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Influence of thermal scanning and local inhomogeneity on the shape of AC susceptibility curves of high-T_C superconductors

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Abstract

The effects of the temperature gradient (p) and heat transfer's velocity (q) parameters on the shape of AC susceptibility curves were experimentally explored. At homogeneous $Y_{1.01}Ba_{1.95}Cu_{2.97}O_{6.86}$ ceramic samples only broadening with negligible horizontal translation and shape distortions of the χ' and χ'' susceptibility curves were observed. At samples with local inhomogeneity—beside broadening—a proportional peak-deep-hump type distortion and horizontal translation of the χ'', and rotational translation of the χ' curves appeared. The results—leading to a clearer understanding of how inhomogeneities and the scanning parameters influence the shape of the AC susceptibility curves—allow separating the effects of local inhomogeneities from the ones produced by structural modifications. It is confirmed that the spatial resolution of the method for local inhomogeneities is proportional to p and inversely proportional to q until a limit is determined mainly by the relaxation time of the studied phenomenon.

Keywords: AC susceptibility measurement; Thermal scanning; Cuprate superconductors; Spatially inhomogeneous structures; Phase transition

1. Introduction

Glassy systems can display complex collective behavior characterized by the formation of synchronous clusters [1]. In high-T_C superconductors there are many ordered phases which appear to compete and sometime coexist [2]. Due to structural relaxation processes, their response to a change in temperature near the phase transitions is not instantaneous and is inherently non-linear. The dynamics of real systems, especially of cuprate superconductors has been the subject of numerous investigations [3]. Thermal scanning susceptometry (TSS) [4,5], like the different variants of the scanning calorimetry (DSC, MDSC) [6–8], can help to detect the kinetics of structural relaxations and additional phenomena [9–11]. In the TSS method, the presence of the temperature gradient ($p = \Delta T/\Delta x$) in the sample indicates weak thermal coupling between the sample and the thermal bath. In opposition to the isothermal methods, where the global response of the whole volume of the sample is received, at weak coupling, partial volumes of the sample are successively explored (scanning). One of the possibilities—to realize an easily controllable temperature gradient along the sample—is to cool down or heat up one end of the sample via a variable thermal resistor. The speed of its variation determines the temperature scanning rate, $q = \Delta T/\Delta t$. For highest

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spatial inhomogeneity resolution *q* should be small whereas *p* should be high. For the highest sensibility of the method, the requirements are antagonistic; *p* should be small, and *q* should be high. The experimental set-up determines the limits of an acceptable compromise. Sample inhomogeneities influence on the results of the magnetic measurements [12].

The purpose of this work was to study comparatively the effects of the parameter values *p* and *q* and local inhomogeneities on the shape of χ' and χ'' AC susceptibility curves for $Y_{1.01}Ba_{1.95}Cu_{2.97}O_{6.86}$ ceramic samples.

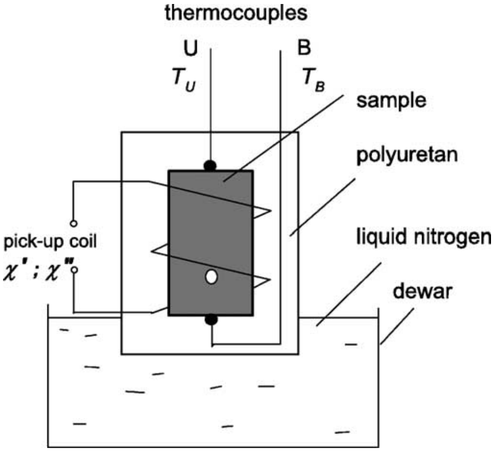

Fig. 1. Schematic drawing of the experimental set-up. The pick-up coil is wound around the sample. χ' and χ'' are measured in phase and out of phase AC voltages proportional to the real and imaginary components of the AC susceptibility. U and B are J-type thermocouples measuring the temperatures of the upper (T_U) and the bottom (T_B) end. The primary AC field coil is not marked.

