Emergent Scaling Laws in Complex Dielectric Materials

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Outline

1. Modelling Complex Dielectric Materials
2. Origin of Power-Law Emergent Response
3. Results of Analytical and Numerical Approaches
Conductor-dielectric composites display *anomalous power-law* scaling in bulk AC conductivity – “Universal Dielectric Response.”

’Jonscher power law’

Emergent property of a complex system resulting from component interaction (not a resultant property)
Microstructure of a Composite

- $\text{Al}_2\text{O}_3 - \text{TiO}_2$
- Variable conductivity ratio (with AC driving frequency $\omega$).

R. Uppal & R. Stevens
Modelling of Complex Composites

- $\text{Al}_2\text{O}_3 - \text{TiO}_2$
- Associate conducting phase with $R$ and dielectric with $C$. 

![Diagram showing the composition of the composite material with labels for $R_{\text{TiO}_2}$ and $C_{\text{Al}_2\text{O}_3}$ at 10 $\mu$m scale.]}
Modelling of Complex Composites

- Model using resistor-capacitor network:
  - Randomly assign bonds on square lattice as either $R \left( y_R = R^{-1} \right)$ or $C \left( y_C = i\omega C \right)$.

  \begin{align*}
  N & \text{: Total number of components} \\
  p & \text{: proportion of } C \\
  h & \text{: } i\omega CR \text{ conductivity ratio}
  \end{align*}

Vainas and Almond, 1999
Response of Networks

- Random percolation
- \[ \text{conductivity} \approx \text{frequency}^p \]
Response of Networks

**Conductivity**

**Frequency**

Random percolation

\[ \text{conductivity} \approx \text{frequency}^\beta \]
Response of Networks

Frequency

conductivity \approx \text{frequency}^p

Random percolation
Response of Networks

Frequency

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Random percolation
Response of Networks

Frequency

conductivity \approx frequency^p

Random percolation
Related to Percolation Theory\(^1\)

**Critical system, as \( p \to p_c \):**

- Infinite system;
  - Correlation length:
    \[ \xi(p) \propto |p - p_c|^{-\nu}. \]
  - Average cluster size:
    \[ \chi(p) \propto |p - p_c|^{-\gamma}. \]

**Phase transition** at \( p = p_c \).

In 2D square lattice: \( p_c = 0.5, \ldots \)

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Response of Networks

- Power $n \approx p$, proportion of variable components (capacitors).

Experimentally verified.

See: Almond and Bowen, 2004
Scaling with the network size at $\rho = 0.5 = \rho_c$
Scaling with the network size at $\rho = 0.4 < \rho_c$
Scale variation as a function of $p$ and $N$

- If $p = p_c = 0.5$ then $\max |Y| / \min |Y| \sim N$
- If $p < p_c$ then
  - $\max |Y| / \min |Y| \sim N$ for small $N$
  - $\max |Y| / \min |Y| \sim C(p)$ for large $N$
  - $C(p) \rightarrow \infty$ as $p \rightarrow p_c$. 
Analytic Explanation for the Origin of the Power-Law Emergent Response

Features of PLER:

1. Admittance $|Y| \propto \omega^n$, $n \approx p$ over several orders of magnitude.

2. $|Y(\omega)|$ independent of details (statistical properties).

3. Percolation limits & width of region can depend strongly on network size $N$ if $p = p_c$ and weakly otherwise.
Matrix Representation of Electrical Networks

Using Kirchhoff’s laws:

\[
\begin{pmatrix}
\Sigma_2 & -y_{2,3} \\
-y_{2,3} & \Sigma_3
\end{pmatrix}
\begin{pmatrix}
v_2 \\
v_3
\end{pmatrix}
=
\begin{pmatrix}
y_{1,2} \\
y_{1,3}
\end{pmatrix}
V
\]

\[
\Sigma_2 = y_{1,2} + y_{2,3} + y_{2,4}
\]

\[
\Sigma_3 = y_{1,3} + y_{2,3} + y_{3,4}
\]

\[
v_1 = V, \quad v_4 = 0, \quad y_{m,n} = 1/z_{m,n}
\]

- Problem reduces to solving:

\[
Kv = bV
\]

- **K** sparse banded (Laplacian) matrix of admittances,
- **v** vector of node voltages,
- **b** vector of boundary elements.
- **V** applied boundary potential.
The Power-Law Emergent Response

