# Fractal analysis of anomalous low-frequency dispersion in $X_2BaCuO_5$ compounds ($X$=La–Y)

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The anomalous low-frequency dispersion process observed in perovskite ceramics ($X_2BaCuO_5$, $X$ =La–Y) is described by a fractal model. The interpretation is given in terms of the Dissado-Hill-Jonscher theory and is based on a cluster description of the structural ordering and fluctuation in carrier-dominated dielectrics. The relationship of the mechanism to that of power-law noise in electrical systems is identified and its dynamical and structural interpretation explored. Particular features of several ceramics related to superconductor compounds are described. [S0163-1829(99)09931-2]

## I. INTRODUCTION

The electronic structure of strongly correlated copper-oxide superconductors such as $XBa_2Cu_3O_{7-x}$ ($X$=La–Y) has been an enduring problem in the last 15 years.[1–6] Together with these superconductors, another phase from the compounds $X_2BaCuO_5$ also appears. These ceramic compounds do not show a superconductivity transition and have been characterized as insulators. There are still numerous unresolved questions such as their dielectric response in the frequency domain.

The susceptibility $\chi(\omega)=\chi'(\omega)-j\chi''(\omega)$ of these compounds shows a peak at the loss component, $\chi''(\omega)$, together with an increase in the real component, $\chi'(\omega)$, corresponding to a relaxation process. The measurements give such loss peaks with a non-Debye shape[7,8]

$$\chi' \propto \chi'' \propto \omega^{-p}, \quad \omega < \omega_c \tag{1}$$

$$\chi' \propto \chi'' \propto \omega^{-(1-n)}, \quad \omega > \omega_c \tag{2}$$

where the frequency $\omega_c$ may be identified with the frequency of maximum loss and $n$ and $p$ are constants that lie between zero and unity.[9,10]

In this paper, a general fractal model is described and applied to explain this behavior. First it is assumed that the two power laws [Eqs. (1) and (2)] arise, basically, from hopping charge carriers. Secondly the mathematical background is developed to describe effective-charge displacements, between binding sites, in terms of the waiting-time distribution for anisotropic displacements (low-frequency limit) and over a fractal structure (high-frequency limit). Finally, using a fractal lattice that mimics the binding site net, the model is applied to the ceramic compounds ($X_2BaCuO_5$, $X$ =La–Y).

## II. DIELECTRIC RESPONSE OF CERAMIC COMPOUNDS

The orthorhombic phase 1:2:3 of $YBa_2Cu_3O_{7-x}$ exhibits a critical temperature of 90 K. Superconductor characteristics were also found in rare-earth compounds such as $XBa_2Cu_3O_{7-x}$ ($X$=La–Y). On the other hand, the phase 2:1:1 of $X_2BaCuO_5$ ($X$=La–Y) (Ref. 11) also appeared. All of them are isostructural from the spatial group *Pnma*, the main difference between them lying in their cation radius $\rho(La^{3+})>\rho(Y^{3+})$. The dielectric response in the frequency domain of these ceramic compounds is reported.

Dielectric properties were measured by an automatized and computer-controlled vectorial impedance meter HP 4227A. The relative dielectric constant $\varepsilon'(\omega)$ and relative loss factor $\varepsilon''(\omega)$ were measured in the frequency range $10^4$–$10^6$ Hz.

From Figs. 1 and 2 it can be seen that $\varepsilon'$ decays on increasing the frequency from $10^4$ to $10^6$ Hz. The $\varepsilon''$ maximum, associated with the delay of spatial charge polarization forms, is absent because of the rather low frequency at which it occurs ($<10^4$ Hz).

