Emergent Scaling Laws in Complex Dielectric Materials

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UNIVERSITY OF BATH

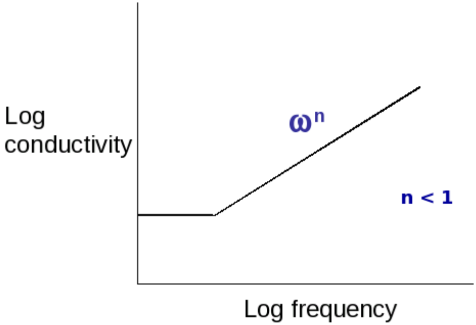
BICS

EPSRC
Engineering and Physical Sciences Research Council

Outline

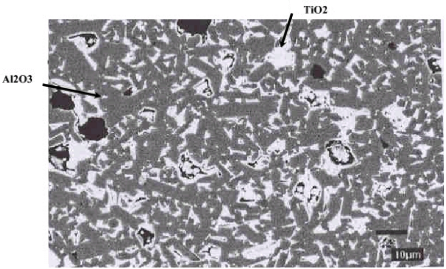
Bulk Response of Composite Materials

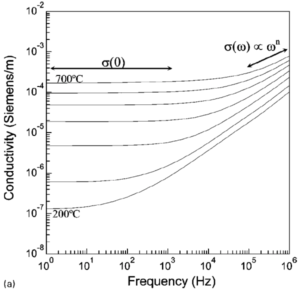
- Conductor-dielectric composites display *anomalous power-law* scaling in bulk AC conductivity – "**Universal Dielectric Response**."
- 'Jonscher power law'
- Emergent property of a complex system resulting from component interaction (not a resultant property)



Microstructure of a Composite

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

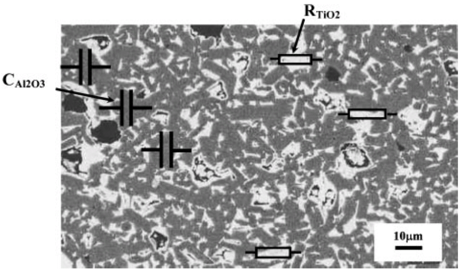




R. Uppal & R. Stevens

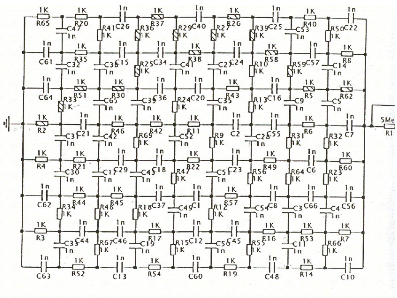
Modelling of Complex Composites

- $Al_2O_3 - TiO_2$
- 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($y_C = i\omega C$).



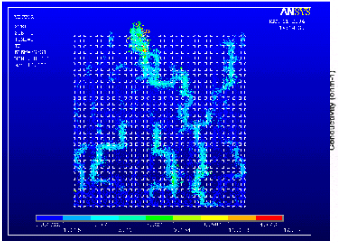
N: Total number of components

p: proportion of C

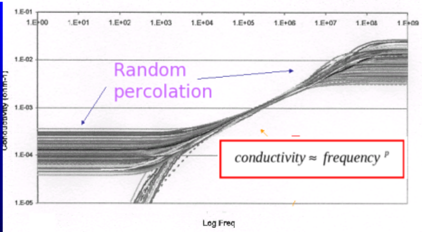
h: $i\omega CR$ conductivity ratio

Vainas and Almond, 1999

Response of Networks

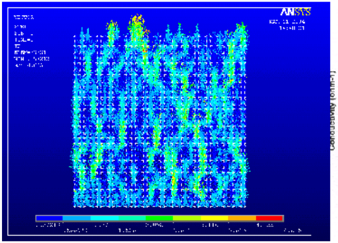


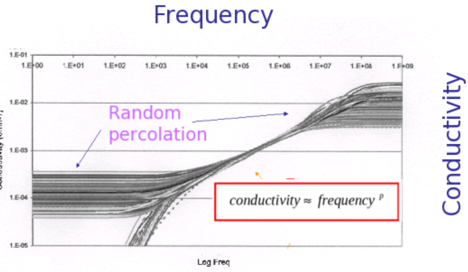
Frequency



Conductivity

Response of Networks





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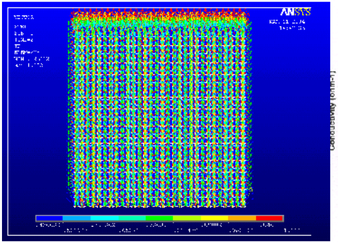

