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Study of AC transport processes in RE(Y,Sm,Nd,Dy)BCO HTSC samples in the normal state

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Abstract   : Using a contactless method, the ac response given by $\sigma_2(\omega)/\sigma_1(\omega)$, the ratio of the real (σ_1) and imaginary (σ_2) parts of conductivity, was measured over wide frequency (0 1 to 10 MHz) and temperature (80 to 300 K) ranges for HTSCs(RE)BCO (RE = Y, Sm, Nd, Dy) Analysis of ac response in the normal state clearly indicates that both tunnelling and hopping of carriers are responsible for ac transport and relaxation in these materials A master curve is obtained for each material when $|\sigma_2/\sigma_1|$ is scaled against reduced frequency ω/T^α and the shape of the master curve is similar for all the materials

1. Introduction

Measurement of frequency dependent response of a sample is a well established technique of studying relaxation processes in a solid [1,2]. After the discovery of oxide superconductors with remarkably high transition temperatures by Bednorz and Muller [3] and others [4], we studied the frequency dependent response (5 Hz to 13 MHz) of $YBa_2Cu_3O_7$ at room temperature and liquid nitrogen temperature by using a contactless method developed by us [5] . A suitable cryostatic arrangement, which did not interfere with ac measurements in our contactless method, had to be developed for measurements at other intermediate temperatures and we reported our measurements on YBCO over wide frequency (0.1 to 10 MHz) and temperature (79 to 254 K) ranges [6]. It was suggested there that for ac conduction, in the normal state of YBCO, relaxation occurs through tunnelling and/or hopping of the charge carriers. In order to be more definite about the nature of transport and relaxation processes, we have extended our

measurements to other 123 type rare earth (RE) HTSCs which have transition temperatures (T_c ~ 90 K) similar to YBCO (Y, At. No. 39, electron configuration [Kr] $4d^1 5s^2$). In this paper, we report our measurements of frequency dependent conductivity of YBCO and other (RE)Ba$_2$Cu$_3$O$_7$ [RE = Nd (60,[Xe]$4f^4 6s^2$), Sm (62,[Xe]$4f^6 6s^2$) and Dy (66,[Xe]$4f^{10} 6s^2$)] samples. Instead of fitting an intuitively suggested formula to the data as was done in our previous work [6], we have tried, in this work, to arrive at an appropriate formula giving ω and T dependence from an analysis of the measured data. We are again led to conclude that in the ceramic 123 HTSCs studied ac conduction occurs through both tunnelling and hopping of carriers.

2. Experimental details

(i) *Sample preparation and characterization :*

Cylindrical ceramic samples were prepared by solid state reaction method. The details of the method are given in a previous paper [6].

These samples were characterized by four probe dc resistance measurement (T_c ≈ 89–91 k) using a Janis CCR. The samples were further characterized by X-ray diffraction pattern using Rigaku Miniflex XRD.

(ii) *Experimental method :*

A contactless method [6] was used for measuring frequency dependent conduction. A coil of 15–20 turns were wound (close fitting) around the cylindrical sample at its central region. The ends of the coil were connected to the input terminals of an L.F. impedance analyser HP4192A while the sample was placed in a gas flow cryostat. The temperature was varied from room temperature to about 80 K. At each temperature the frequency of measurement was swept from 0.1 MHz to 13 MHz in steps of 0.1 MHz. At each frequency the measured quantities were R and X where the impedance of the coil is given by $Z = R + iX$.

We define two dimensionless quantities.

$$A(\omega) = \frac{R_{sample}(\omega) - R_{air}(\omega)}{L_0 \omega} \tag{1a}$$

and

$$B(\omega) = \frac{X_{sample}(\omega) - X_{air}(\omega)}{L_0 \omega} \tag{1b}$$

Here the subscript 'air' means 'without sample' and L_0 is the self-inductance of the coil.

