The Origin of Emergent Scaling Laws in Complex Dielectric Materials.

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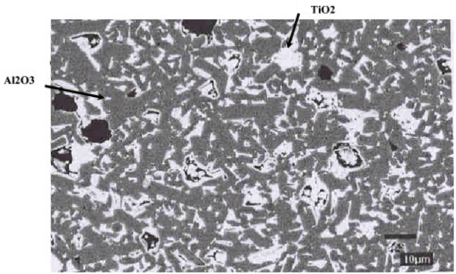
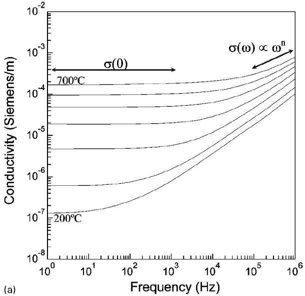
10th April

Microstructure of a Ceramic

- $Al_2O_3 - TiO_2$
- Variable conductivity ratio (with AC driving frequency ω).

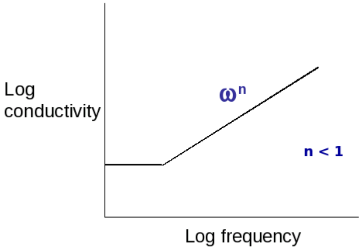




R. Uppal & R. Stevens

Bulk Response of Composites

- Conductor-dielectric composites display *anomalous power-law* scaling in bulk AC conductivity – "**Universal Dielectric Response**."
- 'Jonscher power-law'



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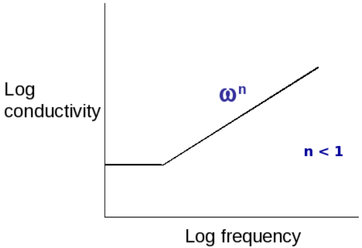
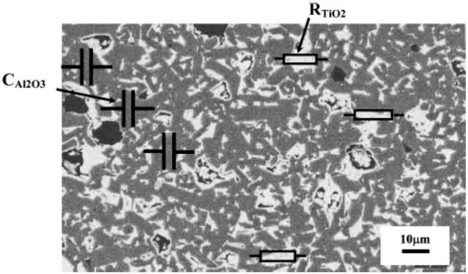


- Emergent property of a complex system resulting from component interaction.

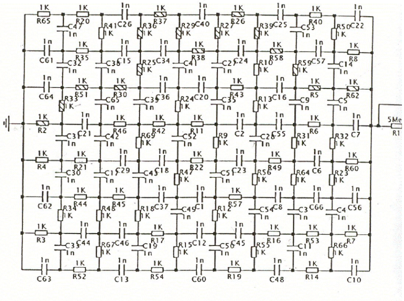
Modelling of Complex Composites

- $Al_2O_3 - TiO_2$
- Associate conducting phase with R and dielectric with C.



Modelling of Complex Composites

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



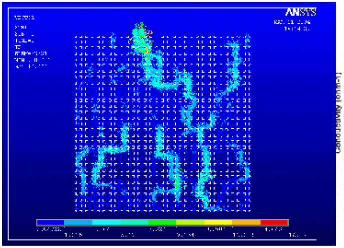
N: Total number of components

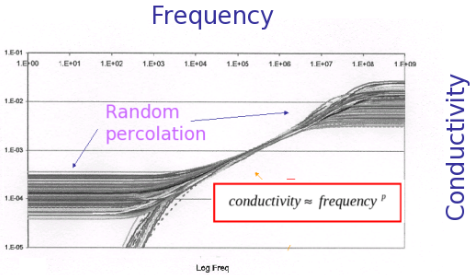
p: proportion of C

h: $i\omega CR$ conductivity ratio

Vainas and Almond, 1999

Response of Networks





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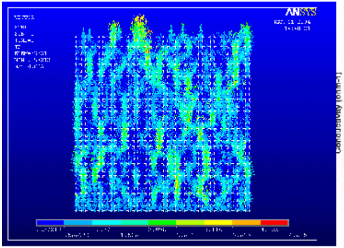