2. Experimental

Three samples (A—5 mm × 5 mm × 4 mm; B1—4 mm × 5 mm × 7 mm; C—4 mm × 5 mm × 7 mm) were prepared from fine powder using the dynamic pressing method. This method produced compact samples possessing very smooth surfaces. These features have a significant importance in thermally non-equilibrium experiments. According to X-ray diffraction studies, these specimens were very close to the single-phase material. Only the component of Y(123) exists in a significant quantity (>95 mass%) in the samples. All important physical and chemical parameters of the $Y_{1.01}Ba_{1.95}Cu_{2.97}O_{6.86}$ samples ($T_C \approx 92$ K) have been published earlier [13]. The studied specimen was inserted in a pick-up (secondary) coil connected to a commercial two phase lock-in amplifier (Brookdeal

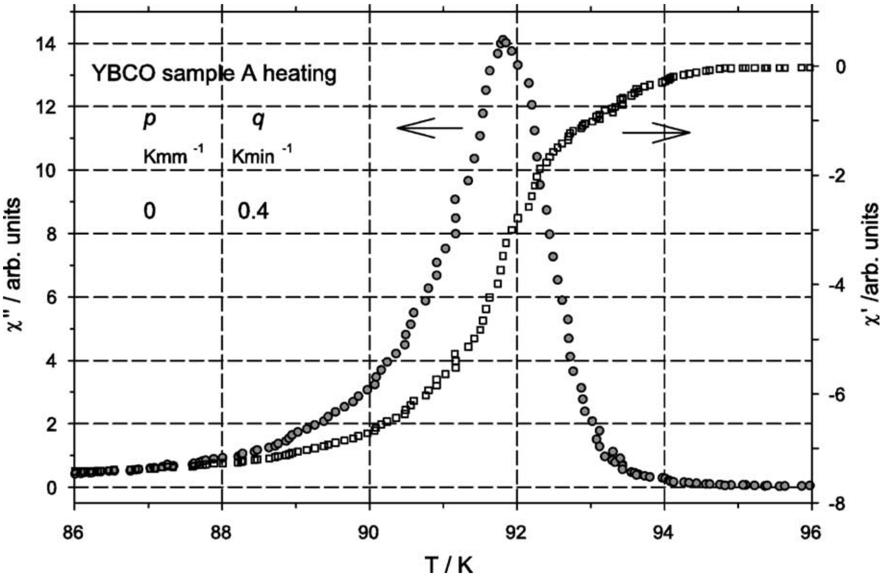

Fig. 2. The χ' and χ'' curves of the sample A (5 mm × 5 mm × 4 mm) measured by conventional method using a commercial lock-in amplifier vs. the temperature *T* measured by J-type thermocouple.

Electronics Princeton Applied Research type 5206) (Fig. 1). For comparison, first the complex susceptibility of a homogeneous sample (A) was determined using the conventional (without temperature gradient) AC method. Following this, the homogeneous B1 sample was studied, using the TSS method. In the sample C (initially identical to sample B1) a hole ($\Phi = 1\,\mathrm{mm}$) was drilled at 1/3 height for local inhomogeneity effect studies. For temperature gradient measurements two J-type thermocouples (U, B) were used; one at the top and another one at the bottom end of the sample. The sample was coated with a neutral silicone rubber layer. The sample–secondary coil-thermocouple system was inserted into the primary coil powered by the lock-in amplifier ($f = 5\,\mathrm{kHz}$). The AC current used produced a magnetic field of the order $H \sim 10^{-4}\,\mathrm{T}$ on the surface of the sample. A polyurethane foam layer (thermal insulator) surrounded hermetically the sample-secondary coil-thermocouple system.

In order for the TSS method to be suitable for detecting fast changes in the sample's parameters, the sample-bath thermal relaxation time has been chosen to be long compared with the relaxation time observed at the normal-superconductor domain fluctuation in the sample.

During the relatively high number (~50) of thermal scanning, the bottom end of the sample was cooled and the top end was heated. For this reason, the sample