Since the measurements were done at high frequencies with samples of low resistivity, a pertinent question is whether the measurements described bulk or surface properties of the samples. The penetration depth δ is given by [7].

with $\mu = 1$

Taking $\rho = 1/\sigma \sim 0.5$ m Ω cm for good samples in the normal state

$\approx 0.5 \times 10^{-3} \times 1/9 \times 10^{-11}$ sec

δ (0.4 MHz) = 2.4 mm

δ (10 MHz) = 0.36 mm

It is thus reasonable to consider the measured properties to be bulk properties. A number of control experiments were done to confirm this.

3. Experimental results

In Figure 1, the values of $A(\omega)$ and $B(\omega)$ for SmBCO have been plotted against frequency ranging from 0.3 MHz to 8 MHz at two temperatures 98 K and 235 K. On this same graph we have also plotted the function graphs of

$$A(\omega) = \sum_{i=1}^{5} C_i \frac{\omega \tau_i}{1 + \omega^2 \tau_i^2} \qquad (2a)$$

and

$$B(\omega) = \sum_{i=1}^{5} C_i \left[\frac{1}{1 + \omega^2 \tau_i^2} - 1 \right] \qquad (2b)$$

where the major relaxation times τ_i (i = 1 to 5) were obtained from the plot of $|B(\omega)|$ vs. $A(\omega)/\omega$. The procedure for obtaining τ_i and C_i has been described in Ref. [5]. The excellent fit of the experimental points with the function plot suggests that our system is consistent with the Debye model so far as ac transport is considered in the normal state of these samples. Results are similar for other (RE)BCO samples reported in this paper.
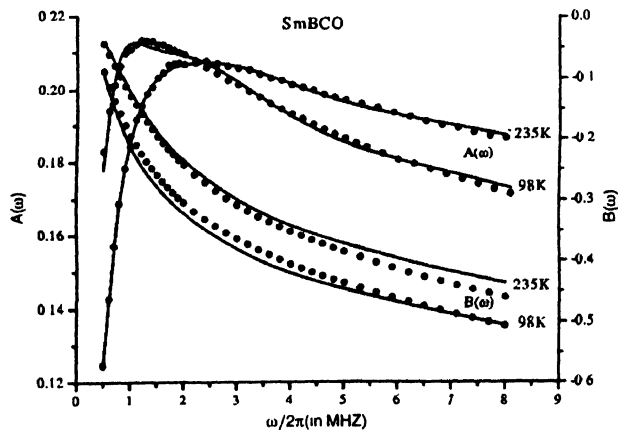
Figure 1. $A(\omega)$ and $B(\omega)$ vs. $\omega/2\pi$ (in MHz) for SmBCO at 98 K and 235 K. The solid lines are function graphs of eq. 2(a) for $A(\omega)$ and eq. 2(b) for $B(\omega)$.

As has been shown in the Appendix, $A(\omega)$ is proportional to $\varepsilon_2(\omega)$ Thus our method gives a simple way of finding how $\varepsilon_2(\omega)$ varies with ω. It is known that ac

dielectric loss starts at ω_{max}, the frequency at which $\varepsilon_2(\omega)$ (or $A(\omega)$), becomes maximum In case of SmBCO, for example, $\omega_{max}/2\pi$ is in the range 1 5 to 2 5 MHz increasing approximately linearly with temperature

The quantities $A(\omega)$ and $B(\omega)$ at a particular temperature T are related to the real and imaginary parts of conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ (see Appendix) It has been shown that the ratio $|B(\omega)/A(\omega)|$ gives $|\sigma_2(\omega)/\sigma_1(\omega)|$ unambiguously In fact

$$|B(\omega)/A(\omega)|_T = |\sigma_2(\omega)/\sigma_1(\omega)|_T = X(\omega, T) \text{ (say)} \tag{3}$$

We next consider the frequency and temperature dependence of $|\sigma_2/\sigma_1|$ ω and T dependence of $|\sigma_2/\sigma_1|$