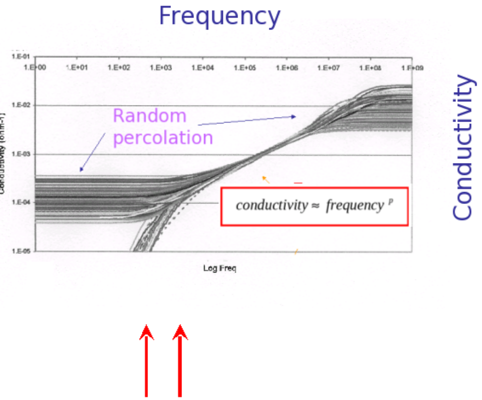

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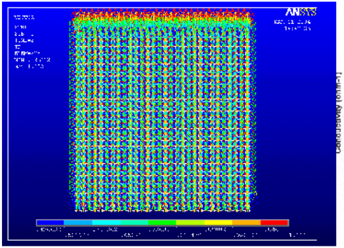

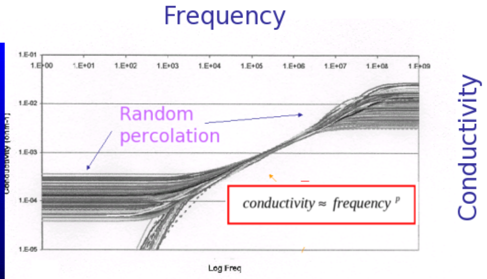

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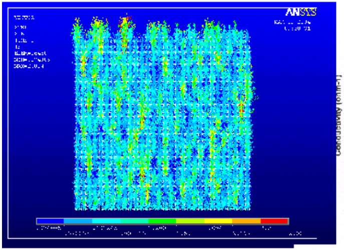

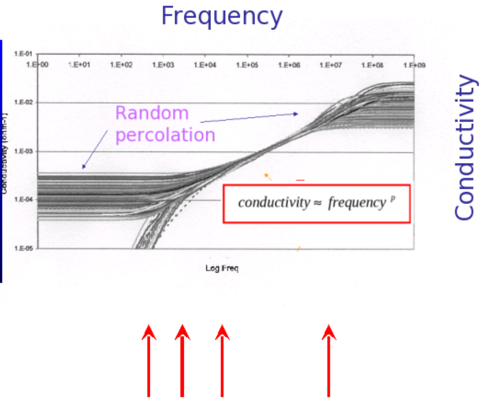

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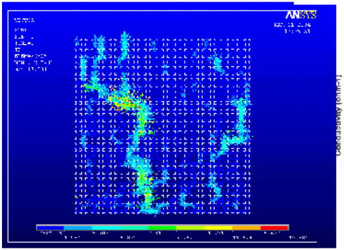
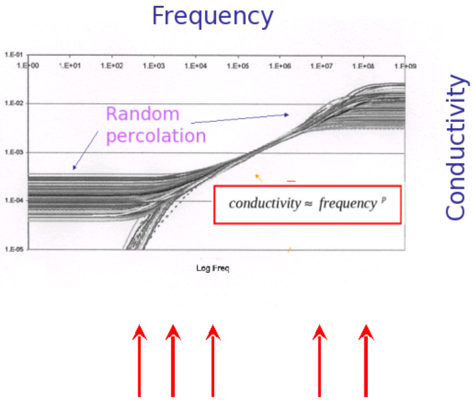
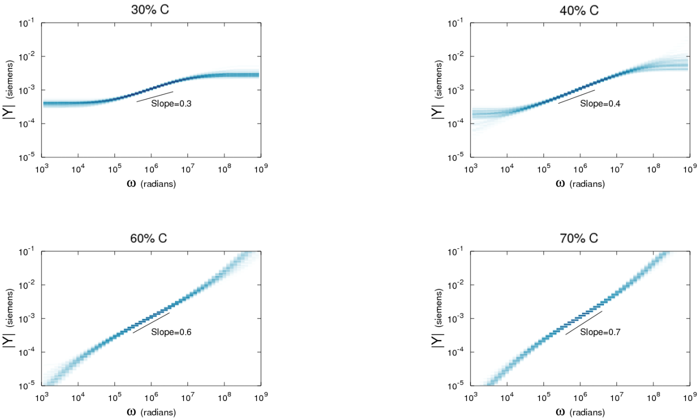




