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Statistical and scaling properties of the alternating-current conductivity in thin metal–dielectric composites

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Abstract. We study in this paper the scaling and statistical properties of the alternating-current conductivity of thin metal–dielectric composite films for different degrees of loss in metallic components and particularly in the limit of vanishing losses. We model the system as a two-dimensional RLC network and calculate the effective conductivity by using a real-space renormalization group method. It is found that the real conductivity fluctuates strongly for very small losses. The correlation length diverges for vanishing losses leading to the failure of the effective-medium theory to describe the dielectric properties of such systems in this limit. We found also that the distribution of the real conductivity becomes log–normal below a certain critical loss R_c which is size dependent for finite systems. There is further discussion on the statistical characterization of the optical modes.

1. Introduction

Dielectric properties of composite materials have constituted for a long time a subject of intensive research [1] due to the wide variation of these properties achieved by changing their composition. The effective-medium theory (EMT) provides the most useful way to describe these properties [1]. In particular, the complex effective conductivity of thin metal–dielectric composites was found from this theory to behave at the percolation threshold (i.e. for the minimum concentration of the metal corresponding to the appearance of a continuous path; in 2D square lattices this metallic concentration is 50%) as [1, 2]

$$\sigma_{eff} = \sqrt{\sigma_m \sigma_d} \tag{1}$$

where the indices m, d and *eff* stand respectively for the metal, dielectric and effective medium. The conductivity of the dielectric component is generally assumed to be purely imaginary (the absorption being neglected) while the real part R of the metallic resistivity corresponds to the loss in the metal. Thus, in the limit of loss-free films ($R = 0$) corresponding to a superconductor–dielectric film, the effective conductivity from equation (1) is purely real and corresponds to an anomalous dissipation of the electric field. An anomalous absorption of light has been observed experimentally [2] and was explained recently as a storage of the electromagnetic energy in small regions of the film [3] or as a localization of the electric field [4]. However, this experimental observation and theoretical interpretation does not mean that the film becomes dissipative. Obviously, a film composed of non-dissipative components

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should not be dissipative. On the other hand, the real conductivity has been found (by two different methods) to vanish for vanishing loss [4] which is opposite to the prediction of equation (1). Therefore, it is interesting to check the validity of this equation in this limit where its predictions remain ambiguously interpreted. Indeed, EMT is valid only if the length scale is much larger than the correlation length [1, 2] and the result obtained recently [4] is probably for sizes smaller than this length scale. The scaling behaviour of the conductivity allows us then to investigate this quantity which is a characteristic length for such systems. This correlation length is defined as the length scale over which the field fluctuations become negligible. It can be measured by the scaling method used recently by Brouers *et al* [5] (at this length, the conductivity saturates). We note here that the scaling study of the conductivity by Brouers *et al* [5] has been restricted to just two values of the loss parameter: $R = 0.1$ and $R = 10^{-4}$, where they found that the real conductivity saturates at about one. However, the above-mentioned discrepancy with the effective-medium theory (1) was observed for much smaller losses [4] and should be enhanced for vanishing losses ($R = 0$). It is then necessary to measure the correlation length (L_c) for vanishing losses in order to check the behaviour of the effective conductivity which remains controversial in this limit.

In addition to the above-discussed discrepancy, a non-monotonic behaviour of the averaged real conductivity was observed previously [4] for $R \ll 10^{-5}$ indicating strong fluctuations of this quantity, while for larger losses it seems to be well averaged. Therefore, a critical loss R_c should exist separating the two different statistical behaviours of the effective conductivity for finite sizes (for sample sizes 512×512 this critical loss seems to be around 10^{-5}). An extensive study of this transition as well as the statistical properties of the effective conductivity is then needed both to correctly average it (since, if its distribution is actually log–normal, its moments can diverge) and also to characterize the phases on either side of R_c as for the electronic systems. Indeed the electric field in the Helmholtz equation plays an identical role to the electronic wavefunction in the Schrödinger equation [6] and a delocalization transition of the optical modes was found at this critical loss for such systems [4]. An analogy is then possible with the electronic systems where localized states (the insulating regime) are characterized by log–normal conductance distributions while for extended or weakly localized states (the metallic regime) these distributions are normal [7]. To the best of our knowledge, such an investigation of the statistical properties of the effective conductivity in such composite films has not been undertaken before.