The raw data of $|B(\omega)/A(\omega)|$ for SmBCO are plotted in Figure 2 as a function of T at different fixed frequencies $\omega/2\pi$ It is seen from inspection that $|B(\omega)/A(\omega)|$ increases monotonically with ω at fixed T and decreases monotonously with T at fixed ω The trend is similar for other samples studied
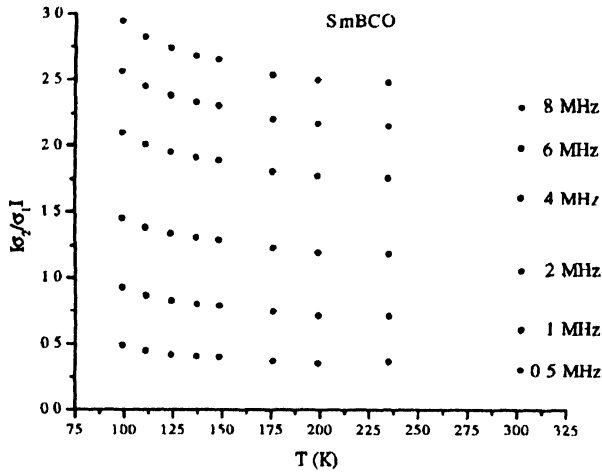

Figure 2. Plot of $|B(\omega)/A(\omega)| = |\sigma_2/\sigma_1|$ vs T at different $\omega/2\pi$ $|\sigma_2/\sigma_1|$ monotonically decreases with T at fixed $\omega/2\pi$ and increases with $\omega/2\pi$ at fixed T

To find the temperature dependence of $|\sigma_2/\sigma_1|$ we consider $T|\sigma_2/\sigma_1|$ vs T at fixed ω's and examine linear (which gives $|\sigma_2/\sigma_1| = A'/T + B'$) and polynomial (which gives $|\sigma_2/\sigma_1| = A'/T + B_1 + B_2T$) fits and consider $T|\sigma_2/\sigma_1|$ vs T^2 and examine linear fit (which gives $|\sigma_2/\sigma_1| = A'/T + B'T$)

It is seen, by examining χ^2 in each case, that the relation

$$|\sigma_2/\sigma_1| = A'/T + B_1 + B_2T \tag{4}$$

best represents the data

Various pair approximation type (PAT) models [8–10] and random walk type [RWT] models [11–13] have been applied to account for temperature and frequency dependence of ac conduction in various systems, particularly amorphous and disordered systems Our samples are polycrystalline ceramic HTSCs

We first show that if small polaron tunneling (SPT) and correlated barrier hopping (CBH) processes [6] occur simultaneously and independently then $|\sigma_2/\sigma_1|$ must have the form given by eq. (4).

Taking linear combinations of eqs (23) and (24) in Ref. [6], we can write $\left|\dfrac{\sigma_2}{\sigma_1}\right| = C_1\left[\ln\omega\tau_0\left(1+\dfrac{A_1}{T\ln\omega\tau_0}\right)\right] + C_2\left[\ln\omega\tau_0\left(1+A_2T\ln\omega\tau_0\right)\right]$, where we have assumed τ_0 to be the same for both the processes.

$$\left|\frac{\sigma_2}{\sigma_1}\right| = (C_1+C_2)\ln(\omega\tau_0)+\frac{C_1A_1}{T}+(C_2A_2)T[\ln(\omega\tau_0)]^2 = a\ln(\omega\tau_0)+\frac{b}{T}+cT[\ln(\omega\tau_0)]^2 \quad (5)$$

Eq (5) has the same temperature dependence as eq. (4)

