Frequency-dependent (ac) Conduction in Disordered Composites: a Percolative Study

Asok K. Sen*

LTP Division, Saha Institute of Nuclear Physics
1/AF, Bidhannagar, Calcutta 700 064, India

and

Abhijit Kar Gupta†

Institute of Mathematical Sciences, CIT Campus
P.O. Taramani, Chennai 600 113, India

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Abstract

In a recent paper [Phys. Rev. B57, 3375 (1998)], we examined in detail the nonlinear (electrical) dc response of a random resistor cum tunneling bond network (\textit{RRTN}, introduced by us elsewhere to explain nonlinear response of metal-insulator type mixtures). In this work which is a sequel to that paper, we consider the ac response of the \textit{RRTN}-based correlated RC (CRC) model. Numerical solutions of the Kirchoff’s laws for the CRC model give a power-law exponent (= 0.7 near $p = p_c$) of the modulus of the complex ac conductance at moderately low frequencies, in conformity with experiments on various types of disordered systems. But, at very low frequencies, it gives a simple quadratic or linear dependence on the frequency depending upon whether the system is percolating or not. We do also discuss the effective medium approximation (\textit{EMA}) of our CRC and the traditional random RC network model, and discuss their comparative successes and shortcomings.

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*asok@cmp.saha.ernet.in : Author for communication
†abhi@imsc.ernet.in
1 Introduction

Electrical response of a composite material made up of conducting and insulating components to an external field is still of much interest. To find the electrical conduction properties of random mixtures one usually models a network with the notions of percolative processes \cite{1, 2} as a basic framework. For simplicity one may consider a lattice-based percolation and assume that the occupied bonds (with a probability $p$) are metallic or good conductors and the rest of the unoccupied bonds (with a probability $1 - p$) are insulators or very poor conductors. So it turns out that one is dealing with a random electrical network where each bond of the regular lattice constitutes an appropriate circuit element. To study the linear direct current (dc) response of a random binary mixture, one considers it to be a random (linear) resistor network ($RRN$) where the conductance $g$ of a circuit element assumes the value $g_1$ with probability $p$, or $g_2$ ($= 0$ for insulators) with the complementary probability $1 - p$. When one attempts to study the alternating current (ac) response of such a system at a frequency $f = \omega/(2\pi)$, the elementary conductances $g_1$ and $g_2$ do, in general, become complex quantities, termed as complex ‘admittances’ (inverse of impedances), because of the presence of inductances and/or capacitances in some parts of the circuit. Electrical conductivities (both dc and ac) of various composite systems, have been the subject of numerous studies \cite{3, 4, 5, 6}. On the theoretical side, analytical calculations involving related Maxwell’s equations with appropriate boundary conditions, effective medium approximation ($EMA$), series expansion method, Monte Carlo simulation, model random resistor network ($RRN$), etc. are employed to study various interesting behaviors.

The key experimental facts to be contended with by a model in the case of nonlinear dc response have been discussed in our recent works \cite{6, 7}. To reproduce the very low percolation threshold, a ‘low’-temperature resistance minimum, and the nonlinear electric (dc) response of metal-insulator composites, we had introduced in our original work \cite{8} a new semi-classical bond percolation model. The distinguish-
ing feature in this model (compared to earlier RRN, etc.) is the possibility of a local tunneling/ dielectric breakdown (beyond a microscopic threshold voltage, \( v_g \)) between two metallic bonds separated by a nearest neighbor lattice distance. We call these specially positioned insulating bonds, tunneling or ‘t-bonds’. All the other insulating bonds separated farther away remain insulating at any voltage however large. Clearly, since the positions of these t-bonds are totally correlated to the positions of the metallic bonds in a particular (random) configuration, this model may be considered as a correlated bond percolation model. The percolative phase transition aspects [8] and the dielectric breakdown aspects [9] for this model have already been discussed elsewhere. Based upon this correlated bond percolation, the circuit made of the random network of resistors and t-bonds has been called a RRTN (random resistor cum tunneling bond network).