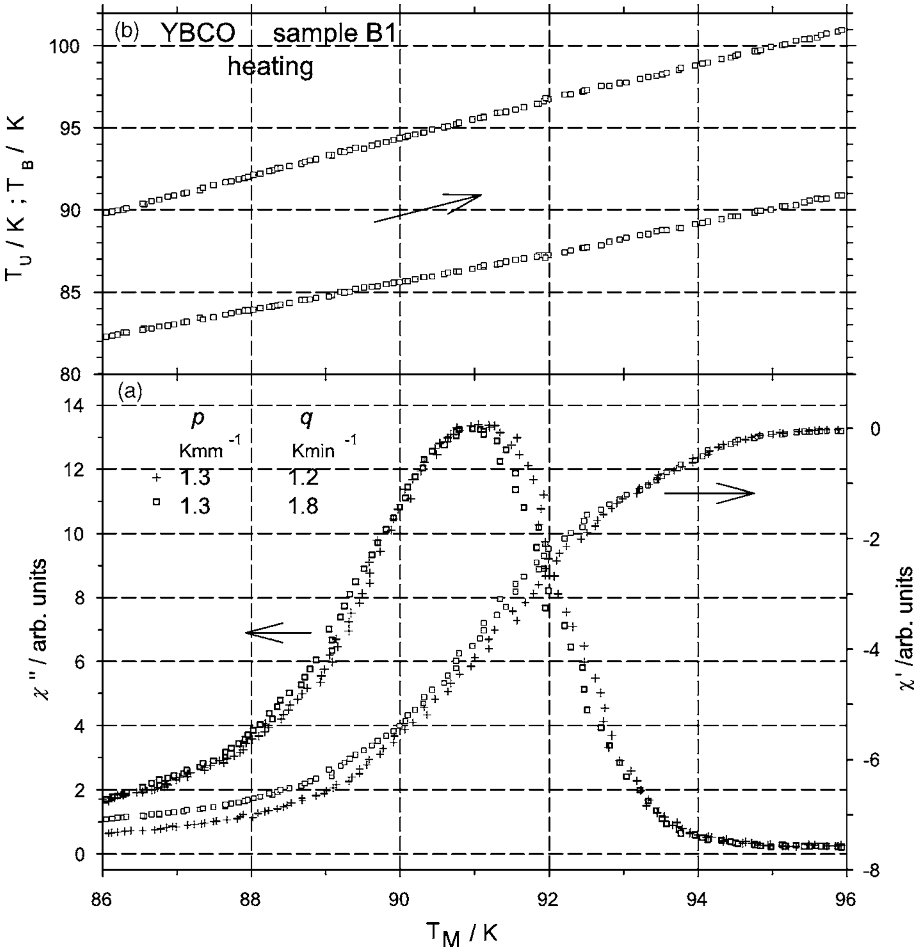

Fig. 3. (a) The χ' and χ'' curves of sample B1 (4 mm × 5 mm × 7 mm) measured by the TSS method, vs. the medium temperature $T_M = (T_U + T_B)/2$ using a commercial lock-in amplifier. (b) The corresponding variation of the temperatures T_U and T_B measured during the TSS measurements by J-type thermocouples at the top and bottom end of the sample B1 vs. the medium temperature T_M.

was slowly submerged in (or raised from) a Dewar containing liquid nitrogen. During this type of cooling (or heating), the heat transport occurs mainly in one direction (quasi-1D temperature gradient). As a result, the boundary plane between the superconducting and normal region sweeps along the sample from the bottom to the top end (or inversely). The temperature accuracy, time and the temperature resolution were 2 K, 1 s, and 0.2 K, respectively. All data were recorded as a function of time using a computer controlled DAS-TC (Keithley) data acquisition card.

Among the advantages of the used TSS method over the conventional one we can mention that it does not permit only the observation of fine structured curves but also the ranging of local phenomena.

3. Results

Fig. 2 presents the results received for the homogeneous A sample, studied by the conventional AC susceptibility method. Fig. 2 also shows the continuous

