Chapter 2: Background
2.1 Identification of polaronic character of carriers by measuring $\sigma_2/\sigma_1$

The study of frequency dependent loss gives much information about the relaxation processes in solids. This study involves measurements of the real $\sigma_1(\omega)$ and the imaginary part $\sigma_2(\omega)$ of the complex conductivity at different frequency regions. The ac conductivity components $\sigma_1(\omega)$ and $\sigma_2(\omega)$ are related to each other via Kramers-Krönig relation. These parameters can also be measured separately by many experiments. Long [1] has derived specific expressions for $\sigma_1$, $\sigma_2$ and the ratio, $\sigma_2/\sigma_1$ as a function of frequency and temperature for polaron hopping and tunneling. To verify the validity of the proposed model of polaronic transport in HTSC cuprates measurement of ac conductivity of such samples at different temperature and frequency has been carried out. The measured data are expected to follow the same frequency and temperature dependence as estimated by Long. Accurate measurement of $\sigma_1$ and $\sigma_2$ separately requires precession geometric construction of the setup whereas $\sigma_2/\sigma_1$ can be obtained with less precession in geometry. By the method developed in this laboratory $\sigma_2/\sigma_1$ is measured in a contact less method. Of course this method can not be used for obtaining $\sigma_1$ and $\sigma_2$ separately. In the following sections measured data of $\sigma_2/\sigma_1$ were analyzed for polaronic characteristic.

2.2 Our contact less method to obtain the ratio $\sigma_2/\sigma_1$

A radiating system carrying current appears as a load having impedance with a resistive and a reactive part. This is true even if the radiating system is made in the form of a simple coil of a conducting wire. In our contact less method, ac signals are sent to an insulated copper coil with the high Tc samples as its core. Thus from the impedance ($Z$) of the coil with and without core, the ratio $(\sigma_2/\sigma_1)$ of the imaginary and the real parts of the complex conductivity of the core material can be calculated by virtue of the well known Complex Poynting Theorem [2]. Here we have assigned two dimensionless quantities to elevate the core effect.

Assuming the field-source time dependence $e^{-i\omega t}$, electric field can be taken in the complex form,

$$\vec{E}(\vec{x}, t) = \text{Re}[\vec{E}(\vec{x})e^{-i\omega t}]$$
For a single charge $q$, the power radiated by external electromagnetic fields $\vec{E}$ and $\vec{B}$ is $q\vec{v} \cdot \vec{E}$ where, $\vec{v}$ is the velocity of the charge. Here the magnetic field contribution is nil since the magnetic force is perpendicular to the velocity. For a continuous distribution of charge and current, the total rate of doing work by the fields in a finite volume $V$ is given by,

$$\int_V \vec{J} \cdot \vec{E} d^3 \vec{x} \quad (2.2)$$

For harmonic variation, the product $\vec{J}(\vec{x},t) \cdot \vec{E}(\vec{x},t)$ can be written as,

$$\vec{J}(\vec{x},t) \cdot \vec{E}(\vec{x},t) = \frac{1}{2} [\vec{J}(\vec{x})e^{-iat} + \vec{J}^*(\vec{x})e^{iat}][\vec{E}(\vec{x})e^{-iat} + \vec{E}^*(\vec{x})e^{iat}]$$

$$= \frac{1}{2} [\vec{J}(\vec{x}).\vec{E}(\vec{x})e^{-2iat} + \vec{J}(\vec{x}).\vec{E}^*(\vec{x}) + \vec{J}^*(\vec{x}).\vec{E}(\vec{x}) + \vec{J}^*(\vec{x}).\vec{E}^*(\vec{x})e^{2iat}]$$

$$= \frac{1}{2} \text{Re}[\vec{J}^*(\vec{x}).\vec{E}(\vec{x}) + \vec{J}(\vec{x}).\vec{E}(\vec{x})e^{-2iat}] \quad (2.3)$$

Since,

$$\frac{\vec{A} + \vec{A}^*}{2} = \text{Re}[\vec{A}]$$

So,

$$\langle \int_V \vec{J}(\vec{x},t) \cdot \vec{E}(\vec{x},t) d^3 \vec{x} \rangle_{\text{timeaverage}} = \frac{1}{2} \text{Re} \int_V \vec{J}^*(\vec{x}).\vec{E}(\vec{x}) d^3 \vec{x} \quad (2.4)$$

$$= \frac{1}{2} \text{Re} \int_V [\vec{J}^*(\vec{x}).\vec{E}(\vec{x}) + \vec{J}(\vec{x}).\vec{E}(\vec{x})e^{-2iat}] d^3 \vec{x} \rangle_{\text{timeaverage}}$$

For time averages of products, the convention is, therefore, to take one-half of the real part of the product of one complex quantity with the complex conjugate of the other.