Response of Networks

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

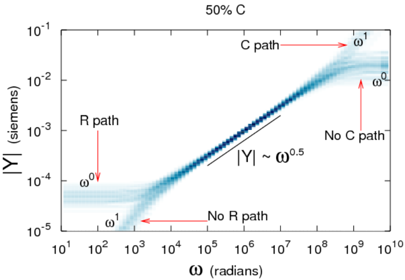


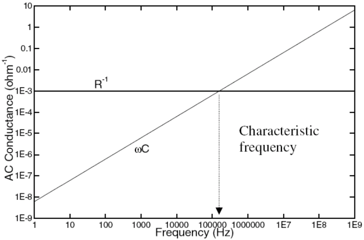
- Experimentally verified.

See: *Almond and Bowen, 2004*

Power-Law Emergent Response

- Emergent power-law response over wide range of ω.

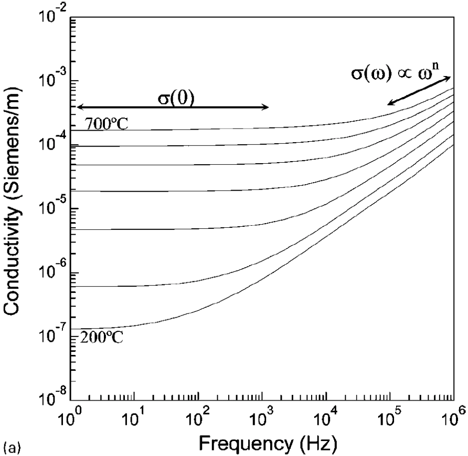




- $p = p_c = \frac{1}{2}$: critical *percolation threshold* for 2D square lattices.

Physical Interpretation

- Typical frequency response of a real material:



- conductivity increasing with frequency:

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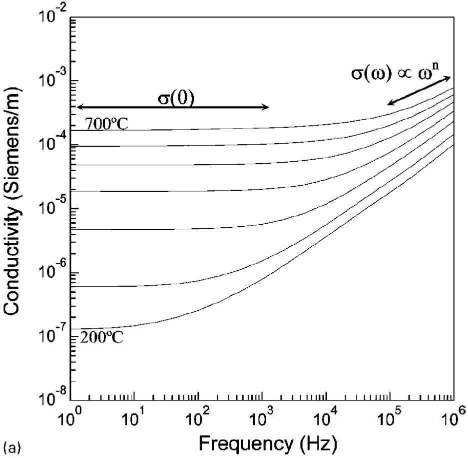
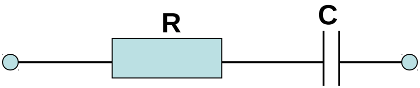
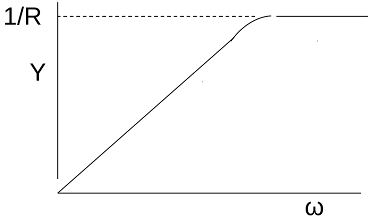
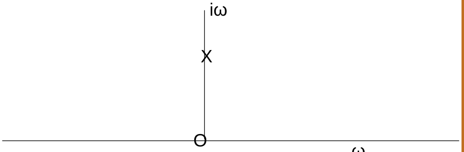


- conductivity increasing with frequency:
- material behaving as a "high-pass filter".

Simple High-Pass Filter



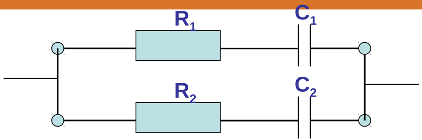




POLE ZERO DIAGRAM

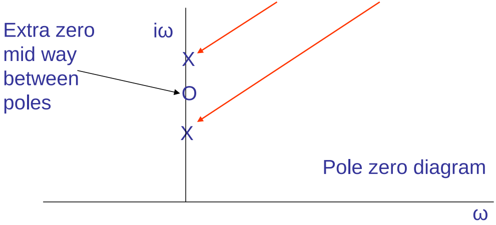
- $Y(\omega) = \dfrac{i\omega C}{1 + i\omega CR}$
- Pole at $\omega = i/CR$
- Zero at $i\omega = 0$