In order that eq. (5) represent our experimental data the following conditions must hold, namely,

$$A'(\omega) = b \quad (6a)$$

$$B_1(\omega) = a\ln(\omega\tau_0) = a\ln\omega + a\ln\tau_0 \quad (6b)$$

$$B_2(\omega) = c[\ln(\omega\tau_0)]^2 \quad (6c)$$

In Figure 3 we plot A' (ω) against $\omega/2\pi$ We find that A'(ω) varies linearly with ω Writing A'(ω) = $b(\omega)$ = $b_0(1 + \beta\omega)$, we determine b_0 and β for all the samples
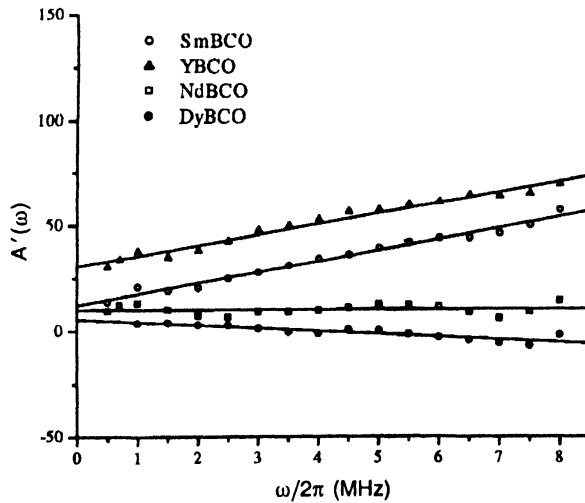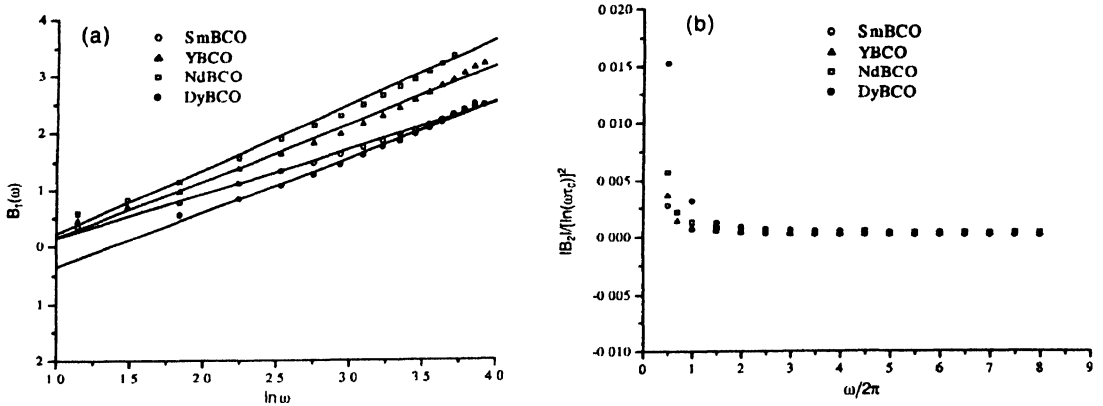

Figure 3. A'(ω) *vs* $\omega/2\pi$ plot for Sm (o), Y(▲), Nd (□) and Dy (•) based 123 HTSCs

We note that if eq. 6(b) is true then $B_1(\omega)$ should be a linear function of $\ln\omega$. In Figure 4(a) we have plotted $B_1(\omega)$ against $\ln\omega$ for all the samples and find that $B_1(\omega)$ varies linearly with $\ln\omega$. From the linear fit we get a and τ_0 for all the samples

In Figure 4(b), we plot $\dfrac{|B_2(\omega)|}{[\ln(\omega\tau_0)]^2} = c(\omega)$ with $\omega/2\pi$. $c(\omega)$ can be given by the

Figure 4. (a) $B_1(\omega)$ vs ln ω and (b) $|B_2(\omega)|/[\ln(\omega\tau_0)]^2$ vs $\omega/2\pi$ for Sm(o), Y (▲), Nd (□) and Dy (•) based 123 HTSCs

relation $c(\omega) = \eta/\omega$, the parameter η being determined from the fit.