The Maxwell’s electromagnetic field equations for harmonic fields are,

\[
\nabla \cdot \vec{D} = 0 \\
\n\nabla \times \vec{B} = 4\pi \rho \quad (2.5)
\]
\[ \nabla \times \mathbf{H} = \frac{4\pi}{c} \mathbf{j} + \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t} = \frac{4\pi}{c} \mathbf{j} - \frac{i\omega}{c} \mathbf{D} \]
\[ \nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} = \frac{i\omega}{c} \mathbf{B} \]

Now,

\[ \frac{1}{2} \int \mathbf{J} \cdot \mathbf{E} d^3 \mathbf{x} = \frac{c}{8\pi} \int \mathbf{E} \cdot \left[ \nabla \times \mathbf{H}^* - \frac{i\omega}{c} \mathbf{D}^* \right] d^3 \mathbf{x} \]
\[ = \frac{c}{8\pi} \int \mathbf{E} \cdot \left( \nabla \times \mathbf{H}^* \right) d^3 \mathbf{x} - \frac{i\omega}{c} \mathbf{E} \cdot \mathbf{D}^* d^3 \mathbf{x} \]
\[ = \frac{c}{8\pi} \int \left( \nabla \cdot (\mathbf{E} \times \mathbf{H}) - \mathbf{E} \cdot (\mathbf{E} \times \mathbf{H}^*) - \frac{i\omega}{c} \mathbf{E} \cdot \mathbf{D}^* \right) d^3 \mathbf{x} \]
\[ = \frac{c}{8\pi} \int \left( -\mathbf{E} \cdot (\mathbf{E} \times \mathbf{H}) + \frac{i\omega}{c} \mathbf{H} \cdot \mathbf{B} - \frac{i\omega}{c} \mathbf{E} \cdot \mathbf{D}^* \right) d^3 \mathbf{x} \]

The complex Poynting Vector is defined as [2]

\[ \mathcal{S} = \frac{c}{8\pi} (\mathbf{E} \times \mathbf{H}^*) \] (2.7)

and the harmonic electric and magnetic energy densities are given by

\[ w_e = \frac{1}{16\pi} (\mathbf{E} \cdot \mathbf{D}^*), \quad w_m = \frac{1}{16\pi} (\mathbf{B} \cdot \mathbf{H}^*) \] (2.8)

Therefore, equation (2.6) can be written as

\[ \frac{1}{2} \int \mathbf{J} \cdot \mathbf{E} d^3 \mathbf{x} = -\frac{1}{8\pi} \int \mathbf{E} \cdot d^3 \mathbf{x} + 2i\omega \int (w_m - w_e) d^3 \mathbf{x} \]
\[ \Rightarrow \frac{1}{2} \int \mathbf{J} \cdot \mathbf{E} d^3 \mathbf{x} + 2i\omega \int (w_e - w_m) d^3 \mathbf{x} + \oint \mathcal{S} \cdot \mathbf{n} d\alpha = 0 \] (2.9)

The real part of left hand side gives the conservation of energy for the time averaged quantities, and the imaginary part relates to the reactive or stored energy.
If the energy densities $w_e$ and $w_m$ have real volume integrals (i.e. $\int_V (w_e - w_m) d^3\vec{x}$ is real) as occurs for the systems with lossless dielectrics and perfect conductors, the real part of equation (2.9) is given as:

$$\frac{1}{2} \int_V \text{Re}(\vec{J} \cdot \vec{E}) d^3\vec{x} + \oint_s \text{Re}(\vec{S} \cdot \vec{n}) da = 0$$

(2.10)

The first term is the steady state time average rate of doing work on the sources in volume, $V$ by the fields. Equation (2.10) shows that this is equal to the average flow of power into the volume, $V$ through the boundary surface $S$ as calculated from the normal component of $\text{Re}(\vec{S})$.