In the present work which is a sequel to our work on the nonlinear dc response of the RRTN [7] (referred to as I from now on), we are concerned with the modeling of the generic ac response of a whole variety of experimental systems within the RRTN framework (reported briefly first in the ref.[8]). Experiments on the complex ac conductance \( G(\omega) \) in various composite systems, dispersed metals etc., as well as many disordered/ amorphous systems [2, 3, 4, 5, 10, 11, 12], report a non-integer power-law behavior of the modulus, \( |G(\omega)| = [(Re G)^2 + (Im G)^2]^{1/2} \). At a fixed voltage and at a moderately low-\( \omega \), \( |G(\omega)| - G(0) \propto \omega^\alpha \), where \( \alpha \approx 0.7 \) (the case of very low \( \omega \)'s is discussed later). Now, at a low voltage \( V \) (see I), \( [G(V) - G(V = 0)] \propto (V - V_g)^\delta \), where \( V_g \) is the macroscopic threshold voltage for dc nonlinearity and the exponent \( \delta \) is dimensionality-dependent. There has been a recent claim [10] for 3D carbon-wax composites that \( \delta = \alpha = 1.4 \). We shall comment on this later.

In the traditional RC network models used to study the ac response of composites, the conducting bonds are pure (real) resistors and all the insulating bonds may behave as capacitors in the presence of an ac-field. A fairly complete review and references (from the percolative aspect) on the linear response may be found in the ref. [2]. Another more recent review in this regard is due to Shalaev [13].
If one applies an ac electric field across our $RRT N$ model, a t-bond between two nearby metallic bonds are expected to behave as a capacitor. Note that perfect capacitors at all the t-bonds correspond to a situation where all the t-bonds have zero dc conductance at low voltages, and hence the $RRT N$ is in its lower linear dc (or, ohmic) regime. Similarly, leaky capacitors with a very low constant conductance for all the t-bonds, implies that the $RRT N$ is in its upper linear dc regime. To obtain the nonlinear ac response in the truly nonlinear dc regime, one has to let the t-bonds be active or passive according to the voltage differences across them. In this work we study the nonlinear ac response in either the upper or the lower linear dc regimes only. Further, we make a simplifying assumption that all the capacitances across insulators farther than the nearest neighbor distance are zero. Based on the $RRT N$ model, this model for studying ac response may thus be called a correlated RC (CRC) model. We believe that the simplicity of our model is physically appealing and realistic enough. Since the capacitors placed in this way never percolates by themselves, the $|G(\omega)|$ is expected to connect nonlinearly between its own lower and upper ac saturation regimes of $\omega \to 0^+$ and $\omega \to \infty$ respectively.

To make the above discussions more concrete we show in Fig. 1, a typical numerical result for the real part of the ac conductance for $p = 0.52$ in a $20 \times 20$ size sample with leaky capacitors. There may be some genuine concerns whether the upper saturation of the ac conductance does actually occur in realistic samples. In response we note that in most of the early experiments, the steady frequency required to approach the upper ac saturation regime were probably too high to be accessible. Further, we find at least one experiment on $Li$-doped $NiO$ sample [14], where the upper saturation is clearly observed.

As a pedagogical example (which will be useful for our interpretations of the $EMA$ and the simulation results in the sequel), one may calculate the complex $G(\omega)$ for some simple prototype circuits using the capacitive conductance $g_t = j\omega c$
where \( c \) is the microscopic capacitance and \( j = \sqrt{-1} \). For the case of Fig. 2(a):

\[
G(\omega) = \frac{(r_1 + r_2 + \omega^2 r_1 r_2^2 c^2) + j \omega r_2^2 c}{(r_1 + r_2)^2 + \omega^2 r_1^2 r_2^2 c^2}.
\] (1)

On the other hand, for the case of Fig. 2(b):

\[
G(\omega) = \frac{\omega [\omega (r_1 + r_2) c^2 + j c]}{1 + \omega^2 (r_1^2 + r_2^2) c^2}.
\] (2)

It may be noted that the dc conductance \( G(\omega = 0) \) of the first circuit is \( 1/(r_1 + r_2) > 0 \) (thus it is a conductor), whereas the dc conductance of the second circuit is zero (an insulator). The extremely low frequency \( (\omega \rightarrow 0^+) \) behaviors for both the circuits is \([\text{Re } G(\omega) - G(\omega = 0)] \propto \omega^2 \) and \( \text{Im } G(\omega) \propto \omega \). Thus, \( \text{Re } G(\omega) \) or \( \text{Im } G(\omega) \) at very low \( \omega \) cannot distinguish between the two types of circuits. But in the same limit, \([|G(\omega)| - G(\omega = 0)] \propto \omega^2 \) for the elementary \( \text{percolating} \) circuit of Fig. 2(a), while \( |G(\omega)| \propto \omega \) for the elementary \( \text{non-percolating} \) circuit of Fig. 2(b). As \( \omega \rightarrow \infty \), the upper ac saturation value for the Fig. 2(a) is \( 1/r_1 \) and that for the Fig. 2(b) it is \( 1/(r_1 + r_2) \), both of which are finite because the capacitor \( c \) does not geometrically extend from one electrode to the other.