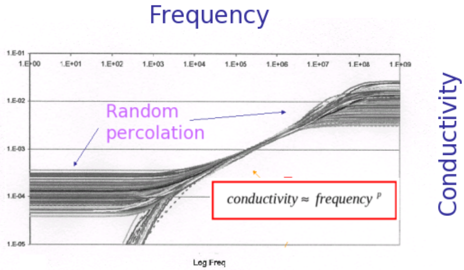

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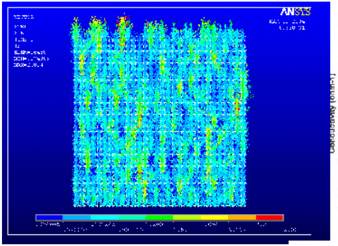

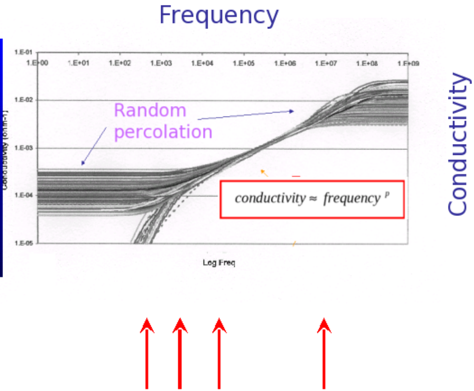

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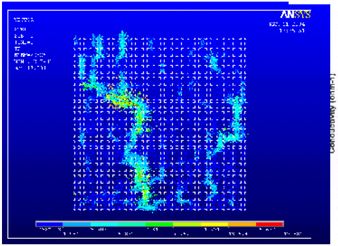
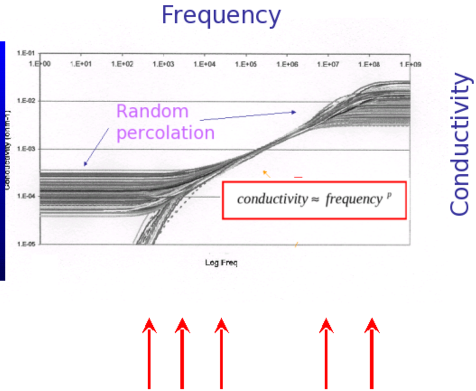
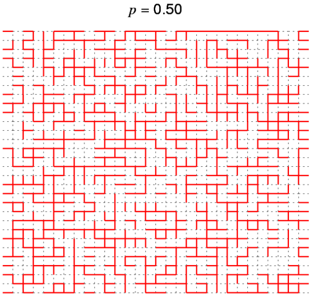




Related to Percolation Theory[1]



Critical system, as $p \to p_c$:
Infinite system;

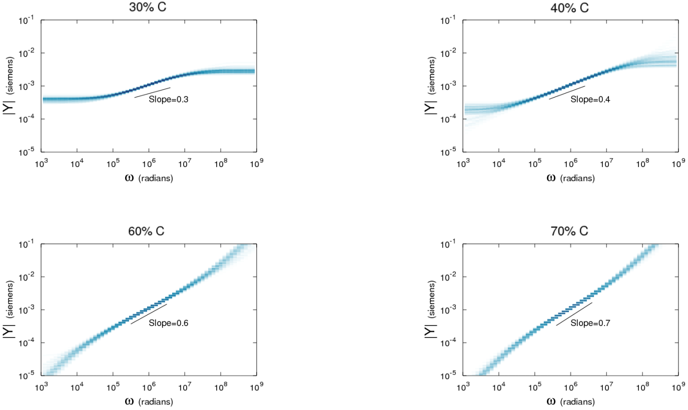
- Correlation length: $\xi(p) \propto |p - p_c|^{-\nu}$.
- Average cluster size: $\chi(p) \propto |p - p_c|^{-\gamma}$.