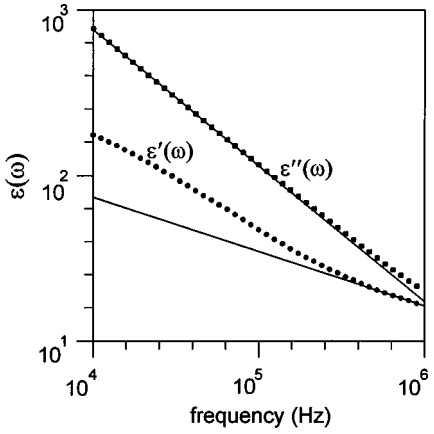


FIG. 1. Relative dielectric constant ($\varepsilon'$) and relative loss factor ($\varepsilon''$) vs frequency log($La_2BaCuO_5$).

 

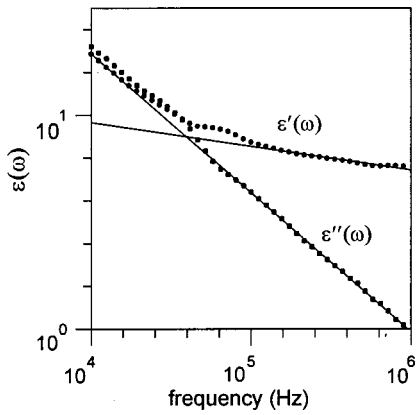


FIG. 2. Relative dielectric constant ($\varepsilon'$) and relative loss factor ($\varepsilon''$) vs frequency log($Y_2BaCuO_5$).

Taking into account the two slopes of the permittivity (Figs. 1 and 2), which are characteristic of systems in which the contribution of polarization comes from hopping charge carriers,[12] it is assumed that hopping charge carriers are the dominant polarization species in the $X_2BaCuO_5$ compounds.

Using the relation $\varepsilon(\omega)=\varepsilon_0[1+\chi(\omega)]$, $\chi'$ and $\chi''$ can be obtained from the experimental values of the dielectric constant $\varepsilon'$ and the relative loss factor $\varepsilon''$. In this way, using Eqs. (1) and (2), the resulting values of indexes $p$ and $n$ are $p=0.81\pm0.08$ and $n=0.67\pm0.07$ on $La_2BaCuO_5$ and $p=0.64\pm0.06$ and $n=0.89\pm0.09$ on $Y_2BaCuO_5$.

## III. THE FRACTAL MODEL

The anomalous low-frequency dispersion (LFD) is identified with the behavior of the perovskite ceramics $X_2BaCuO_5$ ($X$=La and Y). In these systems, a mechanically rigid lattice forms a regular array of binding sites for ions, most of which are occupied. This large occupancy factor allows the ions to interact with one another so as to modify the regularity of the intersite spacing of the substrate potential. As a result, the translational reproducibility will be destroyed over a distance $L_c$, which defines a positional correlation length for the cluster of ions formed. In the static structure the ions will reside in potential wells and vibrate around equilibrium sites mainly determined by the substrate lattice; therefore cooperative motions can be established.

An applied electric field will polarize the system by the hopping of ions to sites at a lower potential. In these cluster systems a distinction must be drawn between ion hopping over a range $L$, which can be either less than or greater than the cluster correlation length $L_c$. In the former case, the vibrations of the ion at the acceptor site are correlated with those of the rest of the cluster of which the donor site is part. Therefore, in this case, ion hopping constitutes a displacement of charge inside the cluster, which thereby becomes a polarized corporate entity. In the latter case, the motions of the ion are correlated with those of the acceptor rather than with those of the donor cluster, both of which become charged.[13–15]

The transport is assumed to take place between localized states $\mathbf{r}$, $\mathbf{r}'$, which are commonly present in insulators. $P(\mathbf{r},t)$ denotes the probability of finding a charge carrier at $\mathbf{r}$ at time $t$, if it was at $\mathbf{r}_0$ at time $t=0$. The generalized master equation (GME) for $P(\mathbf{r},t)$ is

$$\frac{\partial P(\mathbf{r},t)}{\partial t}=\int_0^t dt'\,\phi(t-t')\sum_{\mathbf{r}'}\left[V(\mathbf{r}-\mathbf{r}')P(\mathbf{r}',t')-V(\mathbf{r}'-\mathbf{r})P(\mathbf{r},t')\right], \tag{3}$$