Note that on adding together many such elementary circuits (i.e., on increasing \( L \)), the rational algebraic function type behavior of \( \text{Re } G(\omega) \) obtained from Eq. (1) and Eq. (2) changes over to a sigmoidal type function as shown in the Fig. 1 and looks qualitatively very similar to the nonlinear dc conductance as a function of \( V \) \cite{1, 7}. Here, \( \text{Im } G(\omega) \) is zero for both \( \omega = 0 \) and \( \infty \), with a broad peak in-between. An identical fitting function as the one for \( G(V) \) in I, fits (solid line) the simulation data (open circles) very well for six decades. In the same spirit, \( |G(\omega)| \) in the ac case is writen as:

\[
|G(\omega)| = G(\omega = 0) + G_d(p)[1 - \exp(-\lambda \omega^n)]^\gamma.
\] (3)

To obtain the power-law behavior, one linearizes the exponential function in Eq. (3) for small \( \omega \) (much below the upper ac saturation regime) and one gets: \([G(\omega)] - G(0)] \propto \omega^\alpha \), where \( \alpha = \mu \gamma \). Now, for considering the ac response in our \( CRC \) model,
we have virtually a three component mixture of the ohmic bonds (conductance $g_o = g$), the t-bonds (in general leaky with a complex $g_t = g_c + j\omega c$), and the insulating bonds ($g_t = 0$). We assume that a sinusoidal voltage $V = V_0 \exp(j\omega t)$ is applied across the network. We set $V_0 = 1$, $c = 1$ and $g_o = 1$ for convenience, thereby setting the scales for the voltage, the frequency and the conductance respectively. We shall concentrate on either the $Re \ G$ and $Im \ G$, or the $|G|$ as a function of $\omega$. We would also study the phase-angle of the complex $G$ relative to the phase of the voltage source at any time $t$.

The rest of the paper is organized as follows. In the Sec. 2, we discuss the effective medium approximation ($EMA$) results for our CRC model. Next in the Sec. 3, we present the numerical Kirchhoff’s law solution for the same model and compare them with the EMA (for the CRC) and some basic experimental results. Finally, in the Sec. 4, we summarize our findings with some concluding remarks.

### 2 The EMA result for the CRC Model

For studying the ac conductance of the correlated $RC$ model within the $EMA$, we assume for simplicity that the t-bonds behave as perfect capacitors ($g_t = j\omega c$). So, we are in the lower linear dc regime. The probabilities of a bond to be ohmic, tunneling (capacitive) or purely insulating in a 2D square lattice are:

\begin{align*}
P_o & = p, \quad (4) \\
P_t & = (p^3 + 3p^2q + 3pq^2)^2q, \quad (5) \\
P_i & = 1 - P_o - P_t = \left[1 - (p^3 + 3p^2q + 3pq^2)^2\right]q, \quad (6)
\end{align*}

where $q = 1 - p$. The EMA equation for the effective (i.e., average) complex conductance $G_e$ for this three component system may be written as [4, 15]:

\begin{equation}
AG_e^2 + BG_e + C = 0, \quad (7)
\end{equation}
where \( A = (d-1)^2 \), \( B = (d-1)\left[ (1-dP_o)g_0 + (1-dP_t)g_t \right] \) and \( C = -\left[ (d-1)-dP_t \right]g_o g_t \).

The solution of the quadratic Eq. (7) is

\[
G_e = \frac{-B \pm \left( B^2 - 4AC \right)^{1/2}}{2A}.
\]

We show below that only the `+` sign in front of the square-root is the physical one for our purpose and henceforth use that only. In passing we note that the new percolation threshold for our \( RRTN \) on a square lattice is \( p_{ct} = 1/4 \) using the \( EMA \) (see I) and \( p_{ct} \approx 0.181 \) \[8\] using a real space renormalization \[16\] with finite size scaling analysis.