The complex Poynting Theorem (2.9) can be used to define the input impedance of a general two terminal, linear, passive electromagnetic system. Let the system in the volume $V$ is to be surrounded by the protruding, as shown in Figure 2.1 [1]. If the complex harmonic input current and voltage are $I_i$ and $V_i$, the complex power input is $\frac{1}{2} I_i^* V_i$.

![Figure 2.1 Schematic diagram of a simple EM radiating system](image-url)
This can be written in terms of the Poynting Vector by using equation (2.9) applied to all of space on the outside of \( S \) as

\[
\frac{1}{2} I_i^* V_i = -\oint_S \vec{S} \cdot \vec{n} \, da
\]  

(2.11)

where the unit normal \( \vec{n} \) is outwardly directed as shown in Figure 2.1, and it has been assumed that the input power flow is confined to the area \( S_i \). Using equation (2.9), the right hand side of equation (2.11) can be written as

\[
\frac{1}{2} I_i^* V_i = \frac{1}{2} \int_{V} \vec{J}^* \cdot \vec{E} d^3 \vec{x} + 2i\omega \int_{V} (w_e - w_m) d^3 \vec{x} + \oint_{S-S_i} \vec{S} \cdot \vec{n} \, da
\]  

(2.12)

The surface integral in equation (2.12) represents power flow out of the volume \( V \) through the surface \( S \) except for the input surface \( S_i \).

The input impedance \( Z=R+jX \) follows from equation (2.12) with the definition of \( Z \) given by \( V_i = Z I_i \)

Now,

\[
\frac{1}{2} I_i^* V_i = \frac{1}{2} Z |I_i|^2
\]

So, after Jackson (1978), \( Z=R+jX \)

\[
= \frac{1}{|I_i|^2} \left[ \int_{V} \vec{J}^* \cdot \vec{E} d^3 \vec{x} + 4j\omega \int_{V} (w_e - w_m) d^3 \vec{x} + 2 \oint_{S-S_i} \vec{S} \cdot \vec{n} \, da \right]
\]  

(2.13)

Equation (2.13) is to be applied to the actual experimental arrangement used as shown in Figure 2.2. A closely wound coil \( C \) of enameled copper wire is fed with voltage \( V_i \), and a current \( I_i \) flows in through the input terminals A and B. The material under test in the form of a cylinder of length \( L \) is slipped through the coil of width \( W \) (\( W<<L \)). The impedance across AB is measured with and without the sample. These are designated as \( Z_{\text{sample}} \) and \( Z_{\text{air}} \) respectively.
In applying equation (2.13) it is assumed that the power flow out through (S-Sj) is real and it remains substantially unchanged with or without the sample. This is justified since power flowing out will be very small unless the frequency is very high.

Since $\vec{J}$ in the first term in equation (2.13) will be confined within the copper wire because of its high conductivity and its enamel coating, the first term should be equal to the ohmic resistance $R_0$ of the coil. Unless the frequency is very high, the contribution of Skin effect to $R_0$ will be negligible.

Thus it is to note that

(i) \[ \frac{1}{|I_j|^2} \int \vec{J}^* \cdot \vec{E} d^3\vec{x} = R_0 = R_{oc} \text{ (say)} \]

(This, of course, depends on temperature)

(ii) The second term yields

\[ \frac{4}{|I_j|^2} \int w_v d^3\vec{x} = \frac{1}{|I_j|^2} \frac{1}{4\pi} \int (\vec{E} \cdot \vec{D}^*) d^3\vec{x} \]

\[ = \frac{\varepsilon^*}{|I_j|^2} \frac{1}{4\pi} \int |\vec{E}|^2 d^3\vec{x} \]

\[ = \varepsilon^* C_e \text{ (say)} \]

where, $C_e$ is a geometrical factor
\[ \frac{4}{|I|^2} \int \omega n d^3 \vec{x} = \frac{1}{|I|^2 4\pi} \int (\vec{B} \cdot \vec{H}^*) d^3 \vec{x} \]
\[ = \frac{\mu}{|I|^2 4\pi} \int \vec{H}^2 d^3 \vec{x} \]

where, \( C_m \) is a geometrical factor.