Phase transition at $p = p_c$.
In 2D square lattice: $p_c = 0.5, \ldots$

[1] S. R. Broadbent and J. M. Hammersley, *Percolation processes. I, II* Proc. Cambridge Philos. Soc. (1953)

Response of Networks

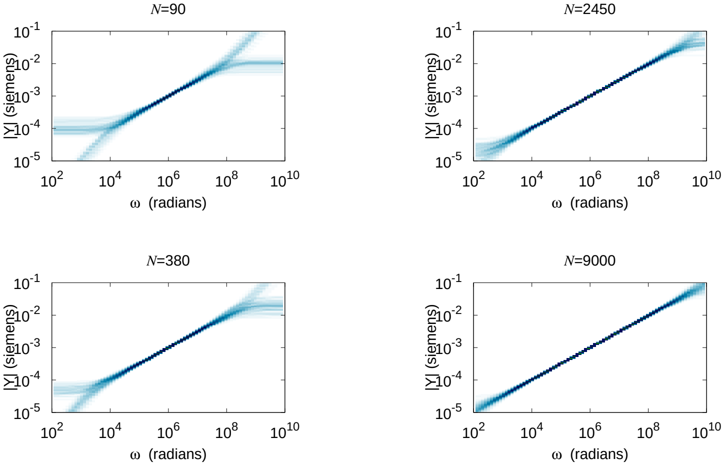
- Power $n \approx p$, proportion of variable components (capacitors).



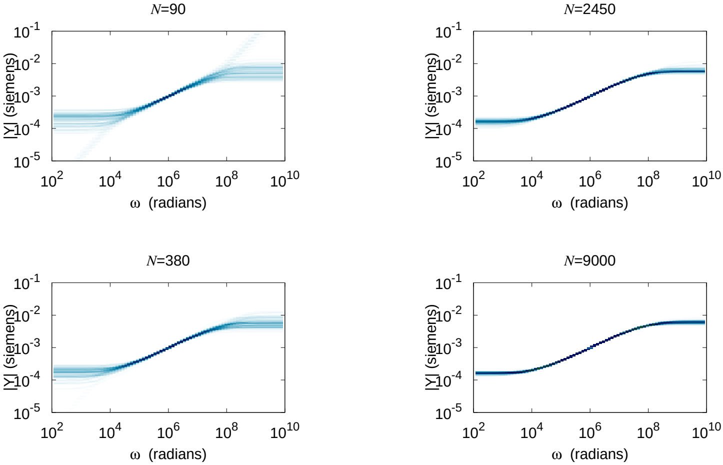
- Experimentally verified.

See: *Almond and Bowen, 2004*

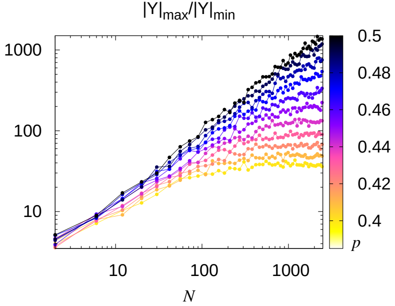
Scaling with the network size at $p = 0.5 = p_c$



Scaling with the network size at $p = 0.4 < p_c$



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
  - $C(p) \to \infty$ as $p \to p_c$.

Analytic Explanation for the Origin of the Power-Law Emergent Response

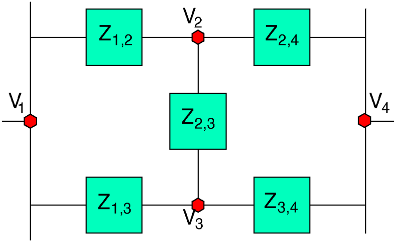
- Features of PLER:

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:

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

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

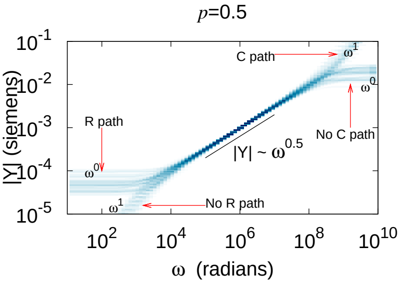
- Problem reduces to solving:

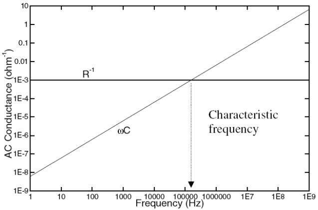
$$\mathbf{K}\underline{v} = \underline{b}V$$

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

The Power-Law Emergent Response

- Admittance $Y(\omega) = \underline{b}^T \mathbf{K}^{-1} \underline{b}$
- $\mathbf{K} = \mathbf{K}_R + i\omega \mathbf{K}_C$





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

Poles and Zeroes of the Transfer Function

- Admittance $Y(\omega) = \underline{b}^T \mathbf{K}^{-1} \underline{b}$
  - $\mathbf{K} = \mathbf{K}_R + i\omega \mathbf{K}_C$

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

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  - Zeros $\omega_{z,k}$ are the finite generalised eigenvalues of a symmetric block-bordered extension of $\mathbf{K}$.

