basic material was the same. Seven samples (cut from the same high quality, one component $Y_{1.01}Ba_{1.95}Cu_{2.97}O_x$ pellet having $T_c \sim 92$ K, and 4.82 g/cm^3 density),[8] of different dimensions (from $2\times2\times6$ to $6\times6\times24$ mm^3) have been studied. The top of the samples was kept at the temperature (T_t) of liquid nitrogen. The bottom temperature (T_b) was varied between room and liquid-nitrogen temperatures by using a cold finger. T_b was monitored by a Fe-constantan thermocouple, in close contact with the bottom of the sample. The resolution was $\sim$0.01 K.

In order for the TSS method to be suitable for detecting fast changes in the sample's parameters, the thermal relaxation time τ_1 of the sample (in the heat sink direction) has been chosen to be short compared with the sample-bath relaxation time τ_2 ($\tau_1 \ll \tau_2$). We have $\tau_1 = C_s d_s^2/\eta_s$ and $\tau_2 = C_s d_s d_r/\eta_r$, where d_s and d_r are the sample and the thermal resistor lengths (distance between the sample and the heat sink), η_s and η_r are the sample and the resistor thermal conductivity, respectively, and C_s is the sample specific heat.[3]

Between the room temperature and the onset temperature of superconductivity (T_0), the curves obtained for the empty and filled coils practically coincide with each other. For this reason, we considered the variation of the empty coil's parameters as a base line between T_0 and 77 K. While this variation remains, both for L and D measurements, below 5%, it can be neglected as a first approximation (Fig. 2).

One can relate the changes in inductance (dissipation factor) of the coil to the real (imaginary) component of the susceptibility. For evaluation of the real part of the ac susceptibility (χ'), the following two conditions have been assumed: the $\chi'=0$ value (zero diamagnetism) corresponds to the base line inductance L (at T_0 or 77 K), and the $\chi'=-1$ value (total diamagnetism, when the entire geometric volume of the granular sample is shielded) corresponds to the inductance $L=0$ (Fig. 3). As a first approximation, a linear scale has been used between the 0 and -1 susceptibility values.

Because the ac magnetic field penetrates into the sample along the junctions between the grains and in the grains to the extent of the skin depth, the field inside the sample cannot be considered to be uniform. Now, this problem is

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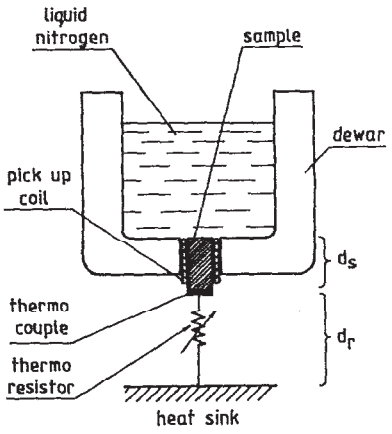

FIG. 1. Schematic drawing of the experimental setup.

automatically resolved. The pick-up coil's inductance depends on the volume integral of the square of magnetic field ($L \propto \int H^2 dV$): so its measured value corresponds to the real field distribution in the sample.

For the cases $\chi' = 0$ and $\chi' = -1$, the magnetometric demagnetizing factor (N), depending mainly on the length-to-diameter ratio of the sample, can be calculated. According to the $N < 0.1$ data of Ref. 9, the surface field can be found within a percent of the applied field. Hence, N can be neglected as a first approximation. The ratio of our sample's density to the ideal density gives a filling factor of $\sim 75\%$. Since there are 5% nonreacted Y phases present,[8] a superconducting volume fraction greater than $\sim 70\%$ shows that the whole NS transition is practically finished. This is true for all the samples used at 77 K. So, for $\chi' \leq -0.7$ cases, in calculating the internal susceptibility, we used the same demagnetizing factor as at $\chi' = -1$. In cooling or in heating, the longitudinal demagnetizing