where $V(\mathbf{r}-\mathbf{r}')\phi(t-t')$ are time-dependent transition rates (the spatial and temporal dependence were decoupled). $\phi(t)$ is called the memory function and takes into account the memory effect of this kind of non-Debye relaxation process.[16] By making the nearest-neighbor approximation for $V(\mathbf{r}-\mathbf{r}')$, applying an electric field $E$ in the $x$ direction, and taking the continuum limit of Eq. (3) it follows that

$$\frac{\partial P(\mathbf{r},t)}{\partial t}=\int_0^t \phi(t-t')\left(D_0\nabla^2 P(\mathbf{r}',t')-\mu_0 E\frac{\partial P(\mathbf{r},t')}{\partial t}\right)dt'. \tag{4}$$

The frequency-dependent diffusion coefficient can be derived from linear-response theory[17,18] as

$$D(\omega)=D_0\phi(\omega)=-(\tfrac{1}{6})\omega^2\langle r^2(\omega)\rangle \tag{5}$$

and the dielectric susceptibility is[17]

$$\chi(\omega)=j(ne^2/kT)D_0[\phi(\omega)/\omega]. \tag{6}$$

Now, $R(\mathbf{r},t)$ denotes the probability to reach site $\mathbf{r}$ in the time interval $(t,t+dt)$, $\beta(t)$ the probability to remain on a site for a time $t$ after arrival, and $\psi(t)$ the probability of a charge carrier arriving at $t=0$ to make a transition between $t$ and $t+dt$. Then,

$$\beta(t)=1-\int_0^t \psi(t')dt', \tag{7}$$

$$P(\mathbf{r},t)=\int_0^t \beta(t-t')R(\mathbf{r},t)dt', \tag{8}$$

$$R(\mathbf{r},t)-R_0(\mathbf{r},t)=\sum_{\mathbf{r}'} V(\mathbf{r}-\mathbf{r}')\int_0^t \psi(t-t')R(\mathbf{r}',t')dt', \tag{9}$$

where $R_0(\mathbf{r},t)$ is the initial condition which is a delta function $\delta(\mathbf{r}-\mathbf{r}_0)\delta(t-0)$. Note that because of normalization $\Sigma V(\mathbf{r}-\mathbf{r}')=1$.

To give the connection with the GME, the Laplace transform is applied to the above expressions:

$$\beta(\omega)=[1-\psi(\omega)]/\omega, \tag{10}$$

$$P(\mathbf{r},\omega)=\beta(\omega)R(\mathbf{r},\omega), \tag{11}$$

$$R(\mathbf{r},\omega)-\delta(\mathbf{r}-0)=\sum_{\mathbf{r}'} V(\mathbf{r}-\mathbf{r}')\psi(\omega)R(\mathbf{r}',\omega), \tag{12}$$

where $\omega$ is the Laplace-transformed variable using the formula for the transform of a convolution. When $\mathbf{r}\neq\mathbf{r}_0$, the above equations yield the relation

$$P(\mathbf{r},\omega)=\sum_{\mathbf{r}'} V(\mathbf{r}-\mathbf{r}')\psi(\omega)P(\mathbf{r}',\omega). \tag{13}$$

The Laplace transform of the GME, Eq. (3), can be written as (if $\mathbf{r}\neq\mathbf{r}_0$)

$$\omega P(\mathbf{r},\omega)=-\phi(\omega)P(\mathbf{r},\omega)+\phi(\omega)\sum_{\mathbf{r}'} V(\mathbf{r}-\mathbf{r}')P(\mathbf{r}',\omega), \tag{14}$$

where Eq. (9) was used. Now if Eq. (13) is inserted into Eq. (14) the following relation between the memory function $\phi(t)$ and the waiting-time distribution $\psi(t)$ is obtained:

$$\phi(\omega)=\omega\psi(\omega)/[1-\psi(\omega)]. \tag{15}$$

### Fractal time process

The intercluster motion is considered first as a fractal-time process. In a fractal-time process the distribution of waiting time can be described by the average number of events $N(t)$ within time $t$ and can be written as

$$N(t)\propto t^{D_t}, \tag{16}$$

where $D_t$ is called the fractal dimension of the process.