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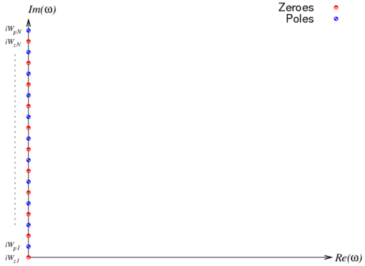
  - Poles $\omega_{p,k}$ are the finite generalised eigenvalues of K.
  - Zeros $\omega_{z,k}$ are the finite generalised eigenvalues of a symmetric block-bordered extension of $\mathbf{K}$.
- Study distributions of Zeroes, Poles and statistics of spacings between them.

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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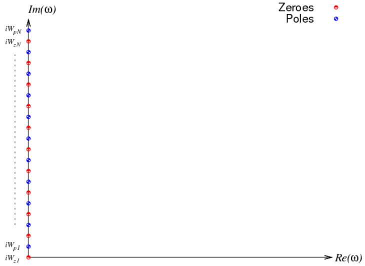

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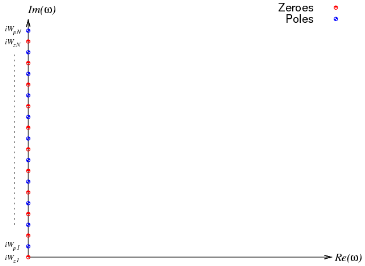
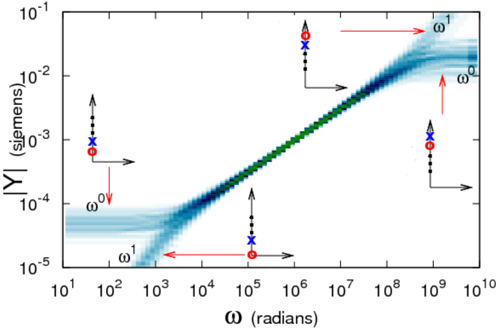
4. Boundaries of P,Z set correspond to transition between PLER and percolation/saturation.



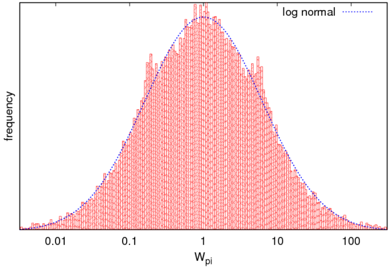
Boundaries of PLER



Observations on P,Z Distributions

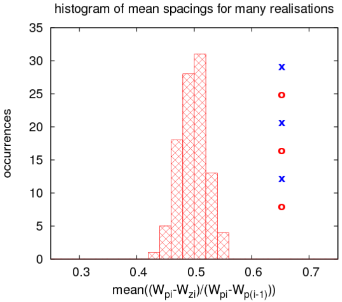
From analysis of large number of networks:

- 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

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

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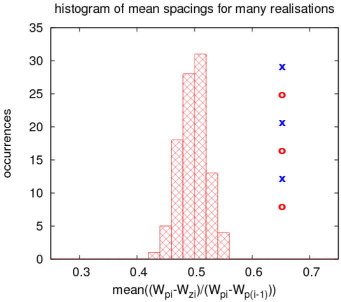
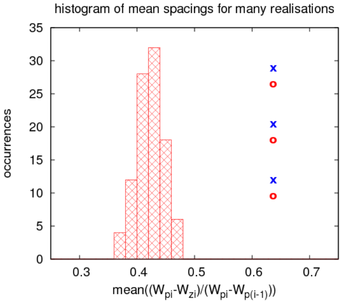


  - For $p \neq 0.5$ ($p = 0.4$):



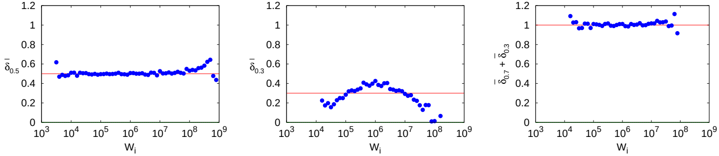
$\longrightarrow$ Mean (over i) spacings equal

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

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.

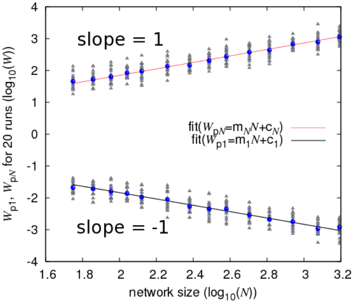


Observe

- $\bar{\delta}_i(0.5) \approx 0.5$,
- $\bar{\delta}_i(p) \approx \bar{\delta}_{N-i}(p)$,
- $\bar{\delta}_i(p) + \bar{\delta}_i(1-p) \approx 1$,
- $\text{mean}_i\ \bar{\delta}_i(p) \approx p$.