Separating the real and the imaginary parts of \( G_e(\omega) \), one gets

\[
Re \, G_e(\omega) = \frac{(2P_o - 1)}{2} + \frac{1}{2\sqrt{2}}[X + (X^2 + Y^2)^{1/2}]^{1/2},
\]

and,

\[
Im \, G_e(\omega) = \frac{\omega(2P_t - 1)}{2} + \frac{1}{2\sqrt{2}}[-X + (X^2 + Y^2)^{1/2}]^{1/2},
\]

where \( X = (2P_o - 1)^2 - \omega^2(2P_t - 1)^2 \) and \( Y = 2\omega[(2P_o - 1)(2P_t - 1) - 2(2P_t - 1)] \).

It may be noted here that \( P_t(p) \) has a single broad peak structure with a maximum value of about 0.3840 at a \( p = 0.4800 \). Thus, the quantity \( \omega(2P_t - 1) \) is always negative. Further, \( X \) is also negative for \( \omega > |(2P_o - 1)/(2P_t - 1)| \) and approaches \(-\infty\) quadratically as \( \omega \to \infty \). We will take only the absolute value of the square-rooted expression for two reasons; (i) \( Re \, G_e(\omega) \) in Fig. 3 achieves the necessary upper ac saturation since the built-in square-root function in the computer uses exactly that, and (ii) this procedure keeps \( Im \, G_e(\omega) > 0 \) for all \( \omega > 0 \) (needed since \( Im \, g_t > 0 \)).

Let us first check the \( \omega \to \infty \) limit. The dissipative part of the complex conductance, \( Re \, G_e(\omega) \), should be positive, finite, and greater than \( G_e(\omega = 0) \) in this limit. Further, the reactive part \( Im \, G_e(\omega) \) of the \( CRC \) network must become zero (i.e., show no response) when the driving field oscillates much faster than the network’s relaxation time (or, the \textit{time-constant}). Now, in this limit,

\[
Re \, G_e(\omega) = \frac{(2P_o - 1)}{2} + \frac{1}{2} \left| \frac{2(2P_t - 1)}{(2P_t - 1)} - (2P_o - 1) \right|,
\]
and,
\[
Im G_e(\omega) = \frac{\omega(2P_t - 1)}{2} + \frac{1}{2} |\omega(2P_t - 1)|, \quad (12)
\]
and thus for all \( p > p_{ct} \), \( G_e(0) < Re G_e(\omega = \infty) < \infty \). Also, clearly \( Im G_e(\omega) \) in this limit is zero. Thus, both the conditions hold in the \( \omega \to \infty \) limit.

Next we check the asymptotic expansion as \( \omega \to 0^+ \). In this limit, \([Re G_e(\omega) - G_e(0)] \sim \omega^2\) and \( Im G_e(\omega) \sim \omega \). We find that this extremely low-\( \omega \) behavior of the complex \( G_e \) in the EMA is generic for all \( p_{ct} < p < 1 \) (except at \( p = p_c \), see below), and \([|G_e(\omega)| - G_e(\omega = 0)] \sim \omega^2\). Hence the EMA cannot distinguish between a percolating and a non-percolating configuration from their low-\( \omega \) behavior.

Now, in the special case when \( p = p_c (= 1/2 \) for a square lattice), we find that in the limit \( \omega \to 0^+ \), both the \( Re G_e(\omega) \) and \( Im G_e(\omega) \) varies as \( \omega^\alpha \), where \( \alpha = 0.5 \). Obviously, \( |G_e(\omega)| \sim \omega^{0.5} \) in this limit. In passing we would like to quote the extremely low-frequency EMA exponent in the case of 3D. By looking at the EMA expressions for \( G_e(\omega) \) which are the analogues of Eqs. (9) and (10) for a simple cubic lattice at its EMA percolation threshold (\( p_c = 1/3 \) in 3D), one finds again that \( \alpha = 0.5 \) (in 3D).