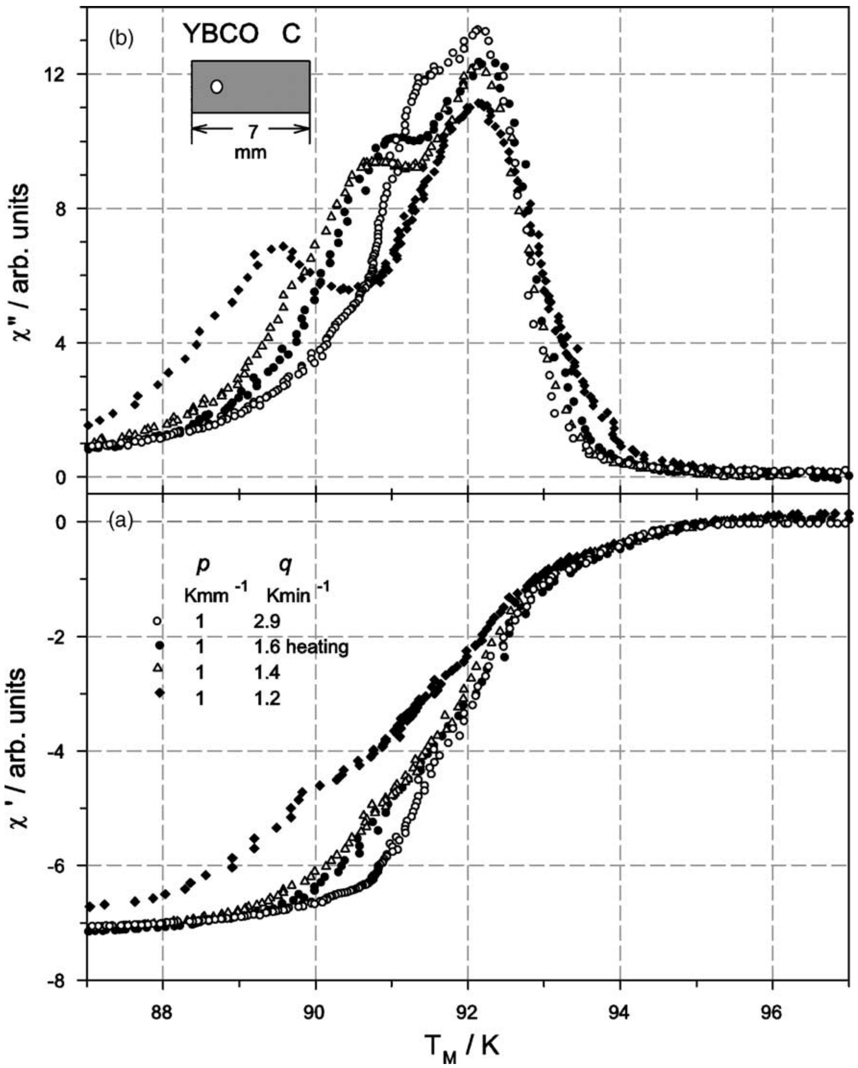

Fig. 4. Effect of different scanning rates ($q = 2.9$, 1.6, 1.4, and 1.2 K min^{-1}) on χ' (a) and on χ'' (b), curves of sample C (4 mm × 5 mm × 7 mm with a hole as local inhomogeneity) obtained at a constant ($p = 1$ K mm^{-1}) temperature gradient vs. the medium temperature T_M.

sigmoid drop of χ' (from 100% to as low as ~20%,) as a function of temperature T. This curve reflects the increase of the ratio of the partial and perfectly coherent fraction to the normal part's volume from 0 to 80%. (That is, the variation of χ' from 0 to as low as approximately -0.8 in dimensionless (SI) units.) The χ' curve begins to deviate from its linear variation at the onset temperature 96 K, when the volume of partial coherent domains becomes high enough to be perceived by the given set-up. The variation of the imaginary component χ'' presents a maximum, reflecting the energy dissipation in the sample. The maximum appeared (at $T_C = 92$ K, and at about 50% coherent volume ratio) when the flux lines and shielding currents fully penetrated the sample. The χ'' peak developed as a compromise between the increase of the absorbed power density and the decrease of the field penetration depth [14].

The χ' and χ'' curves of sample B1 were recorded with the TSS method, under the temperature gradient