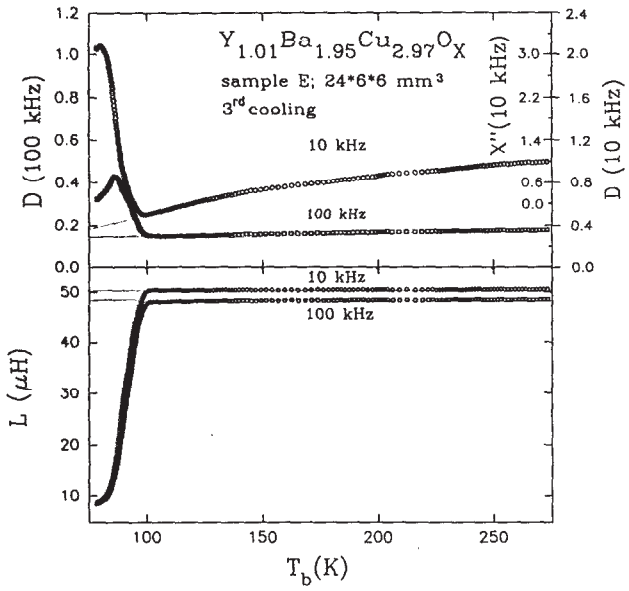

FIG. 2. Inductance L and dissipation factor D of sample E vs temperature measured at the bottom of the sample (T_b). The dotted lines denote the base lines.

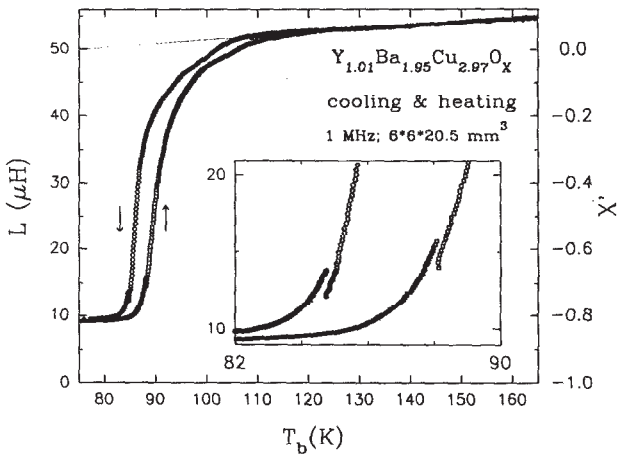

FIG. 3. Inductance and the corresponding real part of the external ac susceptibility vs T_b for cooling and heating. The inset shows in detail a part with discontinuities.

factor varies together with the superconducting volume fraction. This variation is hard to determine. (In our samples, for a variation of the sample's length with 400%, N varies by 63% only. Therefore, the influence of N should not be overestimated.) For these reasons, the factor calculated for constant susceptibilities has been used between 0 and -0.7 susceptibility values, too.

The imaginary part χ'' can be determined through the measured dissipation factor $D = R/\omega L$ using $\chi'' = 2\,\Delta D$, where ΔD is the variation of D from its value measured at T_0 or 77 K (Fig. 2).

The ac magnetic field intensity employed (< 1 Oe) was not enough to separate the intrinsic and coupling components.

III. RESULTS

In all the measurements, χ' shows a drop from 0 to as low as ~ -0.7, ~ -0.8, (dimensionless and in SI units), depending on temperature (Fig. 3). The variation of χ' reflects the change in the ratio of the superconducting to normal volume fractions. The variation of D (and χ''), being proportional to the losses, exhibits a large, frequency-dependent maximum (Fig. 2). The maximum occurs when the ac field extends to the middle of the grains. These results are typical and correspond to data obtained with standard ac susceptometry. The advantages of the TSS method became evident if an extended temperature scale is used: fine details (even discontinuities) of the transition can then be observed (Figs. 3 and 4). The discontinuity begins both for cooling and for heating at the same χ' and T_b values, respectively (on Fig. 4 at $\chi'_b \sim -0.56$ and $T_{bb} \sim 93.5$ K). T_{bb} does not depend on the frequency used (Fig. 5), while χ'_b does not depend on the number of thermal cycles (Fig. 6). The amplitude of discontinuity decreases with the thermal cycles.[8] It is probable that the spectrum is a superposition of a continuous (global flux creep) and of a discontinuous (local flux jump) component, because they are two slightly different superconducting phases of the pellet.[10] The origin and na-