The purpose of this paper is to examine in a first step the scaling properties of the real effective conductivity in two-dimensional (2D) metal–dielectric composites for different losses and to determine the correlation length L_c in the region of vanishing losses ($R \to 0$). We investigate in a second step the statistical properties of this conductivity on either side of the critical loss R_c. In order to check the validity of (1) in the limit of vanishing losses, we restrict ourselves in this work to the concentration of the metallic component corresponding to the percolation threshold p_c (in this case $p_c = 0.5$) where this equation is valid.

When the wavelength is sufficiently large compared to the grain size, we can neglect the skin effect, and this composite system is modelled by a 2D RL–C square network where the inductance L stands for the metallic grains with a loss (resistance) R, while the dielectric grains correspond to the capacitance C which is assumed without dissipation. Indeed, the conductivity of the metallic grains is well described by the Drude dielectric function and can be seen as an inductance with a resistance in parallel with a capacitance [8]. In the limit of small relaxation rate compared to the field frequency, itself being smaller than the plasmon frequency, the metallic component can be approximated as an RL-bond. The electrostatic potential in this random composite system satisfies

$$\nabla \cdot (\varepsilon \, \nabla \Phi) = 0 \tag{2}$$

which can be evaluated numerically as a set of coupled difference equations for the lattice by using a discrete approximation for the gradient operator. These resulting difference equations are identical to Kirchhoff's laws for a random impedance network. Such a random network resembles then the composite that it is intended to model. Therefore, such a network is expected to reproduce the effective conductivity, and its statistical properties, that we intend to study in this work. We restrict ourselves overall to the characteristic frequency ω_{res} where the ac conductivity of the dielectric component has the same magnitude as the metallic one for small losses. We can then use without loss of generality the framework where $L = C = \omega_{res} = 1$ (this frequency can be easily determined from the real values of the dielectric constants of the two components; in fact, for a gold–glass composite, $\omega_{res} \simeq 0.8 \times 10^{16}$ Hz and corresponds to the far-infrared region [9]). For frequencies different from the characteristic one we normalize them with ω_{res} (ω/ω_{res}). The metal and dielectric conductivities will then read at the characteristic frequency ω_{res} in this framework

$$\sigma_m = (\mathrm{i} + R)^{-1} \tag{3}$$

and

$$\sigma_d = -\mathrm{i}. \tag{4}$$

We use for the calculation of the effective conductivity the real-space renormalization group method (RSRG) extensively studied during the two last decades [4, 5, 10, 11] which consists in a representation of the network in Wheatstone bridges transformed into two equivalent conductivities following the directions x and y (see figure 1). This method has been shown to be a good approximation for the calculation of the conductivity and the critical exponents near the percolation threshold [5, 10, 11]. Metallic and dielectric components are generated (with an equal concentration 0.5) from a uniform distribution. The effective conductivity is averaged from 100 samples which leads to sufficient accuracy except in the case of very small losses where the effective conductivity is not self-averaged and its moments diverge.

2. Scaling properties and characteristic lengths

In addition to the coherence length and the correlation length [5], the localization length (L_{loc}) seems to be the third characteristic length for such systems. Indeed, electromagnetic modes were found to be localized in 2D fractal films [12], rough surfaces [13], non-linear Raman scattering [14] and also in the present metal–dielectric films [4] due to strong field fluctuations. However, this length seems to be equivalent to L_c, because the localization length for optical waves is defined as the mean size of the sample above which the local field strength (the equivalent to the wavefunction in the Helmholtz equation) decays by a factor e. This means also that above this length the field fluctuations become small, which is the definition of the correlation length L_c. In the previous work [4], we found a delocalization of the eigenmodes for vanishing losses ($R \to 0$) indicating the divergence of the localization length. Therefore the correlation length should also diverge in this limit.

Before examining the correlation length, let us first reconsider the scaling properties of the real part of the effective conductivity, as was done by Brouers *et al* [5], but for a wider range of losses extended to vanishing ones ($R \to 0$). Obviously our results are identical to those of Brouers *et al* for the values of the loss studied by them, i.e. $R = 0.1$ and $R = 10^{-4}$. As they found, the real conductivity increases and saturates at the correlation length (see figure 2). Furthermore, for very small losses the effective conductivity is shown to fluctuate strongly as opposed to the case for larger losses, where it seems to behave '*well*' statistically. We note also that, as expected, the saturation takes place at larger sizes when the loss is small, and

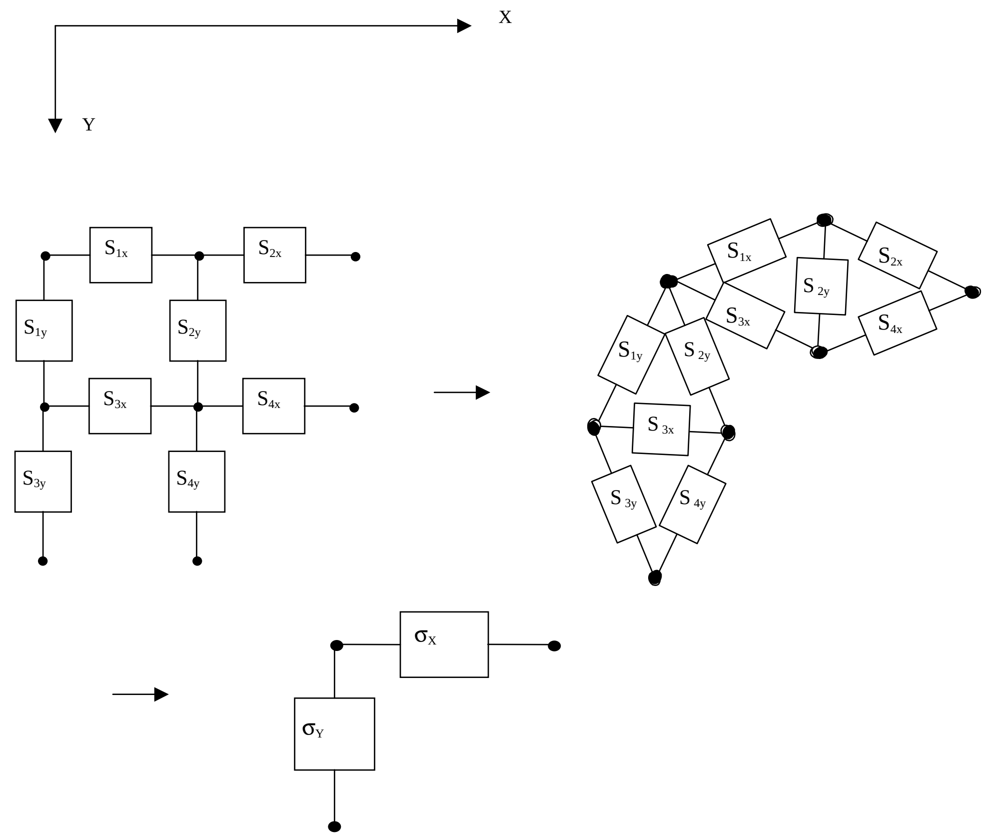

Figure 1. The real-space renormalization group for a square network.

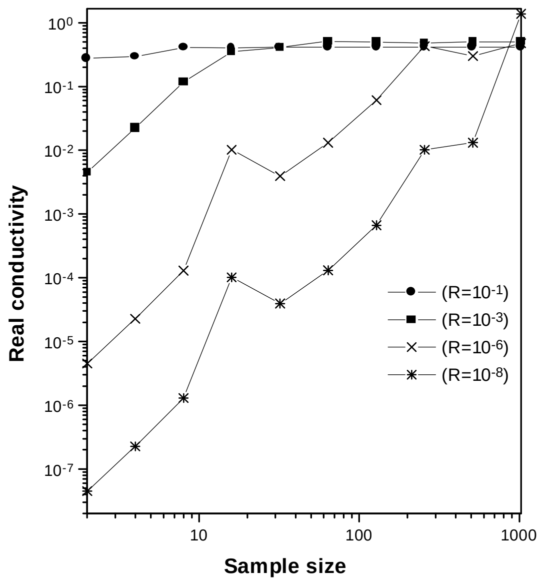

Figure 2. The real part of the conductivity versus the sample size for different losses R.

becomes much greater than 1024 for losses $R < 10^{-6}$. Therefore, the strong fluctuations of the effective conductivity appear when the size is smaller than the correlation length L_c (or equivalently, if the loss is smaller than the critical loss R_c for a fixed system size). The critical loss R_c is then strongly related to the correlation length L_c.

In figure 3 the correlation length is shown to increase rapidly when the loss decreases and shows power-law divergence for vanishing losses (see the inset of figure 3) with the power-law exponent 0.41 (close to the value 0.5 anticipated by Brouers *et al* [5]). This divergence is then in agreement with the delocalization effect found previously for such systems [4] as discussed above. Note that the divergence of the correlation length can also be predicted by EMT models like Bruggeman and Ping Sheng models [15]. Therefore, for loss-free films the correlation length is infinite and for any finite-size film equation (1) ceases to be valid. Indeed, our numerical calculations indicate exactly zero real conductivity for any finite size while equation (1) yields a real conductivity equal to one (from (2) and (3)). The anomalous absorption observed experimentally [2] is then not due to the dissipation but originates from a localization of the electromagnetic waves [4] or equivalently from the storage of the electromagnetic energy in the film [3].

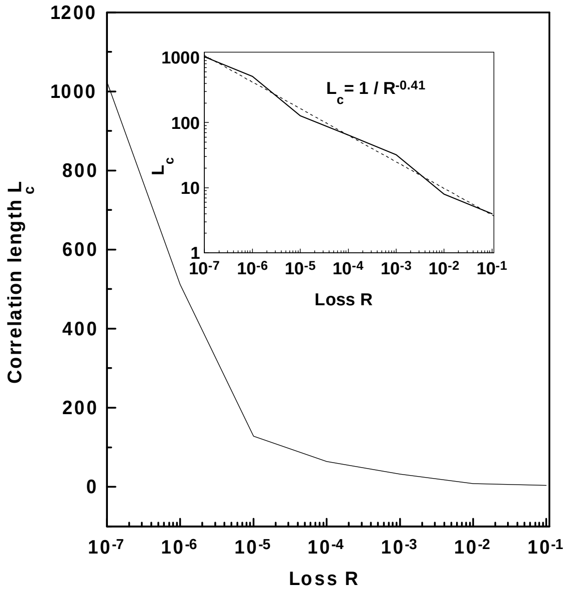

Figure 3. The correlation length as a function of the loss R. The inset is a log–log plot of this figure for the estimation of the power-law exponent.

3. Statistical behaviour of the conductivity

As shown in figure 2, when the loss is very small the effective conductivity (averaged over 100 samples) becomes strongly fluctuating. Therefore, as discussed in section 1, this conductivity may not obey the central-limit theorem [16]. It is then important to study its statistics before averaging it. This investigation can also be used to characterize the optical eigenmodes in such systems by analogy with the quantum counterpart. The conductance fluctuations for classical systems have been studied for a long time [17] but the electromagnetic eigenmodes have not been characterized.

In figures 4, we show the distribution of the conductivity for various losses. As shown in these figures, in the region of large losses ($R = 10^{-1}$ and 10^{-3}) the distribution of the real conductivity seems to be Gaussian and becomes narrower when the length scale increases, confirming that in this region the real conductivity obeys the central-limit theorem [16]. When the loss decreases these distributions become broadened, and consequently the conductivity fluctuations increase (as clearly shown in figure 5). For much smaller losses ($R = 10^{-6}$ and 10^{-9}) the distribution becomes Poissonian and narrows for larger sample sizes for $R = 10^{-6}$ (figure 4(c)) while it seems to be less affected by the size for $R = 10^{-9}$ (figure 4(d)). We note also from figure 4(c) and figure 4(d) that the distribution of the real conductivity narrows when the loss decreases and tends to a delta peak at zero conductivity for $R \to 0$, confirming our previous discussion of the dissipation for vanishing losses. In fact these Poissonian distributions of real conductivity for $R = 10^{-6}$ and 10^{-9} correspond to a log–normal distribution (not shown here to avoid a lengthy paper). Therefore for finite-size systems there is a phase transition at a

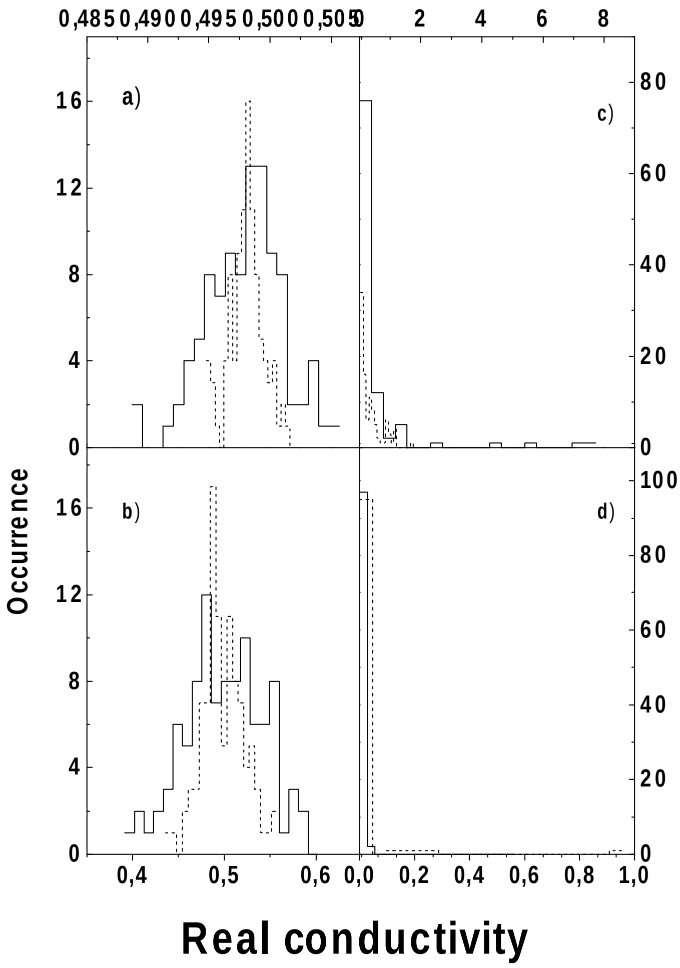

Figure 4. The distribution of the real part of the effective conductivity for sample sizes 512×512 (solid curve) and 1024×1024 (dashed curve) and for four values of the loss: (a) $R = 10^{-1}$, (b) $R = 10^{-3}$, (c) $R = 10^{-6}$ and (d) $R = 10^{-9}$.

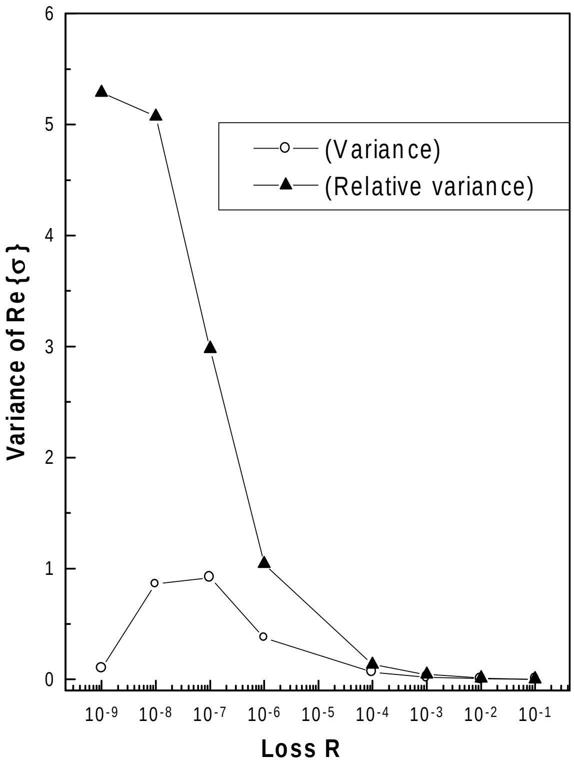

Figure 5. The variance of the real part of the effective conductivity (open circles) and the relative variance (filled triangles).