(iii) The third term is the radiation loss term and may be taken to be the same with
\[ = \mu C_m \text{(say)} \]
or without the sample, as the temperature remains constant.

Thus,
\[ Z_{\text{sample}} - Z_{\text{air}} = i\omega \left( (\varepsilon_{\text{sample}}^* - \varepsilon_{\text{air}}^*) C_e - (\mu_{\text{sample}} - \mu_{\text{air}}) C_m \right) \quad (2.14) \]

Two non-dimensional quantities are used in this study after Nag et al. [3] as given in the following:

\[ A(\omega) = \frac{(R_{\text{sample}} - R_{\text{air}})}{\omega L_0} \quad (2.15) \]

and

\[ B(\omega) = \frac{(Z_{\text{sample}} - Z_{\text{air}})}{\omega L_0} \quad (2.16) \]

Where, \( L_0 \) is the self inductance of the coil.

It is also known that

\[ A(\omega) + iB(\omega) = \frac{1}{\omega L_0} [Z_{\text{sample}} - Z_{\text{air}}] \]
\[ = \frac{i\omega}{\omega L_0} [(\varepsilon_{\text{sample}}^* - \varepsilon_{\text{air}}^*) C_e - (\mu_{\text{sample}} - \mu_{\text{air}}) C_m] \]
\[ = \frac{i}{L_0} [(\varepsilon_{\text{sample}}^* - \varepsilon_{\text{air}}^*) C_e - (\mu_{\text{sample}} - \mu_{\text{air}}) C_m] \quad (2.17) \]

In the normal state of the sample:
\[ \mu_{\text{sample}} = \mu_{\text{air}} = 1 \text{ for non-ferromagnetic substances and } \varepsilon_{\text{air}}^* = \varepsilon_{\text{air}} = 1. \]
So

\[ Z_{\text{sample}}(\omega) - Z_{\text{air}}(\omega) = i\omega(\varepsilon' - 1)C_\varepsilon \]

\[ = i\omega(\varepsilon_1 - i\varepsilon_2 - 1)C_\varepsilon \]

\[ = \omega\varepsilon_2 C_\varepsilon + i\omega(\varepsilon_1 - 1)C_\varepsilon \]

Then the equation (s) takes the form as

\[ A(\omega) + iB(\omega) = \frac{1}{\omega L_0}[\omega\varepsilon_2 C_\varepsilon + i\omega(\varepsilon_1 - 1)C_\varepsilon] \]

Equating real and imaginary parts, the form of \( A(\omega) \) and \( B(\omega) \) can be written as

\[ A(\omega) = \varepsilon_2(\omega) \frac{C_\varepsilon}{L_0} \]

and

\[ B(\omega) = [\varepsilon_1(\omega) - 1] \frac{C_\varepsilon}{L_0} \]

Since

\[ \varepsilon(\omega) = 1 + i\frac{4\pi}{\omega} \sigma(\omega) \]

\[ \varepsilon_1(\omega) + i\varepsilon_2(\omega) = 1 + i\frac{4\pi}{\omega} [\sigma_1(\omega) + i\sigma_2(\omega)] \]

\[ = 1 + i\frac{4\pi}{\omega} \sigma_1(\omega) - \frac{4\pi}{\omega} \sigma_2(\omega) \]

So,

\[ \varepsilon_1(\omega) - 1 = -\frac{4\pi}{\omega} \sigma_2(\omega) \]

and

\[ \varepsilon_2(\omega) = \frac{4\pi}{\omega} \sigma_1(\omega) \]

For the normal state of the sample,

\[ \frac{B(\omega)}{A(\omega)} = \frac{[\varepsilon_1(\omega) - 1]}{\varepsilon_2(\omega)} = -\frac{4\pi\sigma_2(\omega)}{4\pi\sigma_1(\omega)} = -\frac{\sigma_2(\omega)}{\sigma_1(\omega)} \]

Or

\[ \left|\frac{B(\omega)}{A(\omega)}\right| = \left|\frac{\sigma_2(\omega)}{\sigma_1(\omega)}\right| \]

(2.18)
So the ratio of $B(\omega)$ and $A(\omega)$ gives directly the ratio of $\sigma_2/\sigma_1$ in the normal state.