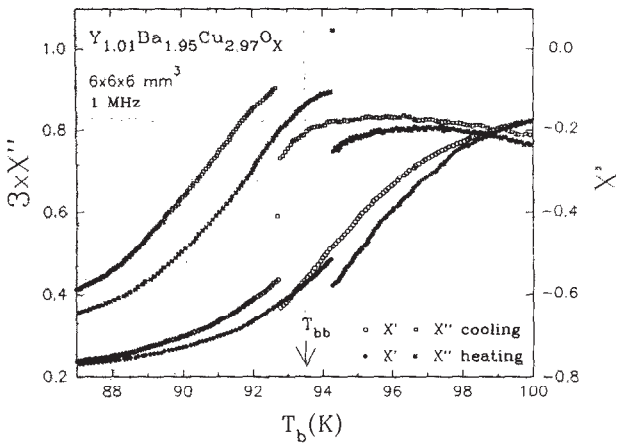

FIG. 4. External ac susceptibility (real and imaginary parts) vs T_b at cooling and heating. The temperature T_{bb} indicates the beginning of anomaly.

ture of the local zones, producing discontinuities in susceptibility spectra, are not really understood yet.

Since the TSS method is an inductive one that senses the changes of the magnetic field in the entire volume of the granular sample—independently of their location and of the temperature profile along the specimen—our external susceptibility data are correct and reproducible. So the existence of the partly known temperature distribution and stresses, along the specimen, only complicates the analysis of susceptibility data, but does not make it impossible. However, a temperature gradient ($\sim$100 K) results in a flux motion and in the generation of dc voltages ($\sim$100 nV) in the direction, and perpendicular to the direction of the gradient.[11] These voltages are of no great importance for ac measurements, as a result of their low-level and dc nature.

The validity of the method has been rigorously verified under different thermal conditions. A fine structure of a

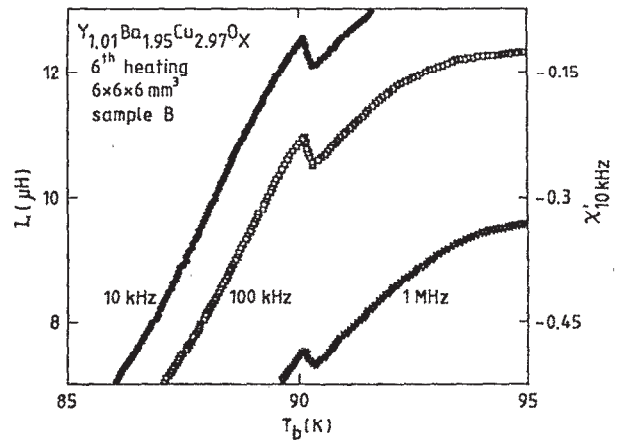

FIG. 5. The anomaly of the inductance (sample B) after six slow (2 K/min) thermal cycling between 300 and 77 K measured during the 6th heating process at 10 kHz, 100 kHz, and 1 MHz frequencies. The plots demonstrate that the position of anomaly does not depend on frequency. The superconducting volume fraction (69% at 10 kHz, 66% at 100 kHz, and 62% at 1 MHz) decreases weakly with frequency.

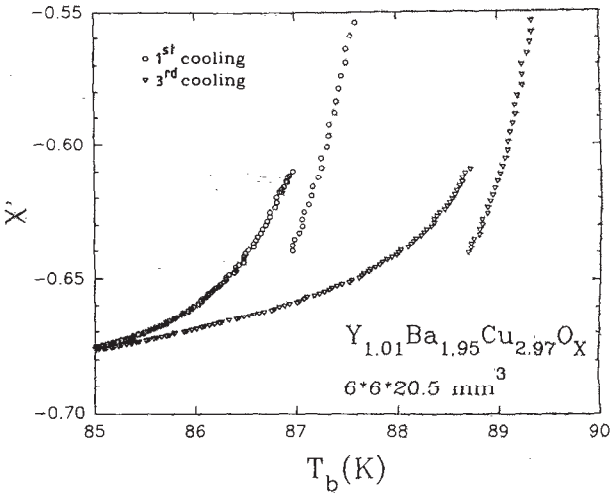

FIG. 6. Real part of the external ac susceptibility vs T_b measured during the first and third cooling process, using a field of 1 MHz frequency.