Parallel Filters



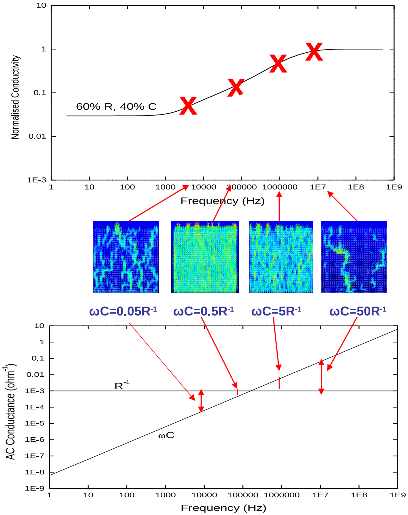
- Admittance:

$$Y(\omega) = Y_1 + Y_2 = i\omega C_1/(1 + i\omega C_1 R_1) + i\omega C_2/(1 + i\omega C_2 R_2)$$
$$= \frac{i\omega C_1(1 + i\omega C_2 R_2) + i\omega C_2(1 + i\omega C_1 R_1)}{(1 + i\omega C_1 R_1)(1 + i\omega C_2 R_2)}$$



Large RC Network Response

TYPICAL LARGE R-C NETWORK RESPONSE



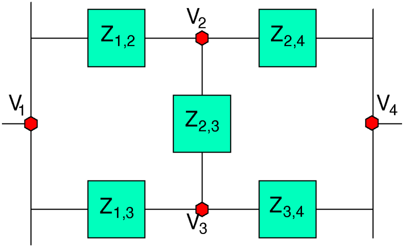
- Net response from:
  - many conduction paths in parallel,
  - Equivalent to a large number of high pass filters with a random distribution of Rs and Cs in parallel.

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 $p = p_C = 1/2$.
  4. If $p \neq 1/2$ percolation limits depend only on p if N is sufficiently large.

Matrices of Electrical Networks



Using Kirchhoff's laws:

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

$$\Sigma_2 = y_{1,2} + y_{2,3} + y_{2,4}$$
$$\Sigma_3 = y_{1,3} + y_{2,3} + y_{3,4}$$
$$v_1 = V, v_4 = 0, y_{m,n} = 1/z_{m,n}$$

- Problem reduces to solving:

$$K\underline{\mathrm{v}} = \underline{\mathrm{b}}V$$

K sparse banded (Kirchhoff) matrix of admittances,
$\underline{\mathbf{v}}$ vector of node voltages,
$\underline{\mathbf{b}}$ vector of boundary elements.
V applied boundary potential.

Poles and Zeroes

- Admittance $Y(\omega) = \underline{b}^T \mathbf{K}^{-1} \underline{b}$
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- Rational function: $Y(\omega) = \frac{N(\omega)}{D(\omega)} = F \frac{(\omega-\omega_{z,1})(\omega-\omega_{z,2})(\omega-\omega_{z,3})\ldots}{(\omega-\omega_{p,1})(\omega-\omega_{p,2})(\omega-\omega_{p,3})\ldots}$.
  - Poles $\omega_{p,k}$ are the finite generalised eigenvalues of $\mathbf{K}$.
  - Zeroes $\omega_{z,k}$ are the finite generalised eigenvalues of a symmetric block-bordered extension of $\mathbf{K}$.

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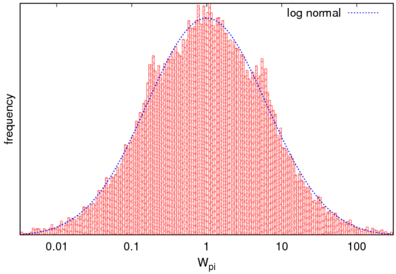
- Study distributions of Zeroes, Poles and statistics of spacing between them.

Observations on P, Z Distributions

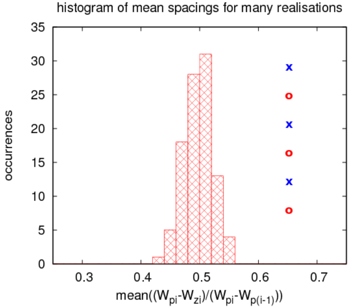
From analysis of large number of networks:

- Poles and Zeroes interlace, as predicted.
- Find a symmetric log-Normal distribution of Zeroes & Poles.