We show in Fig. 3 a log-log plot of \( Re G_e(\omega) \) against \( \omega \) in 2D [Eq. (9)]. In conformity with the asymptotic expansions obtained above analytically for very small \( \omega \)'s, the EMA results shown in the Fig. 3 give \( \alpha(p) = 2.0 \) for all \( p_{ct} < p < 1.0 \) except for the special case of \( p = p_c \). We have \( \alpha(p_c) = 0.5 \) both in the very low and in the moderately low-\( \omega \) regimes much below the upper saturation of \( Re G_e(\omega = \infty) \). Further for each fixed \( p \neq p_c \), there is a characteristic \( \omega_0(p) \) around which \( \alpha(p) \) starts crossing over from 2.0 to the moderately low-\( \omega \) exponent of about 0.5. The jump of \( \alpha(p) \) at an extremely low-\( \omega \) from 2.0 to 0.5 is the hallmark of the inadequacy of EMA. One does also note that for a fixed \( p \neq p_c \), this crossover region becomes smaller for smaller \( p \)'s. Indeed, the crossover region finally tends to vanish as \( p \to p_{ct} \) (\( \simeq 0.18 \) in 2D). Thus, we observe that there are no low to moderate \( \omega \) crossovers for for \( p = p_c \) with \( \alpha = 0.5 \) and similarly for \( p = p_{ct} \) where \( \alpha = 2.0 \).

We did also calculate the EMA results with leaky capacitors at each t-bond
(g_t = g_c + jωc), i.e., in the upper linear dc regime. The only change here compared to the case of perfect capacitors above is that even at $p = p_c$, $|G_e(\omega)| = G_e(\omega = 0) + k\omega^2$ ($k=$constant) for very low-$\omega$. Also for all $p > p_{ct}$, $G_e(\omega = 0) > 0$, as expected.

We would also like to see the behavior of phase-angle of the complex conductance $G_e(\omega)$ with respect to the frequency ($\omega$). The phase-angle ($\phi$) is defined through

$$\tan \phi = \frac{Im \ G_e(\omega)}{Re \ G_e(\omega)}.$$  \hspace{1cm} (13)

In the Fig. 4, we plot the phase-angle ($\phi$) of the complex conductance against frequency ($\omega$) for the EMA. As expected, this angle is zero [just like the $Im \ G_e(\omega)$] both at very small and at very large $\omega$'s. Further the phase has a peak value $\phi = \phi_m$ which increases as $p$ is decreased and the $\omega$ at which the peak occurs is $p$-dependent.

We find from this Fig. 4 that the positions of the peaks tend towards zero and that $\phi_m$ becomes progressively larger as $p$ approaches $p_c$ from higher values. It may be noted that we cannot calculate the phase angle $\phi$ for $p < p_c$ in the EMA with confidence because the quantity $G_e(\omega = 0) = 2P_o - 1$ takes on unphysical negative values. Hence we have not shown any curve for $p < p_c$ in Fig. 4.

3 The Numerical Result for the CRC Model

We now solve Kirchoff’s laws in our 2D complex network at each node of our correlated RC model. We obtain the complex conductance of the macroscopic samples, their real and imaginary parts, the modulus values and the phase-angle through iterative numerical solution using the Gauss-Seidel relaxation method. In the Fig. 5, we have plotted the modulus of the average complex conductance, $|<G(\omega)>|$ against $\omega$ for $0.3 \leq p \leq 0.7$ and for an external sinusoidal voltage $V = \cos(\omega t)$. We let the t-bonds be leaky capacitors with a conductance $g_c = 0.001 << g_o$ and averaged over the same twenty configurations for each $\omega$. So, we are virtually in the upper dc saturation regime. For clarity, we have shown the graphs in Fig. 5 from zero to a moderately low $\omega$ (=0.1) much below the upper ac saturation regime. We
find that for \( p \simeq p_c \), the respective graphs in Fig. 1 and Fig. 5 are fitted very well with the Eq. (3) such that \( \alpha = \mu \gamma \simeq 0.7 \). Thus, in the moderately low-\( \omega \) regime, \( \alpha \) obtained experimentally in varieties of systems [11, 17], and in an ‘Extended Pair Approximation’ (EPA) theory [18] is close to what we obtain here (i.e., 0.7). We remark here that for a 2D system exactly at \( p_c \), \( \alpha = 0.5 \) both in the simple RC model (see e.g., Ref.[2]) and the EMA of our correlated RC model. Indeed, both of them fall short of the realistic value of \( \alpha \) not only at \( p = p_c \), but also at any \( p \neq p_c \). Further, these theoretical and experimental results do not support any evidence for the statement that \( \alpha = \delta \) [10]. For example, for our RRTN model near \( p_c \) in 2D, \( \delta \simeq 1 \), whereas \( \alpha \simeq 0.7 \). Indeed, as we had commented recently [19], there is no good physical argument supporting the equality. The main point in the comment is that since the actual experimental data become flat at large \( \omega \), it is quite well-known that the exponent obtained may depend crucially on the arbitrarily chosen (i.e., without any particular physical reason) upper cut-off in \( \omega \) for fitting purposes. Consequently, the possibility of an unique function should be explored (as we do here) which fits all the way from the lower to the upper saturation range (if sufficient data are available), and then the power-law behavior in the moderately low-\( \omega \) range should be extracted from that function.