- Admittance $Y(\omega) = b^T K^{-1} b$
- $K = K_R + i\omega K_C$

Emergent power-law response over wide range of $\omega$. 
Poles and Zeroes of the Transfer Function

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- $K = K_R + i\omega K_C$
Poles and Zeroes of the Transfer Function

- Admittance $Y(\omega) = b^T K^{-1} b$
  - $K = K_R + i\omega K_C$

- Rational function: $Y(\omega) = \frac{N(\omega)}{D(\omega)} = \frac{F}{(\omega - \omega_z,1)(\omega - \omega_z,2)(\omega - \omega_z,3)\ldots}{(\omega - \omega_p,1)(\omega - \omega_p,2)(\omega - \omega_p,3)\ldots}$
Poles and Zeroes of the Transfer Function

- Admittance \( Y(\omega) = b^T K^{-1} b \)
  - \( K = K_R + i\omega K_C \)

- Rational function: \( Y(\omega) = \frac{N(\omega)}{D(\omega)} = \frac{F(\omega-\omega_{z,1})(\omega-\omega_{z,2})(\omega-\omega_{z,3})\ldots}{(\omega-\omega_{p,1})(\omega-\omega_{p,2})(\omega-\omega_{p,3})\ldots} \)

- Poles \( \omega_{p,k} \) are the finite generalised eigenvalues of \( K \).
- Zeros \( \omega_{z,k} \) are the finite generalised eigenvalues of a symmetric block-bordered extension of \( K \).
Poles and Zeroes of the Transfer Function

- Admittance $Y(\omega) = b^T K^{-1} b$
  - $K = K_R + i\omega K_C$

- Rational function: $Y(\omega) = \frac{N(\omega)}{D(\omega)} = F_{\frac{\omega-\omega_{z,1}}{(\omega-\omega_{p,1})(\omega-\omega_{p,2})(\omega-\omega_{p,3})...}}$

- Poles $\omega_{p,k}$ are the finite generalised eigenvalues of $K$.
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- Study distributions of Zeroes, Poles and statistics of spacings between them.
Large RC Electrical Networks.

Mathematically it can be shown that:

1. Poles at $iW_{p,k}$ and Zeroes at $iW_{z,k}$ are pure imaginary.
2. $W_{p,k}, W_{z,k} > 0.$
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Boundaries of PLER
Observations on P,Z Distributions

From analysis of large number of networks:

- Poles and Zeroes interlace, as predicted.
- Find a symmetric log-Normal distribution of the Zeroes & Poles.
Observations on Pole-Zero Spacings

• Spacings are statistically regular

• For $p = 0.5$:

→ Mean (over $i$) spacings equal

$$W_{p,i} - W_{z,i} = W_{z,i} - W_{p,(i-1)}$$
Observations on Pole-Zero Spacings

- Spacings are statistically regular

- For $p = 0.5$:

- For $p \neq 0.5$ ($p = 0.4$):

$\rightarrow$ Mean (over $i$) spacings equal

\[ W_{p,i} - W_{Z,i} = W_{Z,i} - W_{p,(i-1)} \]
Regularity of the pole-zero spacings over several realisations

Let

$$\bar{\delta}_i(p) = \frac{W_{z,i} - W_{p,i}}{W_{p,i+1} - W_{p,i}}$$

averaged over many network realisations.

Observe

- $\bar{\delta}_i(0.5) \approx 0.5$
- $\bar{\delta}_i(p) \approx \bar{\delta}_{N-i}(p)$
- $\bar{\delta}_i(p) + \bar{\delta}_i(1 - p) \approx 1$
- $\text{mean}_i \bar{\delta}_i(p) \approx p$
Range of Pole-Zero Values

- Smallest and Largest Value:
  - For the $p = p_c = 0.5$ case:
    - $W_{p/z, 1} \sim 1/NCR$,
    - $W_{p/z, N} \sim N/CR$
Range of Pole-Zero Values