Let $g_n(t)$ be the probability that $n$ events happen at time $t$. Then the Laplace-transformed quantity $g_n(\omega)$ is given by

$$g_n(\omega)=(1/\omega)[\psi(\omega)]^n[1-\psi(\omega)]. \tag{17}$$

The Laplace transform of the number of events within time $t$ can be written as

$$N(\omega)=\sum_{n=0}^{\infty} ng_n(\omega)=\psi(\omega)/\omega[1-\psi(\omega)]. \tag{18}$$

In order to recover Eq. (16), $N(\omega)\propto\omega^{1-D_t}$, which is obtained when $\psi(\omega)=1-A\omega^{D_t}$ for a small $\omega$. At long times, this is equivalent to the asymptotic form

$$\psi(t)=t^{-1-D_t}. \tag{19}$$

Then, to write down the asymptotic dielectric response for a fractal time process at low frequencies the memory function $\phi(\omega)\propto\omega^{1-D_t}$ and the dielectric susceptibility are obtained from Eqs. (15) and (6), respectively.

$$\chi(\omega)\propto\omega^{-D_t}. \tag{20}$$

Note that $D_t$ plays the role of $p$ in Eq. (1) and it can only take values in the interval [0,1].

### Fractal time process on fractal structures

The displacement of charge within the cluster and the regression of these displacements, which includes the contribution to the response from the relaxation of the cluster polarization, were analyzed. The cluster is considered to have a fractal structure on length scales between the size of its building blocks and the size of the whole cluster ($L_c$). The fractal structure mimics some of the characteristics of perovskite ceramic lattice geometry.

The rms $\langle r^2(t)\rangle$ displacement of a charge carrier for a time $t$ over a fractal cluster is given by

$$\langle r^2(t)\rangle=\sum_{n=0}^{\infty}\langle r_n^2\rangle g_n(t), \tag{21}$$

where $g_n(t)$ is the probability of performing exactly $n$ steps for $t$ and $\langle r_n^2\rangle$ is the rms displacement of a random walk of $n$ steps on a fractal, which is given by[19] $\langle r_n^2\rangle\propto n^{2/D_w}$. $D_w$ is the random walk dimension. The Laplace transform of the $\langle r^2(t)\rangle$ is given by

$$\langle r^2(\omega)\rangle=[1-\psi(\omega)]/\omega\Gamma(\alpha+1)[-\ln\psi(\omega)]^{-\alpha-1}, \tag{22}$$

where $\alpha=2/D_w$ and $\Gamma(x)$ is the gamma function. Considering $\ln\psi(\omega)\approx\psi(\omega)-1$ Eq. (22) becomes

$$\langle r^2(\omega)\rangle=\Gamma(\alpha+1)[1-\psi(\omega)]^{-\alpha}/\omega. \tag{23}$$

Now, taking into account $\psi(\omega)=1-A\omega^{D_t}$ and writing

$$\langle r^2(\omega)\rangle\propto\omega^{-\alpha D_t-1} \tag{24}$$

according to Eqs. (5) and (6) it follows that

$$\chi(\omega)\propto D(\omega)/\omega\propto\omega^2\langle r^2(\omega)\rangle/\omega\propto\omega^{-\alpha D_t}. \tag{25}$$

Note that $1-\alpha D_t$ plays the role of $n$ in Eq. (2) and it can only take values in the interval [0,1].

## IV. RESULTS

From the experimental data and the proposed model [see Eq. (20)] the following values are obtained for the fractal dimension $D_t$: $La_2BaCuO_5$: $D_t=0.81\pm0.08$ and $Y_2BaCuO_5$: $D_t=0.64\pm0.06$. The definition of $p$ indicates that for large values of $p$ intercluster exchange fluctuations carry the effective charge over long paths, affecting many clusters as transport develops. Then, the greater values of $p$ in $La_2BaCuO_5$ indicate near perfect transport, while the low value of $p$ in $Y_2BaCuO_5$ indicates poor transport.