sample's transition always begins at the same superconducting volume fraction, independent of the thermal conditions, and in the case of identical scanning conditions, at the same $T_b = T_{bb}$ temperature. This excludes accidents and suggests a close correlation between the details of the spectra and the material structure of the samples. The finding of the same T_{bb} value, with both cooling and heating (Fig. 4), indicates the presence of a quasilinear temperature profile. In this case, the analysis becomes more accurate.

IV. DISCUSSION

Combining conventional ac susceptometry with the thermal conditions used in the differential scanning calorimetry (DSS), a new high-resolution susceptometry (TSS) with enlarged possibilities has been created. Just as commercially obtainable modulated DSS (MDSS) installations show, the nonreversing and reversing heat flow components, the present TSS shows the real and imaginary parts of the susceptibility. Owing to the fact that the susceptibility can be measured without contacts, and more exactly than the heat flow, whenever the TSS is applicable (that is, for all thermally activated transitions, connected with magnetic parameter changes), it is more advantageous to use it instead of MDSS.

The main advantage of the TSS method (over the MDSS and conventional susceptometry) is its capability of revealing subtle and fast quantitative changes. The changes can be correlated with the material structure of the sample. Discontinuities in ac susceptibility curves, discovered by the TSS method, both at NS and SN transitions, demonstrate the performance and basis of this new method.

The procedure, however, has some inconveniences. The applied thermal scanning may produce structural changes in the sample. (Even these changes are observable[8] with TSS.) At different scanning speeds and intervals, the transition of a given zone, which always happens at the same critical temperature, can occur at different bottom

temperatures. So, the T_b scale is not absolute. Due to the fairly complicated experimental conditions, the analysis of data is rather difficult, but it is more fruitful after all than for conventional susceptometry.

Although the experimental phenomena reported here are believed to be generic to Y-Ba-Cu-O, similar studies of other materials (including films, single crystals, powders, and pellets with different grain sizes and methods of preparation) are also essential and may reveal additional phenomena.

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[1] S. N. Artemenko, I. G. Gorlova, and Yu. I. Latyshev, JETP Lett. **46**, 654 (1989).
[2] Hoydoo You, U. Welp, and Y. Fang, Phys. Rev. B **43**, 3660 (1991).
[3] S. E. Inderhess, M. B. Salamon, I. P. Rice, and D. M. Ginsberg, Phys. Rev. B **47**, 1053 (1993).
[4] A. C. Bódi, I. Kirschner, and S. Leppävuori, Phys. Lett. A **158**, 318 (1991).
[5] V. Foukis, O. Dobbert, K.-P. Dinse, M. Lehning, T. Wolf, and W. Goldacker, Physica C **156**, 467 (1988).
[6] P. V. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964); M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682 (1969).
[7] R. B. Goldfarb, M. Lelental, and C. A. Thompson, in *Magnetic Susceptibility of Superconductors and Other Spin Systems*, edited by R. A. Hein, T. L. Francavilla, and D. H. Liebenberg (Plenum, New York, 1992), and references therein.
[8] I. Kirschner, A. C. Bódi, S. Leppävuori, A. Uusimäki, I. Dódony, and T. Porjesz, Phys. Lett. A (in press).
[9] S. D. Murphy, K. Renouard, R. Crittenden, and S. M. Bhagat, Solid State Commun. **69**, 367 (1989).
[10] I. Kirschner, S. Leppävuori, A. C. Bódi, A. Uusimäki, and I. Dódony, 3rd World Congress on Superconductivity, Munich, Session Energy/Power 8C, 1992, p. 1; Appl. Superconductivity **1**, 1721 (1993).
[11] M. Zeh, H.-C. Ri, F. Kober, R. P. Huebener, J. Fischer, R. Gross, H. Müller, T. Sermet, A. V. Ustinov, and H.-G. Wener, Physica C **167**, 6 (1990).