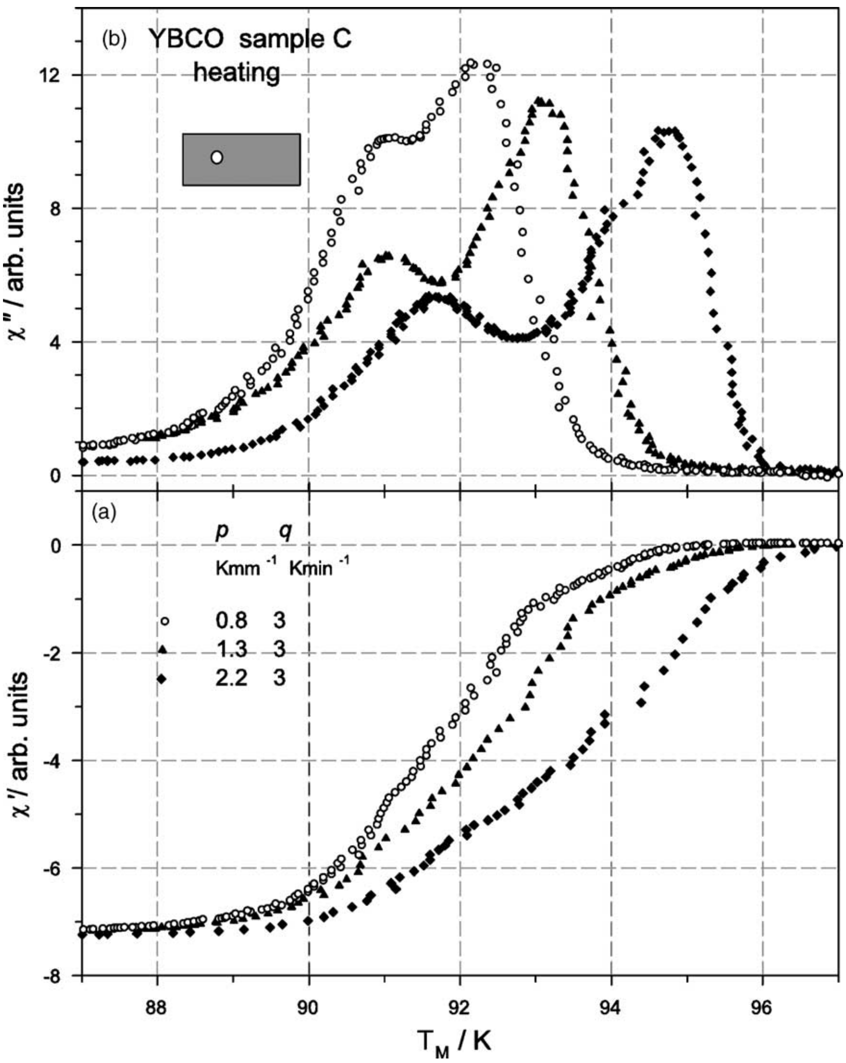

Fig. 5. Effect of different temperature gradients ($p = 0.8$, 1.3, and 2.2 K mm^{-1}) on the χ' (a) and on χ'' (b) curves of the sample C (4 mm × 5 mm × 7 mm with a hole as local inhomogeneity) obtained at a constant scanning rate ($q = 3$ K min^{-1}) vs. the medium temperature T_M.

$p = \Delta T/\Delta x = 1.3 \pm 0.07\,\mathrm{K\,mm^{-1}}$, and presented in function of the medium temperature $T_M = (T_U + T_B)/2$ using two scanning rates ($q = 1.2$ and $1.8\,\mathrm{K\,min^{-1}}$) successively (Fig. 3a). Fig. 3b shows the variation of temperatures T_U and T_B measured by the U and B thermocouples. The main effect of the temperature gradient (scanning) resumes in the broadening of the curves in the direction of lower temperatures.

Comparing the curves of samples A and B1 by curve fitting (using equation type Weinbull for χ'', and sigmoidal for χ' curves) we established that the broadening was not followed by shape distortion. The influence of q on the curves remains negligible.

Sample C was studied with the TSS method. For the shape of the curves χ'' of sample C (with local inhomogeneity) q plays an important role (Fig. 4b). The curves were obtained as the scanning rate varied ($q = 2.9$, 1.6, 1.4, and $1.2\,\mathrm{K\,min^{-1}}$) while maintaining p at a constant $\sim 1\,\mathrm{K\,mm^{-1}}$ value by suitable inclination of the sample's vertical axis. The appearance of a minimum on the χ'' curve (peak-deep-hump structure) reveals the presence of the hole. Slowing down the scanning rate q, the minimum deepens and displaces together with the broadened curves in the direction of lower temperatures. Consequently, with decreasing q the spatial resolution for local inhomogeneities increases, and the main peaks remain at the same (∼92 K) temperature.

Fig. 5a shows the results of sample C obtained with different p at $q = 3\,\mathrm{K\,min^{-1}}$. With increasing gradients ($p = 0.8$, 1.3, and $2.2\,\mathrm{K\,mm^{-1}}$) the χ'' curves broaden in the direction of higher temperatures together with their more and more accentuated peak-deep-hump structures (Fig. 5b).

4. Discussions and conclusions

For homogeneous samples, the TSS results obtained at constant p and q conditions are similar to those obtained under conventional AC susceptometry. The broadening of the curves without distortion—as a consequence of the thermal scanning—increases the spatial resolution of the method for local inhomogeneities.