- **Smallest and Largest Value:**
  - For the $p = p_c = 0.5$ case:
    - $W_{p/z,1} \sim 1/NCR$, $W_{p/z,N} \sim N/CR$
  - Slopes over range of $p$:
    - $W_{p/z,1} \sim 1/N^\alpha CR$, $W_{p/z,N} \sim N^\alpha/CR$, $\alpha \leq 1$
Derivation for Random RC Networks

\[ |Y(\omega, N)| = |g(N)| \frac{\prod_{k=1}^{N} |\omega - iW_{z,k}|}{\prod_{k=1}^{N} |\omega - iW_{p,k}|} \]
Derivation for Random RC Networks

\[ |Y(\omega, N)| = |g(N)| \frac{\prod_{k=1}^{N} |\omega - iW_{z,k}|}{\prod_{k=1}^{N} |\omega - iW_{p,k}|} \]

but: \[ |\omega - iW_{[p,z],k}| = \sqrt{Re(\omega)^2 + W_{[p,z],k}^2} \]
Assuming equal numbers of finite $P,Z$:

$$|Y(\omega, N)| = |g(N)| \prod_{k=1}^{N} \sqrt{\frac{\omega^2 + W_{z,k}^2}{\omega^2 + W_{p,k}^2}}$$
Assuming equal numbers of finite $P,Z$:

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using previous observations of distribution of $P,Z$:

$W_{p,k} \sim f(k), W_{z,k} \sim f(k) - \bar{\delta}_k f'(k)$
Derivation for Random RC Networks

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- using previous observations of distribution of $P, Z$:

$$W_{p,k} \sim f(k), W_{z,k} \sim f(k) - \bar{\delta}_k f'(k)$$

- we obtain:

$$\log (|Y(\omega, N)|) = \log (|g(N)|) + \frac{1}{2} \sum_{k=1}^{N} \log \left( \frac{\omega^2 + (f(k) - \bar{\delta}_k f'(k))^2}{\omega^2 + (f(k))^2} \right)$$
Derivation for Random RC Networks

- Assuming equal numbers of finite P, Z:
  \[ |Y(\omega, N)| = |g(N)| \prod_{k=1}^{N} \sqrt{\frac{\omega^2 + W_{z,k}^2}{\omega^2 + W_{p,k}^2}} \]

- using previous observations of distribution of P, Z:
  \[ W_{p,k} \sim f(k), W_{z,k} \sim f(k) - \bar{\delta}_k f'(k) \]

- we obtain:
  \[ \log(|Y(\omega, N)|) = \log(|g(N)|) + \frac{1}{2} \sum_{k=1}^{N} \log \left( \frac{\omega^2 + (f(k) - \bar{\delta}_k f'(k))^2}{\omega^2 + f(k)^2} \right) \]

- and a few approximations later...
Results for Random RC Networks.

Obtain following expressions with

\[ \bar{\delta} = \text{mean}_{\log(w_i)}(\delta_i). \]

1. Percolation path in R but not C:

\[ |Y(\omega)| = \frac{1}{R} \left( \frac{1 + N^2 C^2 R^2 \omega^2}{N^2 + C^2 R^2 \omega^2} \right)^{\frac{\bar{\delta}}{2}} \]

2. Percolation path in C but not R:

\[ |Y(\omega)| = \omega C \left( \frac{N^2 + C^2 R^2 \omega^2}{1 + N^2 C^2 R^2 \omega^2} \right)^{\frac{1 - \bar{\delta}}{2}} \]

Numerical results for \( p = 0.5 \) for which \( \bar{\delta} = 0.5 \)
Results for Random RC Networks.

- Obtain following expressions with $\bar{\delta} = \text{mean}_{\log(W_i)}(\bar{\delta}_i)$.

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Numerical results for $\rho = 0.5$ for which $\bar{\delta} = 0.5$

- Small Networks:

![Graph showing the relationship between $\bar{\delta}$ and $\omega$ for small networks.](image)
Results for Random RC Networks.

- Obtain following expressions with \( \bar{\delta} = \text{mean}_{\log(w)}(\bar{\delta_i}) \).

1. Percolation path in R but not C:

\[
|Y(\omega)| = \frac{1}{R} \left( \frac{1 + N^2 C^2 R^2 \omega^2}{N^2 + C^2 R^2 \omega^2} \right)^{\frac{\bar{\delta}}{2}}
\]

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\]

Numerical results for \( \rho = 0.5 \) for which \( \bar{\delta} = 0.5 \)

- Small Networks:
- Large Networks:
Summary

- Binary random systems show Power-Law Emergent Response. (Models UDR in solids)
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- Response model give continuous distribution of RC relaxation rates.
  - Possible links to models of relaxation processes.