In terms of burst and gaps[20] $D_t$ can be identified by a Cantor set, i.e., dividing the closed interval [0,1] into pieces of length $1/b$, then removing some of them and preserving only $N$ pieces, and so on to infinity. This gives $D_t=\ln N/\ln b$, $N=7$, and $b=11$ for $La_2BaCuO_5$ and $b=21$ for $Y_2BaCuO_5$.

The corresponding values for $D_w$ [see Eq. (25)] are $La_2BaCuO_5$: $D_w=4.9\pm1.3$ and $Y_2BaCuO_5$: $D_w=11.6\pm5.8$. The definition of $n$ indicates that small values of $n$ correspond to a highly irregular cluster (larger values of $D_w$), and large values of $n$ correspond to a highly ordered cluster. Then, the low value of $n$ in $La_2BaCuO_5$ corresponds to a highly irregular cluster, such as in the case when an interstitial ion is present, while greater values of $n$ in $Y_2BaCuO_5$ indicate a highly ordered cluster structure.

Although a deterministic fractal is not a disordered medium, diffusion on a fractal exhibits anomalies (due to obstacles such as holes, bottlenecks, dangling ends, etc.) similar to those present in disordered media.

The renormalization scheme[21–23] is used for the calculation of the exact values of $D_w$ for the fractal lattices shown in Fig. 3.

Let us consider the mean transit time $T$ needed to traverse a lattice unit from one vertex to another, $O$ [see Fig. 3(a)]. This is done by exploiting the Markov property of the ran-

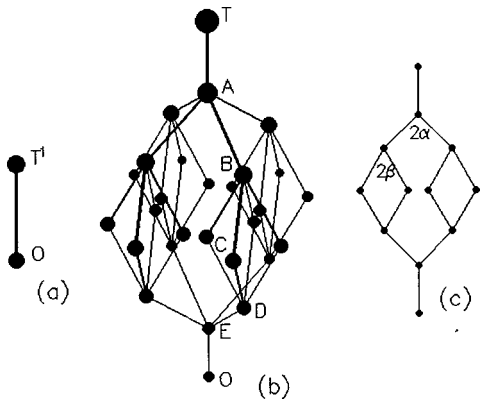


FIG. 3. (a) The lattice unit. (b) The rescaled fractal unit. (c) Section of the lattice (c) showing angles $\alpha$ and $\beta$.

dom walk on the fractal.[23] Thus, $T$ equals the time to exit the first lattice unit, plus $A$, the mean transit time needed to leave the rescaled unit from then on. Using the same reasoning for the times $A$, $B$, $C$, $D$, and $E$ (the mean exit times starting from the decimated nodes)

$$T = T' + A,$$

$$5A = 5T' + 4B + T,$$

$$5B = 5T' + A + 4C,$$

$$2C = 2T' + B + D,$$

$$5D = 5T' + 4C + E,$$

$$5E = 5T' + 4D.$$

The solution is $T' = (\frac{21}{2})^2 T$, which is the rescaling of time for a diffusion process on the lattice upon the rescaling of length by a factor of $2(1+\cos\alpha+\cos\beta)$ [see Fig. 3(c)]. Then, it follows that

$$D_w = 2\ln(\tfrac{21}{2})/\ln[2(1+\cos\alpha+\cos\beta)].$$

A model for the anomalous low-frequency dispersion process that directly relates the mechanism to the disordered structure arising from partial occupancy of binding sites in a material network has been used. This structure is described by a cluster array with low-frequency dispersion arising from the transport of effective-charge displacements when the bound species are constrained to move only in the network. The two power-law regimes in the frequency domain have been identified with effective charge transport on the cluster size, and the intercluster macroscopic size scales with their respective indices defining their appropriate structural order.

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