The (peak-deep-hump structured) curves that are presented here (Figs. 4b and 5b) are distorted by the drilled hole (as artificial local inhomogeneity). They are similar to the χ'' curves of homogeneous materials possessing two critical temperatures. This coincidence may suggest erroneous interpretations. The displacement of peaks in Fig. 5b apparently also may suggest that the critical temperature may depend on the temperature gradient. But our curves reflect particularities produced by not else than the artificial local inhomogeneity but not the temperature dependent variation of the sample's physical and chemical parameters. In order to demonstrate that, we represented in Fig. 6 the maxima of the χ'' curves (reflecting the losses in the superconducting zone situated between the $T_C = 92\,\mathrm{K}$ temperature border-plane and the bottom of the sample) exactly in function of the characterized zone's medium temperature $T_{SC} \approx (T_C + T_B)/2$ (linear interpolation). In the limit of the experimental precision the peaks appear at the same $T_{SC} = 90.15 \pm 0.2\,\mathrm{K}$ temperature. This suggests that the physical and chemical modifications happened always at the same local temperature independently from the applied temperature gradient. That is, the TSS method not altering the course of transition gives objective reproducible results.

Existing mathematical expressions were fitted to the data, but it has not yet yielded a detailed understanding of the structural kinetics. So, the phenomenological interpretation—based on the obtained curves—remains important also in the followings. The distortion on the curves reflects the amplitude and even the location of the particularity.

Before characterization of the studied material the shape of the susceptibility curves must be analyzed in order to separate the effects of eventual local inhomogeneities from the ones produced by structural modifications. For this purpose the χ'' curves—being more affected—are more useful than the χ' ones.

It was confirmed that the spatial resolution of the TSS method is directly proportional to the thermal gradient (p). Under our conditions p must be greater at least five times than $\Delta T/\Delta x = 0.2\,\mathrm{K\,mm^{-1}}$. Here 0.2 K is the temperature resolution and 1 mm is the hole's diameter. The spatial resolution is inversely proportional to the velocity of the heat transfer (q) until the limit $\Delta T/\Delta t = 0.2\,\mathrm{K}/0.8\,\mathrm{min} = 0.25\,\mathrm{K\,min^{-1}}$. Here 0.8 min is the relaxation time observed at normal-superconductor domain fluctuation [9]. At slower scanning rates the spatial resolution of the method for local inhomogeneities decreases. Practically our resolution is better than $\sim 1\,\mathrm{mm^3}$ if the

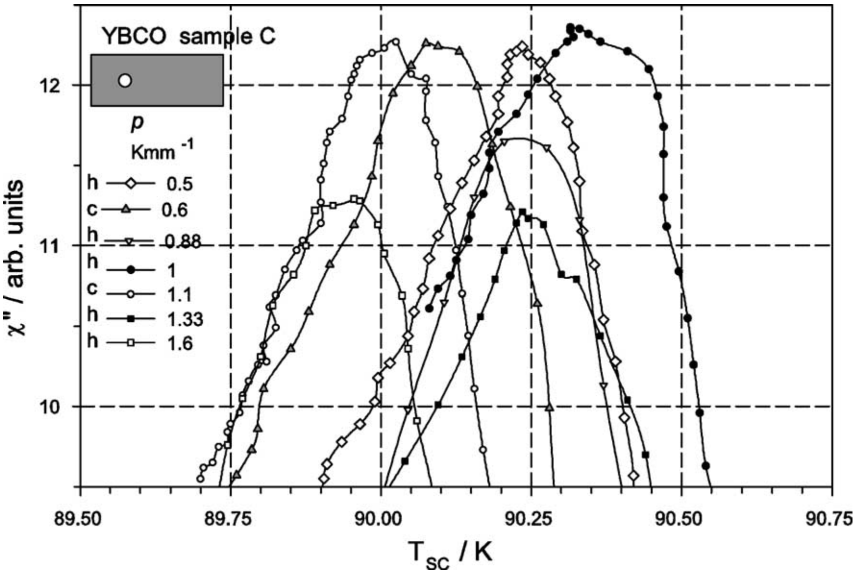

Fig. 6. The peak region of the χ'' curves of sample C for seven different scanning rate vs. T_{SC}, the medium temperature of the superconducting part of the sample (h: heating; c: cooling).

applied temperature gradient becomes greater than $\sim 1\,\mathrm{K\,mm^{-1}}$ and q remains smaller than $\sim 3\,\mathrm{K\,min^{-1}}$. The preceding results allow a better interpretation of the susceptibility curves which can lead to a more clear understanding of the underlying phenomena and characterization of high-T_C superconducting materials.

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