Now, as we had mentioned before, we know of at least one experiment by Pollak et al. [14] on Li-doped NiO single crystals at sufficiently low temperatures, where the upper saturation of the \( Re G_e(\omega) \) may be clearly observed. The frequency range used in that work is from about \( 10^7 \) Hz to about \( 10^{10} \) Hz. We did not try to fit them by our method since no data below \( 10^7 \) Hz was available. In any case we note that the upper saturation is also consistent with the fact that the measured relaxation time for this sample is about \( 2.2 \times 10^{-10} \) s. In passing we do also note that in this system as well, the doping concentration of Li is extremely low: from about \( 13 \times 10^{-6} \) to about \( 136 \times 10^{-6} \), and that the pristine NiO is an insulator. We observe that the upper saturation value of the conductivity for this experiment [14] is about 0.1 S/m, and that the data for all the three concentrations used for Li, fall
closely enough to their fitting function in the high frequency range, but not so well in the low/ moderate frequency range. The power-law exponent $\alpha$ (around $10^7$ Hz) seems to be close to 2.0.

Next, we would like to emphasize that the apparently excellent fitting shown in Fig. 1 with Eq. (3) may be misleading to the eye in the very low-$\omega$ regime. A careful analysis of data in a more revealing log-log plot shows that for very low $\omega$, the fittings may be actually quite bad. We observed for the two elementary circuits (in Fig. 2) for very low $\omega$, $|G(\omega)| \sim \omega^2$ or $\omega$ depending upon whether the circuit is percolating or not. Similarly, we anticipate that for extremely low $\omega$ and in the lower dc regime, the CRC network would also show such simple behaviors (instead of a more complicated percolative behavior in the moderately low-$\omega$ regime as described above). Further this simple behavior is expected to persist (as seen in our EMA results) generically for each $p$ up to some scaled crossover frequency $\omega_0$ depending on $p$, $g_0$, $g_c$ and $c$. For $\omega > \omega_0$, we expect macroscopic percolative effects to gain control and instead of Eq. (3), $|G(\omega)|$ should follow a more general equation closer in form to that used for the dc-conductance [cf. Eq. (4.7) of I]:

$$|G(\omega)| = G(\omega_0) + G_d(p)[1 - \exp(-\lambda[\omega - \omega_0]^{\mu})]^{\gamma}. \quad (14)$$

A typical fit by the above equation is shown in Fig. 6 for a system size $L = 20$ and $p = 0.3$ for about seven decades in $\omega$ using leaky capacitors. In the inset of Fig. 6, we show the log-log plot for the modulus value of $G$ and observe a very convincing quadratic behavior with $|< G(\omega) >| = 0.01219 + 55\omega^2$ for $\omega_0 \leq 0.01$. This observation is totally matching with our analyses for Fig. 2(a). For $\omega > \omega_0$, we get a very good fit for the intermediate frequency range with the general Eq. (14) where $\alpha = \mu \gamma \simeq 3.0$ (which is much larger than 0.7).

Next in the Fig. 7, we show another fit for a single configuration (at $p = 0.45 < p_c$) with perfect capacitors at the t-bonds ($g_c = 0$, non-percolating) of the CRC network with $L=20$. In this case one can easily observe that in the very low-$\omega$ range up to a crossover frequency $\omega_0 \simeq 0.01$, $|G(\omega)| \simeq 6\omega$. This behavior is akin to that
of the elementary circuit of Fig. 2(b). Further, beyond $\omega_0$, we do again have an excellent fitting with the Eq. (14) with $\alpha = \mu \gamma \approx 0.5$. So we have two things to note here for non-percolating configurations (with perfect capacitors, i.e., in the lower dc regime): (i) the very low-$\omega$ behavior is linear in $\omega$, and (ii) the intermediate frequency behavior seems to give a lower value for the exponent compared to that for percolating configurations (e.g., as in the upper dc regimes of Fig. 1 and Fig. 6 where $\delta = 0.7$ near $p_c$). Note that for the non-percolating configurations (only at $p_c$) in the lower linear dc regime, $\alpha = 0.5$ in the case of $EMA$ as well.