Since in all PAT theories σ_2/σ_1 is always proportional to $[-\ln(\omega\tau_0)]$ where τ_0 is some cut-off relaxation time we cast eq (5) in a form in which $\ln(\omega\tau_0)$ has been factored out, i.e.,

$$\left|\frac{\sigma_2}{\sigma_1}\right| = a\ln(\omega\tau_0)\left[1+\frac{b}{a}\frac{1}{T}\frac{1}{\ln(\omega\tau_0)}+\frac{c}{a}T\ln(\omega\tau_0)\right] \tag{7}$$

We summarize in Table 1 the parameters a, τ_0, b_0, β, η for all the four HTSCs. In Figure 5, we fit eq. (7) (unbroken line) to the experimental data [given as •] for SmBCO.

We therefore suggest that the ac conduction in these HTSCs occurs through both tunnelling and hopping of carriers, hopping being the predominant mechanism at higher temperatures and frequencies.

*Master curve and ac universality :*

In many amorphous semiconductors, ionic conductors and polycrystalline substances ac conductivity has the form

$$\sigma(\omega) = A(T)\omega^s \tag{8}$$

at high frequencies, where the exponent s is generally less than or equal to 1 and at low frequencies $\sigma(\omega)$ is frequency independent.

Table 1. Values of the parameters a, τ_0, b_0, β, η for all the four HTSCs.

Sample	a	τ_0 (μ sec)	b_0	β	η
SmBCO	0 77026	0 4476	12.3302	0.0679	0.0048
YBCO	0.98087	0.4339	30.7515	0.0262	0.0055
NdBCO	1.1120	0.4534	10.1958	0.0013	0.0096
DyBCO	0 79155	0.3726	5.5994	−0.0379	0.0120
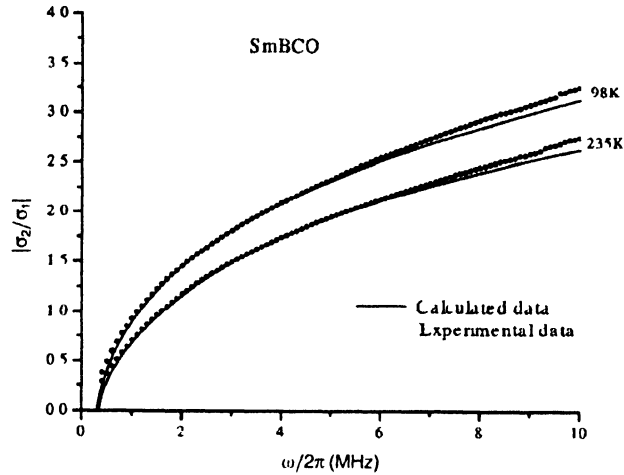
Figure 5. Plot of $|B(\omega)/A(\omega)| = |\sigma_2/\sigma_1|$ *vs* $\omega/2\pi$ for 98 and 235 K The unbroken lines through the exptal points (•) are due to eq (7)

It is usually possible to scale ac data at different temperatures for one solid into one single curve This so called **master curve** gives the dimensionless ac conductivity $\bar{\sigma} = (\sigma(\omega)/\sigma(0))$ as a function of dimensionless frequency $\omega = (\omega/\omega_m)$ The existence of such a curve is referred to as the 'time temperature superposition principle' (TTSP) The shape of the master curve is roughly the same for all disordered solids This is known as **ac universality** [14–17]

We have tried to find whether an appropriate scaling can give a master curve for $|\sigma_2/\sigma_1|$ If the dimensionless quantity $|\sigma_2/\sigma_1|$ is plotted against the dimensionless frequency ω/ω_m the data at different temperatures for one solid do not give a single curve However if we consider $|\sigma_2/\sigma_1|$ as a function $X(x)$ of reduced frequency $x = (\omega/T^\alpha)$ where α is characteristic of the solid we obtain a master curve

In Figures 6(a), 6(b), 6(c) and 6(d) we present respectively the master curves for SmBCO, YBCO, NdBCO, DyBCO In each case the master curve can be represented by a formula

$$X(x) = A_0 x^p \tag{9}$$

We note that $X(x)$ is a homogeneous function of x since

$$X(\lambda x) = \lambda^p X(x) \tag{10}$$

The scaling behaviour is similar for all the HTSCs studied In Table 2 we collect the values of α, p and A_0 for the materials