critical loss R_c from normal to log–normal distributions of real conductivity (R_c is estimated at about 10^{-5} for the sizes 512×512 and 1024×1024 used here). The critical loss is then size dependent as discussed above. For $R < R_c$ the averaged real conductivity should then be estimated from its logarithm, i.e.

$$\langle\sigma\rangle = \exp(\langle\log(\sigma)\rangle) \tag{5}$$

while for $R > R_c$, the conductivity is directly averaged. This method yields much smaller real conductivities for $R < R_c$ than the direct averaging and the decrease of the conductivity observed previously for vanishing losses [4] is enhanced. In particular, at $R = 10^{-9}$ the averaged real conductivity becomes at least one magnitude smaller than the directly averaged one. Therefore, the correlation length should increase more rapidly than in figure 3 for vanishing losses.

This statistical behaviour of the real conductivity characterizes also the two phases observed in reference [4] for a fixed sample size: localized modes for $R > R_c$ and delocalized modes for $R < R_c$. Therefore, from figures 4 the localized eigenmodes are characterized by a normal distribution of the real conductivity while for extended modes this distribution is log–normal. However, for the quantum counterpart the situation is reversed. In the electronic systems, we study the distribution of conductance which is related to the transmission coefficient [18]. The property analogous to the electronic conductance is then the optical transmission coefficient which should show a similar statistical behaviour to the electronic counterpart. Furthermore, a surprising analogy with the universal conductance fluctuations in the electronic systems [7] seems to be shown for the relative variance of the real ac conductivity, which becomes independent of the loss for $R \leqslant 10^{-8}$ (see figure 5) and of the length scale (see figure 4(d)).

4. Conclusions

We have studied in this paper the characteristic lengths and the statistical properties of the real part of the effective conductivity in 2D metal–dielectric composites at the percolation threshold and for a characteristic frequency ω_{res} where the conductivities of the two components have the same magnitude for a vanishing loss. We found that the correlation length (which is equivalent to the localization length) diverges when the loss R vanishes. Therefore the effective-medium theory represented by equation (1) ceases to be valid in this limit and the anomalous absorption observed in this case is not due to any dissipative behaviour of these films but to an energy storage of the electromagnetic waves in small regions of the sample.

We examined also the statistical properties of this effective conductivity and found two different distributions of this quantity on either side of a critical loss R_c. For losses $R > R_c$ the distribution of conductivity is Gaussian while for $R < R_c$ it becomes log–normal. Therefore in the latter case, the effective conductivity ceases to be self-averaged because all its moments diverge. The correct way to average this quantity (and avoid the non-monotonic behaviour of the conductivity in figure 2) is then to average its logarithm (as in equation (4)). For a loss-free system, the distribution becomes delta peaked at zero conductivity. The critical loss separating these two phases is size dependent and is estimated at $R_c = 10^{-5}$ for sizes around 1024×1024 while $R_c = 0$ for infinite size.

These distributions seem to characterize the nature of the optical eigenmodes in such films analogously with the distribution of conductance in the electronic systems. Indeed, it has been found that the eigenmodes are localized for $R > R_c$ while for $R < R_c$ they are delocalized [4]. However, these distributions seem to have long tails and the transition from normal to log–normal distribution is not clearly observed. Therefore, this transition should be investigated by using generalized distributions like Lévy distributions [19]. On the other hand, although the optical properties of loss-free films seem to be explained, some real effects like the local arrangement of the metallic components as well as the Flicker noise [15] are not taken into account in the theoretical calculations. On the other hand, although neglected in the modelling of such composites in RLC-networks, the skin effect can affect these distributions. These questions should be considered further.

Acknowledgments

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