For many practical situations, this intermediate frequency range (away from both the lower and the upper ac saturation regimes) is of prime interest. For t-bonds with leaky capacitors at any $p > p_{ct}$ (i.e., in the upper dc regime), we find that $\alpha$ has a minimum value of about 0.7 near $p_c$, and increases on both sides of it with a value of about 3.0 at $p = 0.3$ (as shown above) and of about 1.35 (not explicitly shown here) at $p = 0.7$. Clearly, this result has a qualitative similarity with the dc nonlinearity exponent $\delta(p)$ as a function of $p$ as shown in the Fig. 11 of I.

Finally, the variation of the phase-angle ($\phi$) with frequency ($\omega$) obtained by numerical (Kirchoff’s laws) solution of our $CRC$ network has been shown in the Fig. 8 for $0.3 \leq p \leq 0.7$. One can easily observe from this figure that in the $CRC$ model, for configurations with $p$ around $p_c$ for a 2D square lattice, the peak value of the phase, $\phi_m \cong 0.7$ (radian). This is close to the universal phase-angle value of $\pi/4$ radian obtained in the simple $RC$ model in 2D at $p_c$ as predicted by Clerc et. al.\[2\]. As noted before, the $EMA$ calculations (i.e., mean-field approach) of the phase $\phi(\omega)$ for our $CRC$ model also obtains $\phi_m \cong 0.7$ rad when $p \cong p_c$. The main problem with the $EMA$ in this case is that the peak occurs at an $\omega$ of about 0.1 rad/s (instead of at around 0.2 rad/s as for the $CRC$ in Fig. 8 above) and that it is too broad. Further, as shown in the Fig. 8 for the $CRC$ network which is expected to be much more realistic (better than the mean-field theory, for one reason) from the demonstrations and the arguments above, $\phi_m$ increases as $p$ decreases: from a value of $\phi_m \cong 1.1$ rad for $p = 0.3$ to a value of $\phi_m \cong 0.4$ rad for $p = 0.7$. 

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4 Summary and Conclusion

In this work we have studied the nonlinear ac response (in the two linear dc regimes) of a variety of materials, e.g., a binary metal-insulator type composite, a dispersed metal, amorphous semiconductors at high field, etc. by modeling them in terms of our recently proposed \textit{RRTN} [6, 7] model. This \textit{RRTN} (Random Resistor cum Tunneling-bond Network) driven by an ac electric field gives rise to a correlated \textit{RC} (\textit{CRC}) model because we place the capacitors only at the t-bonds whose positions are correlated to the positions of the nearest neighbor gaps between two ohmic bonds, as discussed above. For simplicity, the rest of the insulating bonds are assumed to remain inactive at any voltage however large. Thus this model effectively behaves like a three component mixture. We studied the complex ac conductance \(G(\omega)\) of our \textit{CRC} model as a function of \(\omega\) and at a fixed low voltage both by using an effective medium approximation (\textit{EMA}) and by an iterative numerical solution using Kirchoff’s laws. The real part \(\text{Re } G(\omega)\) or the modulus \(|G(\omega)|\) behaves qualitatively very similarly to that for the real, nonlinear dc conductance \(G(V)\) as a function of the external voltage (see I). Thus we fit the \(\text{Re } G(\omega)\) or the \(|G(\omega)|\) with an exponential-related function as in Eq.(14). The fitting with this function seems to be very good just like the fitting of \(G(V)\) with a similar function is optimum. But the similarity ends there.