We are not aware of any theoretical model which predicts such a master curve for $|\sigma_2/\sigma_1|$ and identical scaling behaviour for polycrystalline ceramic 123 type HTSCs in their normal state
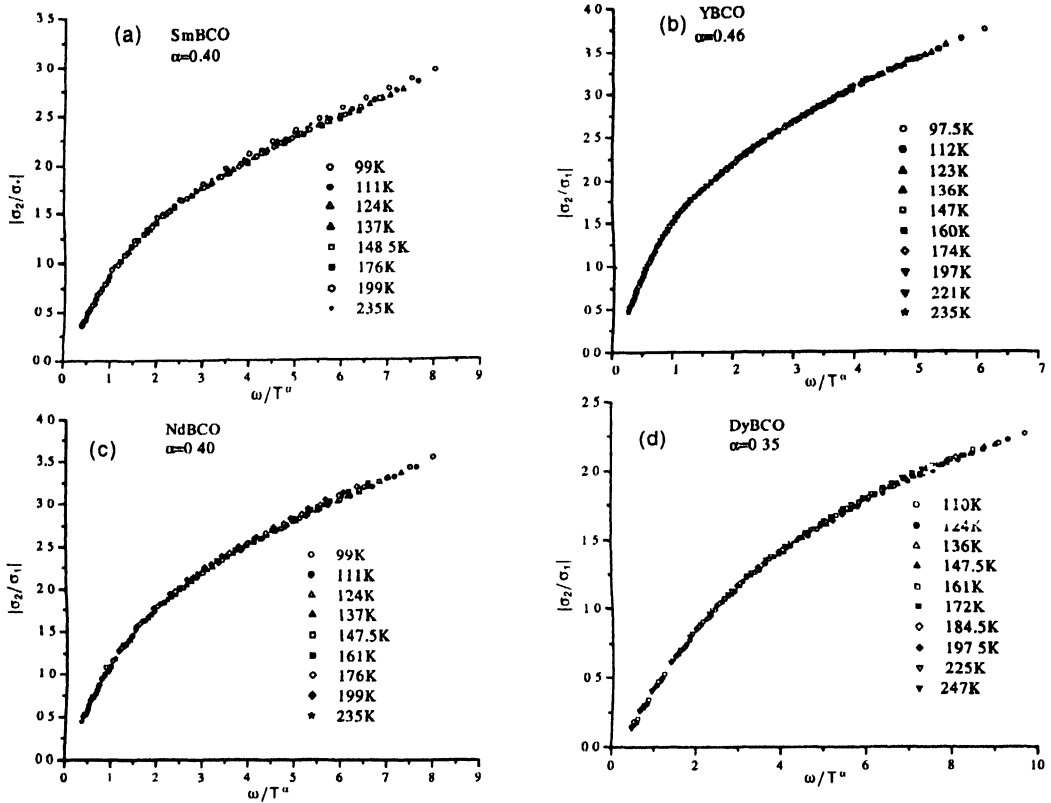
Figure 6. Plots of $|\sigma_2/\sigma_1|$ *vs.* ω/T^α for (a) SmBCO, $\alpha = 0.40$, (b) YBCO, $\alpha = 0.46$, (c) NdBCO, $\alpha = 0.40$ and (d) DyBCO, $\alpha = 0.35$ These master curves incorporating data from wide frequency and temperature ranges are quite impressive.

Table 2. Values of α, p and A_0 for different materials.