2.3 Scaling as a tool and different ideas of scaling

The scaling is a typical analytical procedure for interpreting data including different parameters in disordered system where the plots of any parameter at different conditions e.g. temperatures, magnetic field etc, may be made to collapse on a single curve by using a reduced form of the parameter as a variable. This single curve is called master curve and the process is called scaling.

According to Sidebottom [4], the ability to scale different data plots to a common curve indicates that the process has a common physical mechanism. Scaling could be done in several systems, namely ionic and electronic conduction in glass and similar disordered material Dyre et al [5], gas-liquid coexistence in molecular fluid [6] superfluid density in high temperature superconductors [7], pinning forces in ceramic cuprate superconductors [8]. The common feature in all these systems is the ‘disorder’.

In the recent publications, the scaling behavior in ac conductivity data has been studied by hopping frequency ($\omega_p$), or by the directly measurable or accessible quantities such as temperature, the dc conductivity, the concentration, dielectric strength etc.

Usually two formalisms are followed to study the scaling behavior of the ac conductivity spectra at different temperatures. In the first approach, the loss peak frequency, or hopping frequency ($\omega_p$) is used as the scaling frequency for the frequency axis and dc conductivity for the conductivity axis based on Almond-West (A-W) conductivity formalism [9]. For the second, the magnitude of the minimum jump frequency, based on Dyre’s random free energy barrier model, used as the scaling frequency for the frequency axis.

In the A-W formalism, the scaling behavior of ac conductivity is obtained from the relation,
\[ \sigma'(\omega) = \sigma(0)[1 + \left(\frac{\omega}{\omega_p}\right)^n], \text{ where } \sigma(0) \text{ is the frequency independent conductivity and } n \text{ is the frequency exponent.} \]

In the RFEMB formalism of Dyre, the scaling behavior in ac conductivity is studied on the basis of the minimum jump frequency, \( \gamma_{\text{min}} \), of the distribution of jump frequencies that arise due to distribution of energy barriers in disordered ionic solids.

It has already been mentioned that scaling is an important concept, which serves to reduce the process to simpler parts so that deeper understanding might be achieved. Recently, studies have been made on large numbers of amorphous materials suggesting the temperature independent conductivity relaxation mechanism based on Time-Temperature superposition principle. The frequency dependent conductivity has been normalized with respect to the dc conductivity and measurement temperature by plotting

\[ \frac{\sigma(\omega)}{\sigma_{\text{dc}}} \text{ versus } f / \sigma_{\text{dc}} T \] according to Summerfield scaling law [10]. It has been observed that the ac conductivity curves obtained at different temperatures have merged into one super curve and this suggested that the ionic conductivity relaxation mechanism is independent of temperature.

The dielectric and ac conductivity studies on different types of glasses have been done by Ghosh et al.[11] and by G Govindaraj and his coworkers [12]. They have applied the scaling method to their systems and shown that scaling could be considered as an important technique to shed light on the different features of conductivity mechanisms.

In the high \( T_c \) superconductivity, scaling has also been considered as an important mathematical tool to extract various physics behind the many phenomena. S L Liu and his coworkers [13] have applied the scaling theory for verifying their proposed model regarding vortex-glass theory of HTSC.

Abu-Samreh and Saleh [14] investigated the frequency and field dependencies of the AC susceptibility of polycrystalline Ru-1212 superconductor in the presence of...
an AC applied magnetic field. In their study, the measured curves of AC susceptibility (ACS) components of the said sample were scaled onto a single curve using the peak temperature of the imaginary parts of ACS. The parameters emerged from the scaling analysis could be related to the hopping frequency between potential wells.

H Deguchi and others [15] have measured ac-susceptibilities of ceramic YBCO at various frequency under zero external field and carried out dynamic scaling analysis near intergrain transition temperature. Their scaling analysis is consistent with the results for the ceramic YBa$_2$Cu$_4$O$_8$ and for the d-wave model of a Josephson-junction array.

J Vanacken et al. [16] have formulated a model independent scaling approach to pure and Pr doped YBCO system. They applied the scaling method to the transport properties and obtained a characteristic temperature for each system. Finally they concluded that the transport properties of these systems are dominated by the same underlying scattering mechanism.

Wuyts and others [17] have studied the normal state transport properties of a series of YBCO system. The scaling method, applied by these workers, gives the estimation of some characteristic temperatures which has well agreement with that obtained from other experimental methods.
References: