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# The robustness of the emergent scaling property of random $RC$ network models of complex materials

**N J McCullen, D P Almond, C J Budd and G W Hunt**

Bath Institute for Complex Systems, University of Bath, Bath BA2 7AY, UK

E-mail: n.mccullen@bath.ac.uk

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**Abstract**

The so-called 'universal dielectric response' of composite materials can be reproduced as a power-law emergent response (PLER) of electrical network models. Results are presented for investigations demonstrating the robustness of the PLER of random electrical networks in order to evaluate the usefulness of such models in simulating real composite materials with microstructural disorder. The effect of imposed microstructures has been investigated, looking at both the correlation length and the network size. It is shown that the exact microstructural details may be reasonably omitted, so long as we take care that the general features of the structures, such as their relative smallest and largest scales, are represented.

Anisotropy in the random microstructures is shown to alter the bulk response of the system, with the network responses found to tend towards that expected for purely parallel and series components. The power-law response is shown to be obtainable by taking the geometric mean of the two cases, showing that the bulk response of such systems is an averaged property of these two extreme cases.

It is concluded that, given the longer computing times needed to simulate these more realistic representations, it is reasonable to use the simpler models.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

### *1.1. The importance of composites*

Composite materials are of increasing importance in science and engineering. New materials have to be designed with tailor-made properties to suit a host of demands and meet the many challenges ahead. In coming years advanced energy technologies will require new materials to perform to exact specifications in extreme circumstances.

Composites can exhibit surprising features in their overall response when compared with that of their constituents. This is in common with many other diverse physical systems under the umbrella of *complex systems*, where interactions between many components give rise to new phenomena at the system level [1]. These so-called *emergent properties* can rarely be predicted by simply knowing the behaviour of the individual components as the interaction between them is a defining factor. As such, the microstructure of the composite is crucial to the combined behaviour.

Of particular importance is the understanding of transport properties such as the conduction of energy or fluid, especially in disordered materials. Conduction through random media has applications throughout many disciplines. For example, the Earth's crust is composed of a complex mixture of rock types and understanding the propagation of seismic waves through this medium is of importance to geophysical studies. Similarly, the electrical or thermal conductivity of binary media with different microstructures is important when engineering new composites. A comprehensive review of the historical development of the study of binary disordered dielectric systems can be found in [2].

 

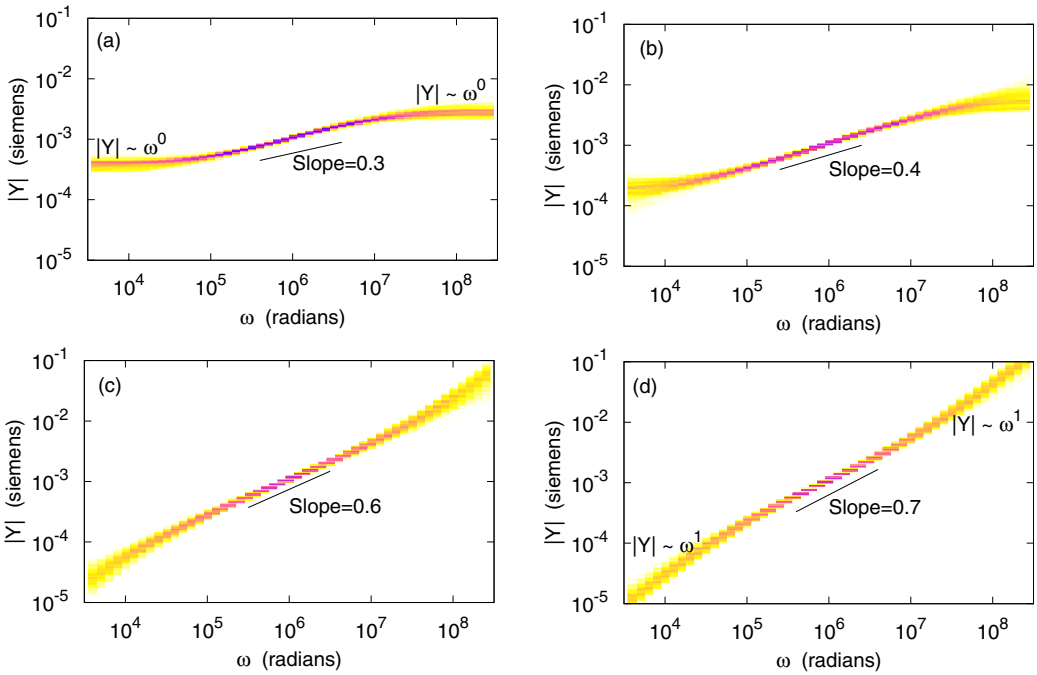


**Figure 1.** Network admittances ($|Y|$) for 20 × 20 node resistor–capacitor network simulations of complex materials (using the network methods described in section 2.1), over a range of the proportion of capacitors: (*a*) 30%, (*b*) 40%, (*c*) 60% and (*d*) 70%. Below the critical percolation value of 50% the responses at low and high angular driving frequency ($\omega$) are determined by the existence of conductive percolation paths and absence of capacitive paths between the terminals, respectively. Similarly, above 50% the limiting response at low and high $\omega$ is determined by the existence of capacitive and absence of conductive paths, respectively. For intermediate values of $\omega$ a power-law response is seen with an exponent close to the proportion of capacitors in the network. Histograms of 100 random realizations are given in this and all subsequent figures.

### *1.2. The universal dielectric response*

The anomalous power-law frequency dependence of the dielectric properties on the AC conductivity of materials [3–5] is a well-known longstanding problem. The majority of theoretical treatments of this effect [6–10] suggest it to arise from intricate, atomic level, interactions between particles within the materials manifesting this phenomenon. We have presented the alternative suggestion [11–16] that the anomalous effects are merely the electrical response characteristics of the two-phase, conductor–insulator, networks formed by the microstructures of heterogeneous/composite materials. It has been suggested that such microstructures can be modelled as large networks of randomly positioned resistors and capacitors (detailed in section 2.1). The electrical characteristics of these networks are found to closely resemble those of the large body of materials that exhibit the anomalous power law frequency dependences of permittivity or AC conductivity. We suggested in [14] that these power-law frequencies dependences were emergent properties of the complex electrical networks formed by the microstructures of heterogeneous materials. The purpose of this paper is to demonstrate the robustness of these emergent properties to variations in the components and their distribution within large electrical networks.

### *1.3. Response characteristics*

In order to demonstrate the various features of the observed responses, we show here some recalculations of results of earlier work from our group. A selection of simulation results using the network methods described in section 2.1 are given in figure 1. The magnitude of the network admittances $|Y|$ versus the angular frequency of the driving AC field ($\omega$) are plotted on a log scale over a range of values of the proportion of the variable (dielectric) inclusions. The networks used for these simulations have the same geometry as seen in later sections (specifically 20 × 20 node *RC* networks, with bonds assigned randomly according to a binary distribution), similar to those shown in later figures (e.g. figure 5(*a*)).

For under the critical value of 50% capacitors, there is a finite chance of a conductive path through the medium but little chance of a percolating path of capacitors. The conductive response in the percolation dominated regions is therefore constant in $\omega$. Above the critical value, where there are likely to be capacitive but no conductive paths, the response is proportional to $\omega$. It can thus be seen that the response is governed by percolation at low (or high) values of the angular frequency ($\omega$) of the applied AC field, where the dielectric (capacitive) elements are effectively holes (or short-cuts) in the network. The exact values in these regions is highly variable and dependent on the exact arrangement of the components in the system, showing up as a wide spread on the histograms of 100 random realizations in figure 1. This is in contrast to the central *emergent* region, where paths are tightly bound and exhibit a power-law response. This central region is thus termed the *power-law emergent response* (PLER).

## 2. Experimental and theoretical approaches

A similar form of response to that shown in the previous section has been found in several other systems composed of a mixture of components with variable contrast ratio ($h$) between the components' individual permittivities ($\sigma_1$, $\sigma_2$). Besides dielectric materials and network simulations thereof, it is also found in the thermal conductivity of binary composites and in simulations of mechanical stresses in a binary disordered network of springs [11]. In all of these cases, the power-law response over several orders of magnitude in $|h|$ was found to be independent of the details of the system, such as the exact microstructural arrangement within a disordered material. The power-law region was found in all cases to have a scaling exponent $\alpha$ close to the volume fraction of the variable component in the composite over a wide range of volume fractions and decades in contrast ratio [12–14].

The combined permittivity ($\sigma$) of a material composed of a mixture of two components, as a function of their contrast ratio, can be studied in various ways. In the current case, the electrical conductivity of dielectric materials is dependent on the angular frequency ($\omega$) of the driving AC electric field. Therefore, the bulk admittance, $Y$, of composites of a dielectric (with the admittance $Y_1 \propto \omega$) and conductor (for which $Y_2$ is independent of $\omega$) can be studied over a range of $|h|$ by varying $\omega$. The form of the response in the power-law region follows a logarithmic mixing rule [17]:

$$\begin{aligned} \ln Y &= \alpha_1 \ln Y_1 + \alpha_2 \ln Y_2, \\ Y &= Y_1^{\alpha_1} Y_2^{\alpha_2}, \end{aligned} \tag{1}$$

where $\alpha_1$ and $\alpha_2$ are the volume fractions of the respective components. Other composite materials with a variable contrast ratio between components can be studied in similar ways. For example, the compressibility of some materials can be temperature dependent, so the overall compressibility of a composite in which one of the components has this property can be studied over a range of $h$ by varying the temperature.

It is shown in [15] that these experimental results are a feature of microstructural electrical networks, by comparison with network simulations, as outlined in the next section. It has been noted that these power-law dispersions are emergent phenomena characteristic of any system that is effectively a complex binary random network [11, 14]. As mentioned in the previous section, the power-law region (PLER) is found in all studies on finite systems to be bounded at high and low $h$ by limits determined by percolation paths within the material. Outside the PLER $Y$ varies from one random realization to another and is proportional to either $Y_1$ or $Y_2$, depending on whether a percolation path exists for the respective component.

### 2.1. Modelling microstructures

Many different approaches exist to model the conductivity of complex composites. The simplest of these is the largely empirical Lichtenecker law (cf equation (1)). There are also analytical approaches such as the effective medium and Bergman–Milton theories [2, 18–20]. Since the advent of modern computing and the ability to solve systems algorithmically using numerical methods, new approaches to studying complex materials have been developed. In the case of conductor–dielectric mixtures, large electrical circuit representations can be used, replacing the constituent conducting and dielectric parts with a network of resistors and capacitors, respectively[1]. For a binary disordered mixture, the different components can be assigned randomly to bonds on a lattice [21]. In most previous studies a 2D square lattice has been used, with bonds assigned randomly as either C or R, with probability $p$, $1-p$, respectively (for examples see later figures). This approach is closely related to percolation models and a large review of this and binary disordered networks can be found in [20].

There are many advantages of using *RC* network representations of these types of system. Particularly, widely available circuit simulation software can be used, which makes use of the available efficient sparse-matrix techniques in solving the equations of the system. Additionally, for bounded 2D square-lattice networks, equivalence transformations can be used to efficiently solve the system by taking advantage of the structure using the circuit analysis methods originally devised by Frank and Lobb [14, 22]. These techniques were used in various studies to show that the PLER exists in any binary random network with variable contrast ratio [11–13, 16, 21]. This indicates that the universal dielectric response found experimentally is a feature deriving from the random nature of the microstructure rather than from any 'new physics' at the microscopic level. These circuit representations also imply that the logarithmic mixing law of equation (1) can be derived from the standard equivalence rules:

$$Y^\nu = \sum_{k=1}^{N} \alpha_k Y_k^\nu \tag{2}$$

with $\nu = 1, -1$ for parallel or series components, respectively; where $Y$ is the admittance and $\alpha_k$ is the proportion of the $k$th component. Following the derivation in [21], we take $\nu \to 0$ and use the approximation $Y^\nu \to 1 + \nu \log Y$ to transform equation (2) into equation (1).

These methods have been useful in explaining the experimentally observed behaviour; however, the validity of results from numerical models can depend on the accuracy of the assumptions upon which they rest. It has largely been presumed that randomly assigning components to bonds on a lattice using a Bernoulli ($p$, $1-p$) distribution results in behaviour equivalent to that produced by microstructural disorder in the real physical systems. Thus, questions remain regarding the extent of the validity of this approach, as the behaviour of the system may depend on the type and scale of the disorder at the microstructural level, and the nature of the mixing. If structure is important, a further question arises as to the degree of refinement needed to sufficiently simulate the smallest scale structures. This is also important because in cases where small-scale structures are represented

[1] Resistor–resistor networks can also be used, by holding $\sigma_{R1}$ constant and varying $\sigma_{R2}$.

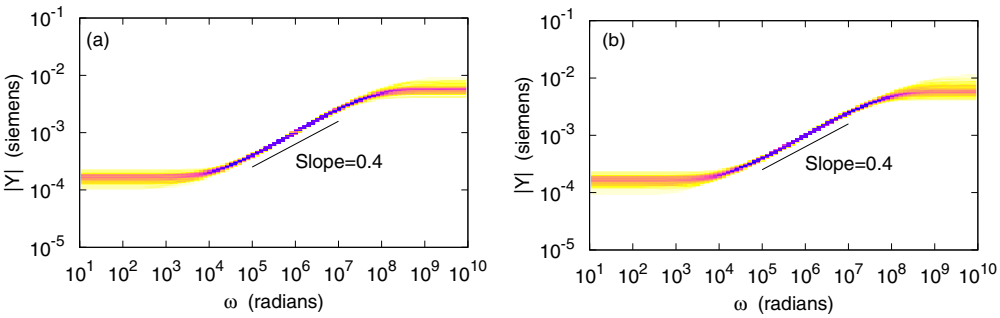


**Figure 2.** Histograms showing the conductive responses of 100 random realizations of 50 × 50 node random networks with 60% $R$ (1 kΩ) and 40% $C$ with ($a$) fixed-values of 1 nF and ($b$) normally distributed $C$ values (1 nF ± 0.333). The PLER can be seen, bounded by percolation and saturation limits at low and high frequencies.

it is necessary to use larger networks to simulate to a higher resolution, which consumes more computing time.

Therefore, here we study the effect on the PLER of both changing the distribution of the respective inclusions and of simulating more 'realistic' microstructures on different scales. The methods used are essentially the same as in [12, 13]; using freely available SPICE based circuit simulation software, which takes advantage of sparse-matrix techniques to solve the linear equations obtained using Kirchhoff's relations. It is shown that the main qualitative features of the results are largely insensitive to such considerations. So long as the network is of sufficient size to ensure a uniform mixture ($\gtrsim$20 × 20 nodes), the response of the system is robust to the exact details of the microstructural disorder. These results are consistent with studies on many complex systems, which can display the striking feature of *universality*, whereby the results are largely independent of the exact details of the system, so long as the correct essential features are represented by the model. An exception to this robust behaviour is found when significant anisotropy is introduced to microstructures in the system, significantly affecting the percolation probabilities. It will be seen that, in these cases, where the geometry of the microstructures begins to resemble series bands of components either parallel or perpendicular to the applied field when the anisotropy is in the respective directions, the power-law behaviour is lost, but can be found strikingly as the geometric mean of the two cases; giving validation to the use of equation (2).

## 3. Results

### *3.1. Modelling different distributions*

We start by investigating the effect of changing the binary distribution of components assigned to the network. This is achieved by replacing the single values of the capacitive elements with a distribution of values, as in real materials individual responses of the dielectric inclusions would not all be identical. Values were assigned randomly from various probability distributions, including uniform and Gaussian. A comparison is shown in figure 2, where the magnitude of the complex impedance of the network $|Y|$ is plotted against the angular frequency $\omega$, on a log scale. Both plots are histograms over 100 random realizations of circuits with 40% capacitors and 60% resistors on a 50×50 node square lattice. The network response is shown between cases where ($a$) all capacitors have the same value of 1 nF and ($b$) values are normally distributed about 1 nF with a standard deviation of 0.333 nF. In all cases the resistors have a fixed value of 1 kΩ and the frequency is varied from $f = 1$ Hz to $f = 10$ GHz, where $\omega = 2\pi f$. Note that the critical angular frequency is $10^6$ rad s$^{-1}$, at which the capacitors individually have the same magnitude of impedance as the resistors. The percolation and saturation limits at high and low $\omega$, where the admittance is constant, can be seen clearly. The PLER consists of tightly bound paths in the region $10^4 \lesssim \omega \lesssim 10^8$, where the results are largely independent of the microstructural details of the realization and have slope $\alpha = 0.4$, equal to the proportion of capacitors $p$ in the networks.

It can be seen from these results that the behaviour is robust to the extra disorder included in the capacitive elements. In particular, the PLER appears not to be significantly disturbed, exhibiting the same features such as power response and range. This robustness was found generally, no matter which type of distribution was used for the spread of values of the elements in the network.

### *3.2. Varying correlation lengths*

The next part of this investigation looks at the effect of modelling realistic microstructures in the simulations in order to test the reliability of models used in previous studies. In such models there was no attempt to represent any physically realistic features at the mesoscopic level, relying instead on the inherent randomness of the allocation of resistors and capacitors to form structures within the simulated material.

The simulated microstructures here were generated from Gaussian random fields [23]; in which the variables in space are governed by probability density functions. In this way sites in the network have a higher probability of being similar to their neighbours, up to some correlation length; imposing a spatial structure on the network. In the following cases a spherical correlation function was used with a defined correlation length, as given in [23].

The fields generated in this way can then be thresholded at the required occupation probability to give a binary image with different proportion of components and have a specified correlation length, defining an average imposed size on the

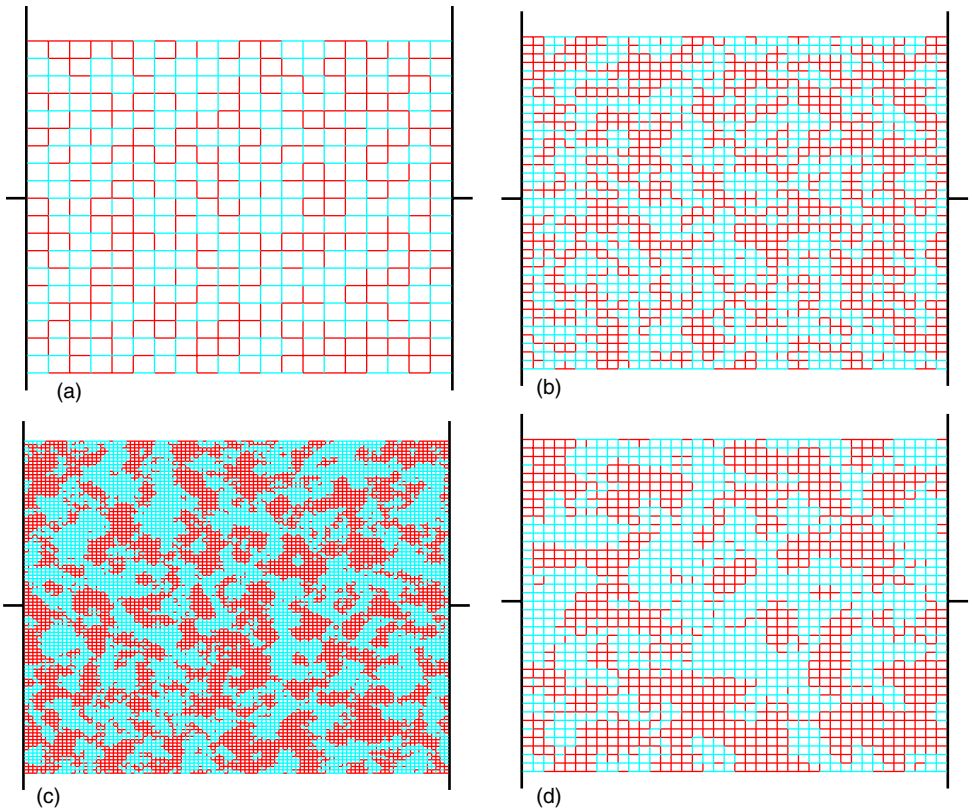


**Figure 3.** Examples of networks generated to resemble random microstructures, each with 50 : 50 mixture of components. In the first three subfigures the correlation length is set to 5% of network size $S$ (components along an edge): ($a$) $S = 20$, ($b$) $S = 40$, ($c$) $S = 100$. In ($d$) the correlation length is 10% of the network size $S = 40$. Note that the total number of components $N = S^2 + (S - 1)^2$.

structures. The networks thus obtained have microstructural features qualitatively similar to those seen in real materials. Examples of networks obtained using this technique are shown in figure 3, demonstrating different correlation lengths and network sizes, which correspond to different resolution simulations. A 50 : 50 mixture of components is used for the examples shown in figures 3 and 4. Parts ($a$)–($c$) show examples where the correlation length is set to 5% of the length along one edge of the network, defined as the network size $S$, which is assigned the values $S = 20$, 40 and 100, respectively, in the three subfigures. The total number of components in the networks is $N = S^2 + (S - 1)^2$, including both horizontal and vertical components. Part ($d$) of figures 3 and 4 has a correlation length of 10% of $S$, where $S = 40$ for comparison with ($b$).

In this way we can compare simulations with different resolutions with respect to a fixed correlation length by looking at ($a$)–($c$), as well as see the effect of varying the correlation length by itself by comparing ($b$) and ($d$). It can be seen immediately that the qualitative features of the plots remain unchanged. The central PLER region has slope $\alpha = 0.5$ on a log scale and extends over several orders of magnitude in $\omega$. Beyond the limits of the PLER the response is dominated by percolation effects and is either constant or directly proportional to the frequency depending on whether there is a resistive or capacitive percolation path. This is true despite there being an imposed correlation length for clusters due to the fact that, as the 50% threshold is approached, clusters in the embedded phase become more dense and join up to make larger clusters, resulting in a transition at 50% where it becomes the matrix phase.

Quantitatively there are some small differences in the response. Particularly the exact extent of the PLER is affected, giving slightly different mean values for the upper and lower limits. This can be seen upon considering the increasing resolution, represented by increasing network size in figures 4($a$)–($c$), whereby the extent of the PLER increases with increasing $N$. This could have consequences when simulating real materials, where the grains tend to aggregate into structures similar to those used in these simulations. However, the similarity between subfigures ($a$) and ($d$) is also striking. In the first case a coarse network with small correlation length is used, whereas both the correlation length and the network size have been doubled in the second. This indicates that low-resolution simulations, which do not include arbitrarily fine-scale detail may be useful, so long as the random spread of structure sizes is comparable to those in the real physical samples, with the smallest structures in the model representing the smallest grains in the sample.

### *3.3. The effect of anisotropy*

In real materials the microstructures may not be completely isotropic, so the effect of anisotropy on the results is studied in this section. Figure 5 shows two example $RC$ networks with different correlation lengths along the directions perpendicular

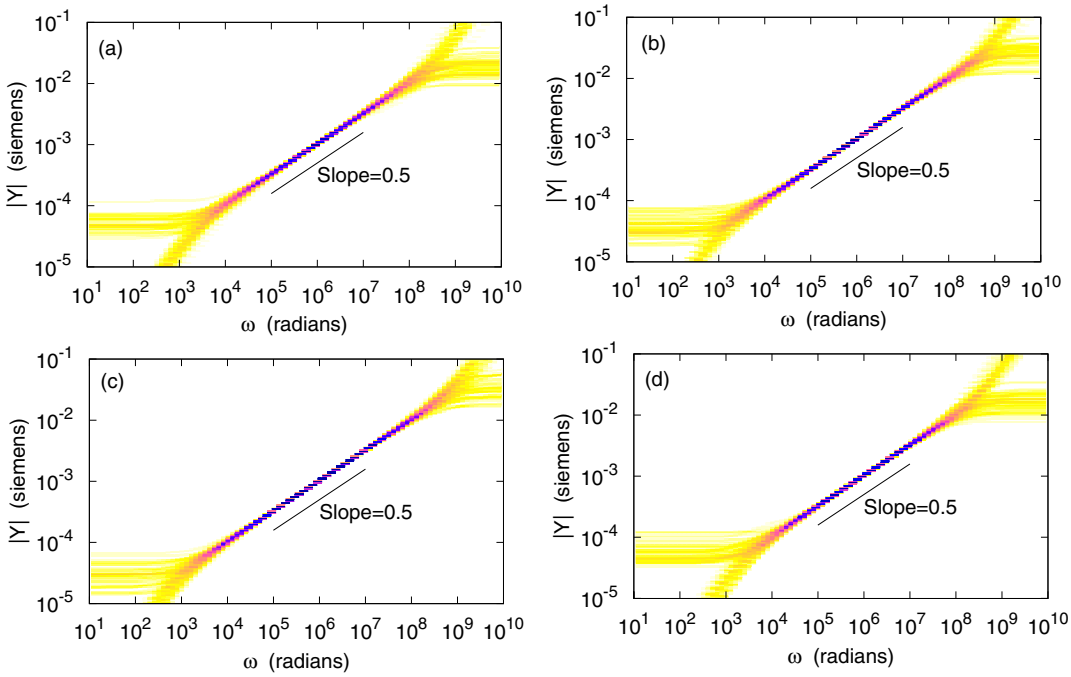


**Figure 4.** Histograms of admittances corresponding to the example networks shown in figure 3, using a 50 : 50 mixture of components. In (*a*)–(*c*) the correlation length (corresponding to structure sizes) is 5% of the network size $S = 20$, $S = 40$ and $S = 100$, respectively, and in (*d*) the correlation length is 10% of $S = 40$.

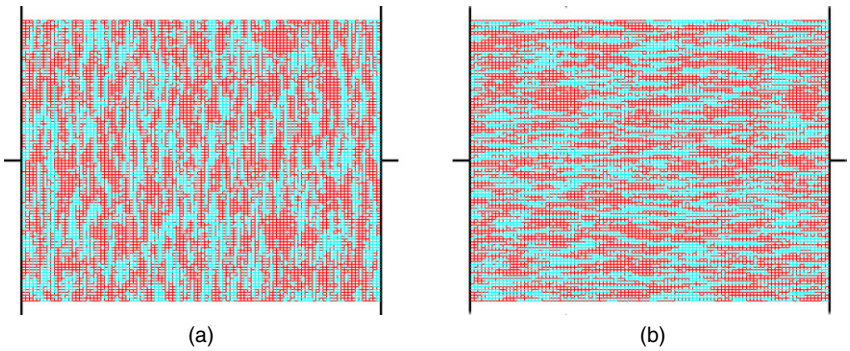


**Figure 5.** Size 100 × 100 node networks representing anisotropic structures, with the direction of longer correlation length running (*a*) perpendicular and (*b*) parallel to the direction of the applied field. A 10 : 1 ratio of correlation lengths was used for these example figures. It can be seen that the systems tend towards circuits with components arranged purely in parallel/series.

and parallel to the applied field. As the ratio of stretching becomes very large, the two cases of where the longest correlation axis is (*a*) perpendicular and (*b*) parallel to the applied field approximate a system where bands of parallel resistors and capacitors are either in series or parallel with this field.

The resulting admittances and phases in cases where the proportion of capacitors $p = 0.4$ are shown in figures 6(*a*) and (*b*) for perpendicular and parallel banding, respectively, where the ratio of long and short axes is 10 : 1. The networks were again derived from a Gaussian random field, stretched in the required direction and applied to 100 × 100 node networks ($N = 19801$ components). The value of $p = 0.4$ was chosen for (*a*)–(*c*) to show how the essential features of the now familiar response curve are distorted by the introduction of anisotropy into the system. It can be clearly seen that in these cases the power law is lost, with the curves tending towards those expected for the ideal cases of perpendicular and parallel bands of resistors and capacitors. However, the central region is still tightly bound and independent of the exact microstructural details of the realizations. In addition, a great deal of symmetry can be seen between the two cases when looking at both the admittances and the phase angles ($\phi$) between the complex voltages at the terminals.

A remarkable result is obtained when taking the geometric mean of the admittances of parallel and perpendicular realizations ($\sqrt{|Y|_{\parallel}|Y|_{\perp}}$). When this is performed, as shown in subfigure (*c*), the PLER is recovered and can be seen to exist over a considerably wide range in $\omega$. Very similar results are obtained for other values of $p$, the most striking being when $p = 0.5$; the critical percolation threshold for a 2D binary network. Here the combined admittance forms an almost

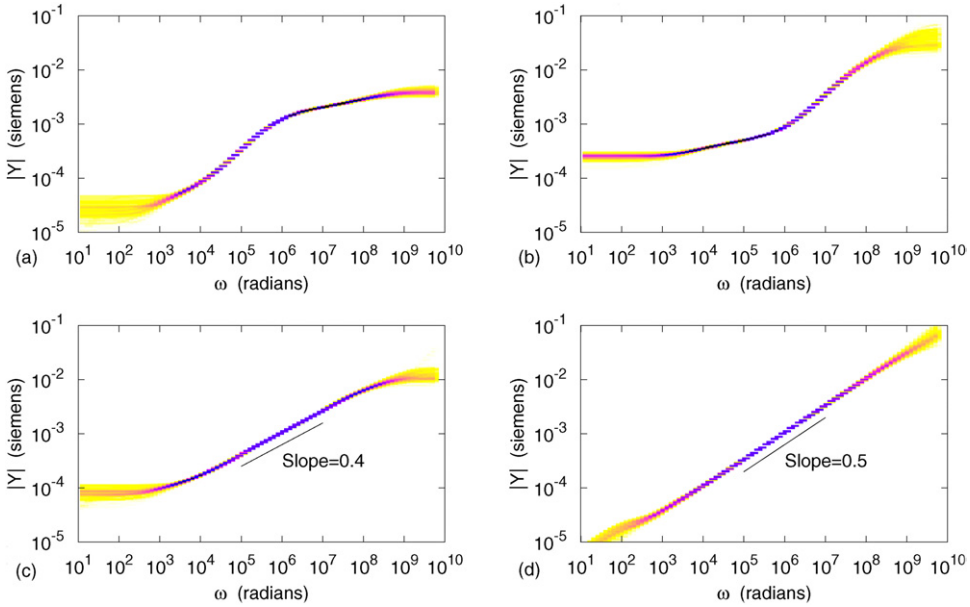


**Figure 6.** Responses of 100 × 100 node networks with microstructures scaled anisotropically perpendicular and parallel to the applied field are shown in (*a*) and (*b*), respectively. In each case the long axis has a correlation length 10 times that of the short, with a mix of 40% capacitors and 60% resistors. The geometric mean of the admittances of the two orientations of each realization was taken to produce the histograms in (*c*), showing how the PLER can be obtained in this way. Figure (*d*) shows the same, but for a 50 : 50 mix of components.

perfect power law (figure 6(*d*)), stretching from near zero to very high values of $\omega$, as would be expected for a random network at $p = 0.5$ where $N \to \infty$.

It can be seen (figures 5(*a*) and (*b*)) that in the limit where the ratio of long to short axes is very large, the systems asymptote towards the Wiener limits, which are equivalent circuits of components purely in parallel/series and are used to give the lower/upper bulk conductivity bounds. Therefore, the results presented in figure 6 provide strong evidence that the effective conductivity of a composite network is given by the geometric mean of the parallel and series equivalent circuits, allowing for the existence or absence of a dc conducting path through the medium; giving validation to equation (2).

## 4. Discussion

In this paper we have addressed many of the outstanding concerns regarding the assumptions made when using network models to simulate the universal dielectric response found in composite materials. The robustness of the PLER of random electrical networks with bimodal distributions of components has been studied in detail. This was carried out in order to evaluate the usefulness of such models in simulating real composite materials with microstructural disorder. In real materials we may not know the details of the microstructures, so it is important to know the relevance of such knowledge and how it impacts on the reliability of our numerical models. To deal with this, the effect of imposed microstructures was investigated, looking at both the size of structures and the network size. It was shown that the exact microstructural details may be reasonably omitted, so long as we take care that the general features of the structures, such as their relative smallest and largest scales, are represented. Therefore, effort and computing time may be saved by taking account of these findings. Such network representations also have applications beyond electrical conduction; having relevance to systems in mechanical systems amongst others [11], thus giving wider validity to these results outside of dielectrics.

It was also shown that the inclusion of anisotropy in the directions parallel and perpendicular to the applied field alters the bulk response of the system, destroying the power law while at the same time retaining the robustness to the exact details of each random realization. In these cases the network responses were found to tend towards what would be expected for purely parallel and series components (cf the Wiener limits). This reveals that it is possible to further tailor the response of complex materials by including such features at the microstructural level. Remarkably, the power law can be obtained by taking the square root of the product of the two cases, indicating that the bulk response of such systems is given by the geometric mean of the parallel and series equivalent circuits. This additionally validates using the logarithmic mixing rule to model the universal dielectric response in composite materials.

A final point to note is that in the majority of studies on network representations of this kind of system two-dimensional grids have been used. While these models have the advantage of making the computational implementation easier to handle, some may doubt the extent of their validity, given that real materials are usually three-dimensional and many critical theoretical quantities and results are sensitive to the dimension of the system. For example the critical probability for bond percolation is $p_c = 0.5$ only on two-dimensional square lattices; on a 3D cubic lattice $p_c \approx 0.249$ [24]. In work previously published by our group [16], results were shown for simulations of conduction using finite element methods to investigate a 20 × 20 × 20 element composite with

a 50% mix of each component. The qualitative features were as we would expect given the results of the 2D calculations and knowledge of the properties of 3D lattice percolation networks, with percolation in both components and the appropriate limits to the observed PLER. Given the much longer computing times needed to simulate in higher dimensions, it is therefore reasonable to use more amenable 2D representations when investigating complex composite materials, so long as we account for the discriminating features of each approach.

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