The Role of the Non Universal Static Dielectric Constant in the Scaling of AC Conductivity Results

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Abstract: The scaling of many experimental results for the AC conductivity plotted against frequency is a well known phenomenon. This paper shows that the scaling of experimental systems can only occur if the non universal static dielectric constant has a specific functional behavior, such as to allow scaling.

Introduction: For the last 50 years it has been observed that logarithmic plots of the real component of the AC conductivity $\sigma_{mh}(\omega)/\sigma_{mh}(\omega=0)$ against the scaled angular frequency ω/ω_C for high resistivity disordered materials, composites and ionic conductors, can be scaled to give superimposed plots, as those given in Fig. 1 [1 and the references therein]. The results are scaled onto each other, using a scaling factor $(1/\omega_c)$, which differs for different temperatures or compositions. The plot shown below is for a series of binary good conductor-bad conductor composites, with different volume fractions (φ) of the good conductor [1]. Several theories have been put forward to explain this ***universal*** conductivity phenomenon, most of which involve percolation concepts [see 1 for references].

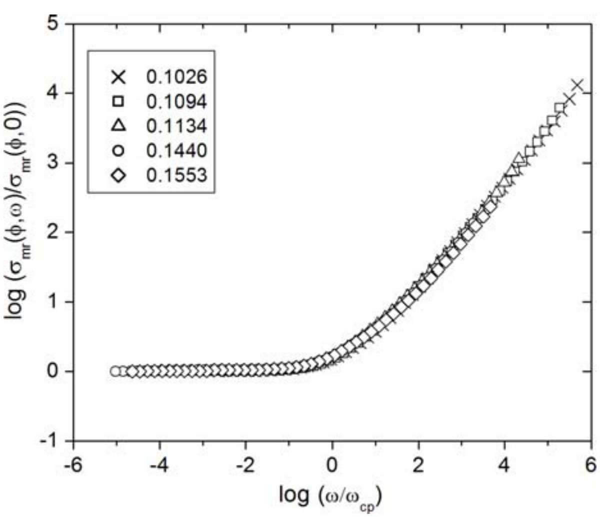

Fig. 1. Plots of experimental data of $\log(\sigma_{mr}(\varphi, \omega)/\sigma_{mr}(\varphi; 0))$ against $\log(\omega/\omega_{cp})$, for the Magnetite-Cellulose Acetate system at 25^0C. The values of φ are indicated and $\omega_{cp} \equiv \omega_{cp}$ was calculated from experimental data, as discussed in the text.

Theory and simulations: In this paper, $\sigma_m = \sigma_{mh}+i\sigma_{mi}$ is the composite conductivity and the conducting component conductivity is $\sigma_h = \sigma_{hr} + i\sigma_{hi}$. For the insulating component, $\sigma_l = \sigma_{lr}+ i\sigma_{li}$, where $\sigma_{li}= \varepsilon_0\varepsilon_{ir}$ and Φ_c is the percolation concentration. It has previously been shown [2 -7] that the Two Exponent Phenomenological Percolation equation (TEPPE), which is,

$$(1-\varphi)\,(\sigma_l^{1/s} - \sigma_m^{1/s})/(\sigma_l^{1/s} + A\sigma_m^{1/s}) + (\varphi)\,(\sigma_l^{1//t} - \sigma_m^{1/t})/(\sigma_l^{1/t} + A\sigma_m^{1/t}) = 0, \qquad (1)$$

with $A = (1 - \varphi_c)/\varphi_c$ and the exponents s and t, best describes experimental results for percolation systems, especially the second order terms [2-7]. When $s = t = 1$ and $\varphi_c = 1/3$, the equation is equivalent to the Bruggeman Symmetric Media Equation. [8]. The TEPPE also reduces to the first order terms of the three standard percolation equations [reviewed in 9, 10], in the appropriate limits. These limits show that s and t are the exponents characterizing the power laws, that are obtained well below and above φ_c respectively. The TEPPE gives the theoretical scaling frequency as,

$$\omega_{ct} = \sigma_{hr}(\varphi,0)[\varphi - \varphi_c)/(1-\varphi_c)]^t/ \omega\varepsilon_0\varepsilon_{lr}(\varphi,0)[(1-\varphi_c)/(\varphi-\varphi_c)]^s, \qquad (2)$$

which is in agreement with the result from the standard percolation equations, except for some necessary normalizing factors. From universality considerations, all experimental results should be able to be scaled by ω_{cr}, given in Eq. 2, using the s and t obtained from fitting the TEPPE to DC conductivity results. In practice it is found that no experimental results for percolation systems can be scaled using Eq. 2 [1], showing that no experimental results are strictly universal. Note that the derivation of the standard percolation equations [9, 10] relied heavily on universality in their derivation.

Experiments [4] where the first to show that the correct scaling frequency could be obtained from the experimental results, by finding the position of the peak of the imaginary Impedance -Z" (Z = Z'- iZ") as a function of ω. Figure 2 shows theoretical values, form the TEPPE, for $(-Z''(\varphi, \omega)/-Z''(\varphi,\omega)_{max})$ plotted against log ω, which are clearer than actual experimental results. The normalization by $-Z''(\varphi,\omega)_{max}$ ensures that all the curves have the same amplitude. The results from these *simulations* to obtain ω_{cp} agree with ω_{ct}, from Eq. 2.