Whereas there is only one exponent for the power-law type low voltage dc response, there are two different power-law regimes for, say \(|G(\omega)|\), much below its ac upper saturation regime. Since any realistic sample should not be able to respond to an extremely large \(\omega\), \(\text{Im } G(\omega)\) must vanish as \(\omega \to \infty\), and that does automatically happen in our \textit{CRC} model. Thus \(|G(\omega = \infty)|\) is real and finite. For non-percolating systems at very low-\(\omega\), \(|G(\omega)| \sim \omega\), whereas if percolating, \(|G(\omega)| - G(0)| \sim \omega^2\). This happens irrespective of the value of \(p\). Next for \(\omega > \omega_0\), i.e., at moderately low \(\omega\)'s (still much below the upper ac saturation regime), the system crosses over to a non-integer power law behavior with a positive exponent \(\alpha(p)\) which depends on
the value of \( p \). If we use leaky capacitors at the t-bonds, the system would percolate for all \( p \geq p_{ct} (\sim 0.18) \). For such situations, if \( p \simeq p_c \), \( \alpha \) has the minimum value of about 0.7 and this value matches with that for many experiments. Away from \( p_c \) (both for higher and lower values), with leaky capacitors at the t-bonds, \( \alpha(p) \) becomes progressively larger than 0.7. Now, if we use perfect capacitors, then for non-percolating configurations close to \( p_c \), \( \alpha(p) \simeq 0.5 \) which is significantly lower than 0.7. It may be noted that both the EMA and the traditional \( RC \) model cannot obtain anything other than the value of 0.5 for \( p \) close to \( p_c \). Further, in the case of EMA, the exponent \( \alpha(p) = 2.0 \) for any \( p \neq p_c \).

We did also look at the phase angle \( \phi(\omega) \) of the complex \( G \) at several \( p \)'s. Since \( \text{Im} \, G(\omega) \) vanishes both at very high and very low \( \omega \)'s (at any \( p \)), \( \phi(\omega) \) would also have this property as a function of \( \omega \) with at least one peak value \( (\phi_m) \) somewhere in-between. There is one curve for each \( p \) with a different peak value. In the case of EMA, \( \phi(\omega) \) cannot be calculated for any \( p < p_c \) and so in Fig. 4, we have shown them only for \( p > p_c \). For the CRC network, we do not have this pathology, and hence in the Fig. 8, we have shown it for \( 0.3 \leq p \leq 0.7 \). In general, \( \phi_m \) increases both in the EMA and our numerical solution of the CRC model as \( p \) decreases. For the latter, it is about 0.4 rad at \( p = 0.7 \) and 1.1 rad for \( p = 0.3 \). It is interesting to note that at \( p = p_c \), \( \phi_m \) takes the universal value of about \( \pi/4 \) rad in the simple \( RC \) model, numerical solution and the EMA of our CRC model. But, whereas the \( \phi_m(p_c) \) occurs at an \( \omega = 0.2 \) rad/s for the CRC model, it occurs at an \( \omega = 0.1 \) rad/s in the case of the EMA. Further the peak around \( \phi_m \) is too much broad for the EMA.

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**Figure Captions:**

**Fig. 1** Real part of the complex $G(\omega)$ for our model correlated $RC$ ($CRC$) network at $p = 0.52$, with leaky capacitors placed at the tunnel junctions. The solid line given by Eq. (3) appears to give a very good fit.

**Fig. 2** Two prototypical elementary circuits each with two ohmic resistors and one capacitor. Note that the Fig. 2(a) corresponds to a metal-like zero-frequency behavior (i.e., $G_e(\omega = 0) > 0$), whereas the elementary circuit in Fig. 2(b) corresponds to an insulator (i.e., $G_e(\omega = 0) = 0$).

**Fig. 3** The EMA result in 2D for the $CRC$ model: real part of the effective conductance $Re G_e(\omega)$ against $\omega$ for a set of values of $p$.

**Fig. 4** The EMA result for $CRC$ model in 2D: the phase-angle ($\phi$) against $\omega$ for different values of $p$.

**Fig. 5** The numerical results in 2D for the $CRC$ model with leaky capacitors with a finite real conductance $g_c > 0$: the $|< G(\omega) >|$ against $\omega$ for a set of values of $p$. For each $p$ and $\omega$, the average over 20 different configurations were taken.

**Fig. 6** The $|< G(\omega) >|$ for $p = 0.3$ and a square lattice of size $L = 20$. Just as in Fig. 5, each t-bond represented a leaky capacitor and an average over 20 configurations were taken. The inset shows an $\omega^2$-dependence up to a crossover frequency of $\omega_0 \approx 0.01$. Above $\omega_0$, the Eq. (14) gives a very good fit as shown.

**Fig. 7** Another example of $|G(\omega)|$ against $\omega$ for $p = 0.45$ and for a typical configuration on a square lattice of size $L = 20$. The very low-$\omega$ part shows a purely linear behavior. But for $\omega > \omega_0 \approx 0.01$, Eq. (14) gives the optimum fit.

**Fig. 8** Phase-angle ($\phi$) as a function of $\omega$ from the numerical solutions of our $CRC$ model.
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