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## Localization and absorption of the local field in two-dimensional composite metal–dielectric films at the percolation threshold

L Zekri<sup>†</sup>, R Bouamrane<sup>†</sup>, N Zekri<sup>†§</sup> and F Brouers<sup>‡</sup>

<sup>†</sup> USTO, Departement de Physique, LEPM, BP 1505 El M'Naouar, Oran, Algeria

<sup>‡</sup> Universite de Liege, Institut de Physique, Sart Tilman 4000, Liege, Belgium

E-mail: nzekri@meloo.com

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**Abstract.** We study in this paper the localization of the electric field and the dielectric properties of thin metal–dielectric composites at the percolation threshold. In particular, the effects of the loss in the metallic components are examined. To this end, such systems are modelled as random  $RL-C$  networks, and the local field distribution as well as the effective conductivity are determined by using an exact resolution of Kirchhoff equations in addition to a real-space renormalization group method for comparison. We find a delocalization of the eigenmodes which remain weakly localized for vanishing losses. This result seems to be in agreement with the anomalous absorption observed experimentally for such systems.

### 1. Introduction

Thin metal–dielectric films have been shown experimentally to exhibit an anomalously high absorption in the visible, near-infrared and microwave regimes at the percolation threshold [1–4]. This effect was interpreted as cluster plasmon absorption [4, 5]. Naturally, these mixtures should show an absorption since the dielectric constant of the metallic component is complex and the effective dielectric constant of the whole system should then also be complex, particularly at the classical percolation threshold (which is the point of transition of the effective dc conductivity from insulating to conducting due to the appearance of a continuous path of the conducting region through the sample), where from the effective-medium theory [6]

$$\epsilon_{eff} = \sqrt{\epsilon_m \epsilon_d}. \quad (1)$$

The indices  $m$ ,  $d$  and  $eff$  stand respectively for the metal, dielectric and effective medium. This equation was shown to be a good approximation if the system size is much larger than the correlation length. Therefore, equation (1) will not be valid if this quantity diverges. From this equation, both the real and imaginary parts of the dielectric constant of the metallic component contribute to the optical absorption of the system. However, a film with non-dissipative components naturally should also be non-dissipative and we expect in this case a vanishing absorption for the whole system, while equation (1) yields a non-vanishing absorption. Therefore, this description of the effective properties of such a medium may not be sufficient to explain the behaviour experimentally observed for such systems [1–4].

§ Fax: +213 6 421581.

The above-discussed experimental observation of the light absorption may then originate from physical phenomena other than the dissipation.

Indeed, since the light has a wave behaviour, the backscattering and the interference effects can strongly affect its propagation through a disordered system [7–9] and its localization is enhanced by disorder [8] as well as by absorption [10]. Therefore, the localization properties of the electromagnetic field in such systems, where absorption and disorder compete, can be a good tool for explaining the anomalous behaviour observed in such thin composite films, particularly at the classical percolation threshold. The classical percolation threshold in 2D square lattices of disordered bonds corresponds to a concentration of the metallic bonds  $p_c = 0.5$  [11, 12] (note that some composites carry current even below the classical percolation threshold due to the fact that tunnelling through disconnected (dispersed) metallic regions can give rise to some virtually connected percolating clusters [13]. This effect is not considered in this paper).

On the other hand, giant local field fluctuations have been recently found numerically in such films [14] at the percolation threshold and for frequencies close to a characteristic one  $\omega_{res}$  where the conductivities of the two components have the same magnitude ( $|\sigma_m| = |\sigma_d|$ ). Large local field fluctuations have been also found in fractal 2D films [15], 3D rough surfaces [16] and non-linear Raman scattering [17]. In both the 2D and 3D systems the electromagnetic modes were found to be localized, probably due to such fluctuations. Furthermore, Brouers *et al* [14] showed for the metal–dielectric films that the local field distribution is asymptotically log–normal. However, from the electromagnetic field theory investigated recently by Sarychev *et al* [4], we can easily deduce that the high strengths of the current (or equivalently the high local field intensities in this case) behave as the inverse of the local transmission of light. We conclude then that the local transmission also has a log–normal distribution for high field strengths in such films. Therefore, if we use the analogy between the electric field in the Helmholtz equation and the electronic wave-function in the Schrödinger equation [7], the local transmission of the electromagnetic waves is equivalent to the electronic conductance at zero temperature [18] where a log–normal distribution is a signature of localization [19, 20].