Range of Pole-Zero Values

- Smallest and Largest Value:
  - For the $p = p_c = 0.5$ case:



$\rightarrow$ $W_{p/z,1} \sim 1/NCR$, $W_{p/z,N} \sim N/CR$

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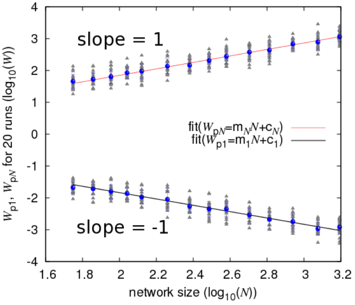

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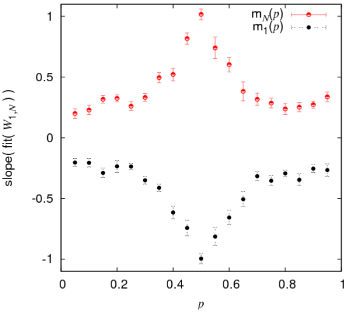
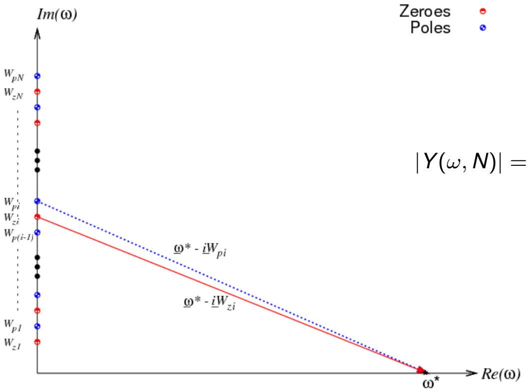
  - Slopes over range of p:



$\rightarrow$ $W_{p/z,1} \sim 1/N^{\alpha}CR$,
$W_{p/z,N} \sim N^{\alpha}/CR$, $\alpha \leq 1$

Derivation for Random RC Networks



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

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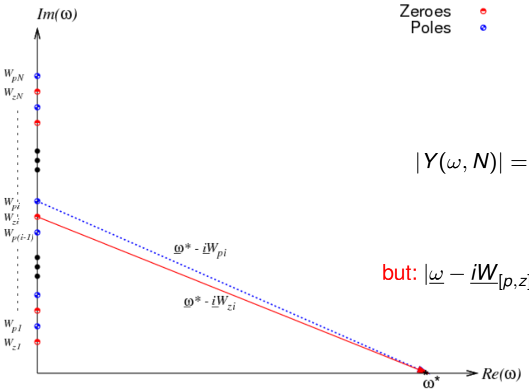

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but: $|\underline{\omega} - \underline{iW}_{[p,z],k}| = \sqrt{Re(\omega)^2 + W_{[p,z],k}^2}$

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 $\bar{\bar{\delta}} = \text{mean}_{\log(w_i)}\left(\bar{\delta}_i\right)$.

(1) Percolation path in R but not C:

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

(2) Percolation path in C but not R:

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

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

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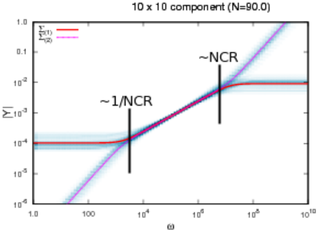
- Small Networks:



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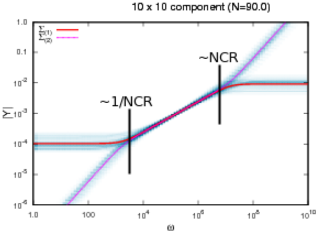
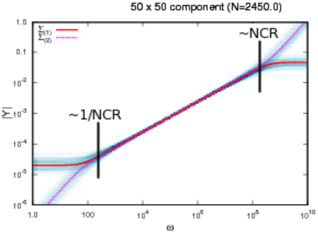
$$|Y(\omega)| = \omega C\left(\frac{N^2 + C^2R^2\omega^2}{1 + N^2C^2R^2\omega^2}\right)^{\frac{1-\bar{\bar{\delta}}}{2}}$$

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

- Small Networks:



- Large Networks:



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- Response model give continuous distribution of RC relaxation rates.
  - Possible links to models of relaxation processes.