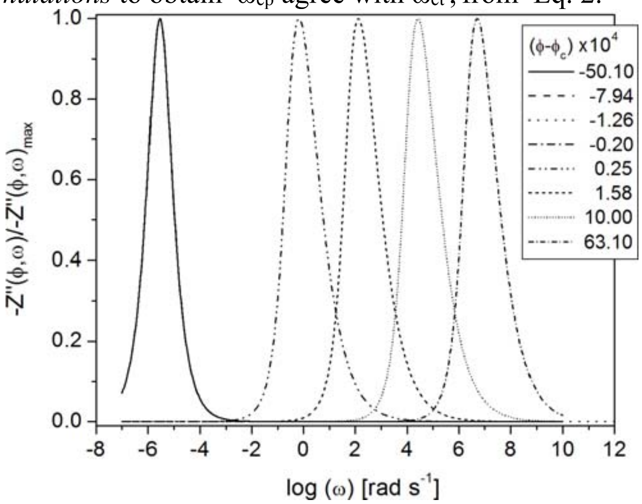

Fig. 2. Plots of the normalized imaginary Impedance ($-Z''(\varphi, \omega)/-Z''(\varphi, \omega))_{max}$ plotted against log ω for various values of $\varphi - \varphi_c$. Here s = 0.87, t = 2, $\varphi_c = 0.16$, $\sigma_{hr} = 10^2$ $(\Omega m)^{-1}$, $\sigma_{lr} = 10^{-16}$ $(\Omega m)^{-1}$, $\varepsilon_{hr}=10$ and $\varepsilon_{lr}=4$. Note the first arcs for $\varphi < \varphi_c$ lie almost on top of each other.

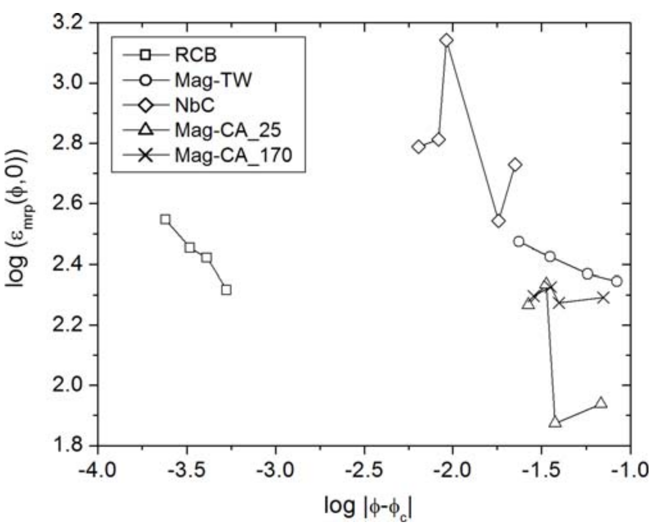

Fig. 3. Plots of $\log(\varepsilon_{mrp}(\varphi, 0))$ against $\varphi - \varphi_c$ for the Raw Carbon Black, Magnetite and NbC-Talc wax systems [1,3], as well as the Magnetite-Cellulose Acetate system [1,4] at both 25^0C and 170^0C. Note that all the samples are above φ_c. The solid lines are a guide to the eye.

***Experimental Results**:* The results from [3] and three sets of experiments from [5] were examined and experimental values of $(-Z''(\varphi, \omega)/-Z''(\varphi, \omega)_{max})$ against ω were plotted in order to obtain ω_{cp}. In all cases the scaling plots, as illustrated in Fig. 1, using these values for ω_{cp}, gave excellent results. As the DC conductivity, given by $\sigma_{hr}(\varphi,0)[\varphi - \varphi_c)/(1-\varphi_c)]^t$ in Eq. 2, can easily be measured, experimental values for $\varepsilon_{lr}(\varphi,0)$ can be calculated. The results of these calculations are plotted in Fig. 3. Note that as these are indirect calculations of a second order component,≈ a certain amount of irregularity in the results is to be expected.

Plots of log $(\varepsilon_{lrp}(\varphi,0))$ against log $(\varphi - \varphi_c)$ using the TEPPE (not shown) where made. These results, for low σ_{lr}/σ_{cr} ($<10^{-18}$), show a divergence, which is always predicted by the standard percolation equations [9, 10]. However, the results for $\sigma_{lr}/\sigma_{cr} \approx 10^{-10}$ show a behavior qualitatively similar to those in Fig. 3.

Conclusions: The universality of scaling results shown for experimental percolation systems are not universal in that the slopes of the $\log(\sigma_{mr}(\varphi, \omega)/ \sigma_{mr}(\varphi; 0))$ against $\log(\omega=\omega_{cp})$ curves are not in accord with theory, when the DC values of s and t are used in Eq. 2. Universal plots for such systems are only observed due the behavior of the dielectric constant $\varepsilon_{lrp}(\varphi,0)$, which was obtained for the first time in [1]. A theoretical explanation for $\varepsilon_{lrp}(\varphi,0)$ does not exist, but the behavior of $\varepsilon_{lrp}(\varphi,0)$ is in semi qualitative agreement with the phenomenological TEPPE. Further analysis of work on non percolation systems, that show universal $\log(\sigma_{mr}(\varphi, \omega)/ \sigma_{mr}(\varphi; 0))$ against $\log(\omega=\omega_{cp})$ curves, needs to be made in order to calculate $\varepsilon_{lrp}(\varphi,0)$ for such systems and compare with the results presented in this paper and [1].

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