Observations on P–Z Spacings

- Spacings are statistically regular
  - For $p = 0.5$:



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- $\overline{W_{p,k} - W_{z.k}} = \overline{W_{z.k} - W_{p,(k-1)}}$

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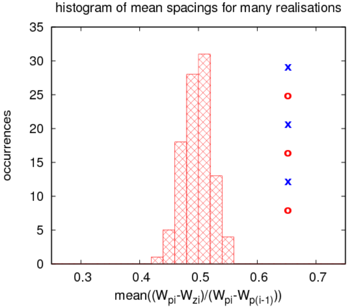

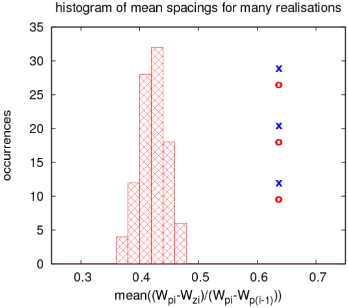

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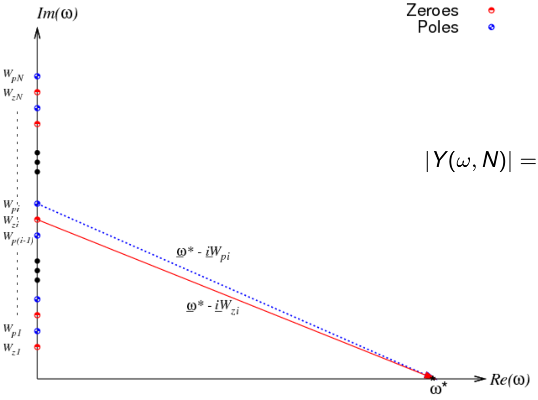
Derivation for Random RC Networks



$$|Y(\omega, N)| = |g(N)| \frac{\prod_{k=1}^{N} |\underline{\omega} - \underline{iW}_{z,k}|}{\prod_{k=1}^{N} |\underline{\omega} - \underline{iW}_{p,k}|}$$

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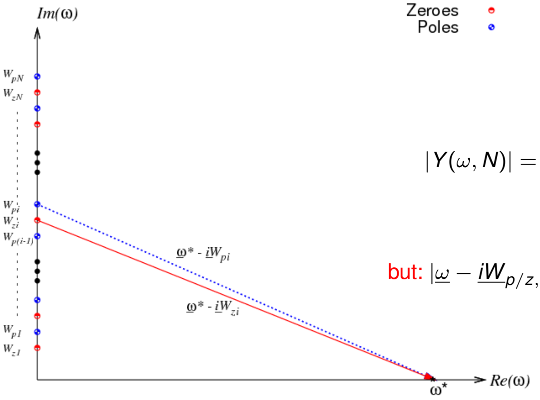

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but: $|\underline{\omega} - \underline{iW}_{p/z,k}| = \sqrt{Re(\omega)^2 + W_{p/z,k}^2}$

Derivation for Random RC Networks

- Assuming equal numbers of finite P, Z:

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

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- and a few approximations later...

Results for Random RC Networks.

- Obtain following expressions with $\bar{\bar{d}} = \text{mean}_{\{\log(w_i)\}}(\bar{d}_k)$:

(1) Percolation path in R but not C:

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

(2) Percolation path in C but not R:

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

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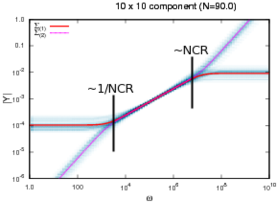
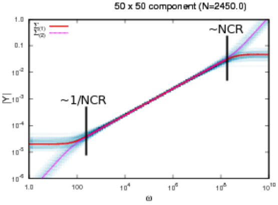
Numerical results for $p = 0.5$ for which $\bar{\bar{d}} = 0.5$:

- Small Networks:



- Large Networks:



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- Almond, Budd, Freitag, Hunt, McCullen & Smith, *Physica A* (2013).