Sample	α	p	A_0
SmBCO	0.40	0.52967	0.9757
YBCO	0.46	0.50155	1.5274
NdBCO	0.40	0.50716	1.2433
DyBCO	0.35	0.64858	0.55501

4. Discussion

We chose to represent the experimental data as a ratio σ_2/σ_1 of the imaginary and real parts of ac conductivity $\sigma(\omega)$ as this ratio is directly related to $\left[-\dfrac{B(\omega)}{A(\omega)} \right]$. The reason for representing data in terms of the components of conductivity $\sigma(\omega)$ is two fold. Firstly, $\sigma(\omega)$ is directly related to equilibrium current-current fluctuation through the Kubo formula

$$\sigma(\omega) = \frac{1}{3k_b T V} \int_0^\infty <J(0).J(t)> e^{-i\omega t} dt$$

where J is the total current in volume V. Kubo formula may be the starting point for any theoretical treatment of $\sigma(\omega)$. Secondly, $\sigma_1(\omega)/2$ times the absolute square of the current density gives the dissipation per unit volume per unit time. The ratio σ_2/σ_1 gives the extent of dispersion *vis-a-vis* dissipation. In the present paper we analysed the experimental data step by step and arrived at a form (giving ω and T dependence) involving only five frequency and temperature independent parameters. This equation (eq. (7)) with only five parameters represent the vast amount of experimental data extremely well. It is, however, more intriguing to discover that the same data can be represented by a master curve when $|\sigma_2/\sigma_1|$ is scaled against ω/T^α, α being characteristic of a particular material. In a wide temperature and frequency range the data can be represented by a homogenous equation involving only three parameters. The existence of the master curve and the universality in its shape can not be related to disorder since the HTSCs we are dealing with, are polycrystalline (composed of granular crystals). In going from grain to grain, however, there is a random change in the orientation of the crystal axes of the grains. We suggest that this element of randomness is responsible for the existence of such a master curve and its universality. The dichotomy between a five parameter equation suggesting tunnelling and hopping as transport mechanism and a three parameter homogenous equation describing a master curve can be explained by noting that the former describes the microscopic aspect of transport within the crystalline grains and the latter is a manifestation of randomness of orientation of the grain crystals on a macroscopic scale.

Acknowledgment

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Appendix

$$Z = R + iX = \frac{1}{|I_i|^2}\left[\int (J^* \cdot E)d^3x + 4i\omega\int(w_e - w_m)d^3x + 2\int_{s-s_i}(s \cdot n)da\right]$$

$$= R_{coil} + i\omega(\varepsilon^* C_e - \mu C_m) + \frac{1}{|I_i|^2}2\int(s \cdot n)da$$

Therefore, $Z_{sample}(\omega) - Z_{air}(\omega) = i\omega\left[(\varepsilon^*_{sample}(\omega) - \varepsilon^*_{air}(\omega))C_e - (\mu_{sample}(\omega) - \mu_{air}(\omega))C_m\right]$
In the normal state of the sample

$$\mu_{sample} = \mu_{air} = 1 \text{ for non-ferromagnetic substances}$$

$$\varepsilon^*_{sample} = \varepsilon_1 - i\varepsilon_2$$

$$\varepsilon^*_{air} = 1$$

So,

$$Z_{sample}(\omega) - Z_{air}(\omega) = i\omega (\varepsilon_1 - i\varepsilon_2 - 1)C_e$$

$$= \omega\varepsilon_2(\omega) C_e + i\omega (\varepsilon_1(\omega) - 1)C_e$$

$$\therefore A(\omega) = \frac{(R_{sample}(\omega) - R_{air}(\omega))}{L_0\omega} = \frac{C_e}{L_0}\varepsilon_2(\omega) = \frac{4\pi c C_e}{L_0\omega}\sigma_1(\omega)$$

$$B(\omega) = \frac{(X_{sample}(\omega) - X_{air}(\omega))}{L_0\omega} = \frac{C_e}{L_0}(\varepsilon_1(\omega) - 1) = -\frac{4\pi c C_e}{L_0\omega}\sigma_2(\omega)$$

Therefore, $\dfrac{B(\omega)}{A(\omega)} = -\dfrac{\sigma_2(\omega)}{\sigma_1(\omega)}$

The relation $\dfrac{B(\omega)}{A(\omega)}$ gives $\dfrac{\sigma_2(\omega)}{\sigma_1(\omega)}$ unambiguously in the normal state of the sample.