The aim of the present paper is to investigate the localization and absorption properties of such films modelled by a square  $RL$ – $C$  network, and use this approach to explain the anomalous absorption at the percolation threshold. The local field and the effective conductivity are calculated by using two different methods in order to compare with the results of [14]: an exact resolution of Kirchhoff equations for such networks which we call the exact method (EM) from now on, and a real-space renormalization group (RSRG) self-similar scheme [21]. The RSRG method has been shown to be sufficient for the computation of the effective conductivity [21], but the electric field has not been checked before although there is increasing use of this method [14, 22]. The degree of localization is measured by means of the inverse participation ratio (IPR) [23] applied to the electric field, while the absorption corresponds to the real part of the complex effective conductivity. We compare in a first step the field distribution obtained by the two methods for different losses and frequencies, and then examine the effect of the loss (resistance) in the metallic component on the localization as well as the absorption at the percolation threshold.

## 2. Method of calculation

From Maxwell equations, one can easily relate the complex conductivity of metallic and dielectric grains to their dielectric constant by

$$\sigma_{m,d} = -i\omega\varepsilon_{m,d}\varepsilon_0 \quad (2)$$

where  $\varepsilon_0$  is the vacuum dielectric permittivity and  $\omega$  the field frequency. Here the film thickness and the size of the components must be smaller than the light wavelength in order to allow neglect of the magnetic field variation. The dielectric constant of the insulator (assumed to be non-dissipative) is real while that of the metal is complex and, from equation (2), its absorption (imaginary part) is related to the real part of the conductivity.

When the field frequency is large compared to the relaxation frequency, composite metal–dielectric films can be modelled by 2D resistor networks [4, 5, 24]. The effective properties of these networks have been extensively studied during the last two decades [5, 24–26]. In the  $RL$ – $C$  picture, if the frequency is smaller than the plasmon frequency  $\omega_p$ , the capacitors  $C$  are the dielectric grains with a conductivity  $\sigma_C = -iC\omega$  and a concentration  $1 - p$ , while the inductances  $L$  are the metallic grains with a conductivity  $\sigma_L = (-iL\omega + R)^{-1}$  ( $R$  being the loss), and a concentration  $p$  is deposited or evaporated over the substrate. Therefore we can take in this case, without loss of generality,  $L = C = \omega_{res} = 1$  (the characteristic frequency  $\omega_{res}$  corresponds to the case where  $|\sigma_m| \simeq |\sigma_d|$  for vanishing losses, i.e.,  $C\omega_{res} = 1/L\omega_{res}$ ).  $L$  and  $C$  being constants near  $\omega_{res}$ , this assumption can be generalized to any frequency  $\omega$  (smaller than the plasmon frequency) normalized to  $\omega_{res}$ . The frequency  $\omega$  for a given film can easily be 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 this corresponds to the far-infrared frequencies [27]). The metallic conductivity then becomes for small losses

$$\sigma_L = \frac{1}{-i\omega + R} \approx \left( i + \frac{R}{\omega} \right) / \omega \quad (3)$$

while the dielectric conductivity is

$$\sigma_C = -i\omega. \quad (4)$$

The first method (the EM), used for the calculation of the local field distribution and the effective conductivity, consists in solving exactly Kirchhoff equations for the corresponding 2D square resistor network. This implies the use of matrices of size  $M^2 \times M^2$  (where  $M$  is the size of the network), which are impossible to handle numerically for large samples (for memory and computational time reasons). However, we take advantage of the sparse configuration of such matrices and their organization in blocks in their tri-diagonal region [27]. Indeed, since the Kirchhoff equation for a node  $i$  uses only the voltages of its nearest-neighbour nodes, the matrix involved here has in its diagonal region only blocks of sizes equal to the size of the film— $M \times M$  in this case. The diagonalization and inversion of such matrices is then achieved merely by the diagonalization and inversion of the constituent blocks. This reduces considerably the memory and computational time requirements. We note that this method provides exactly the same results on the effective conductivity as the Frank and Lobb method [28] but also calculates the local field distribution through the lattice which cannot be done by the other method. However, the time consumed remains large with this method (in particular when averaging over a large number of configurations), and the maximum size that we reach by this method with our computational systems is  $512 \times 512$  (which is sufficient for our statistical treatment).

This is one of the reasons for also using the RSRG method which requires much less computational time. This method, extensively described in previous works (see [14, 21, 29]), consists in a transformation of the 2D square lattice into Wheatstone bridges in the  $x$ - and  $y$ -directions (see figure 1). Each bridge is transformed into an equivalent admittance and the size of the system is divided by 2 in each step up to a final step where the lattice is reduced to two equivalent admittances following these directions. It is then easy to calculate by means of this transformation the effective conductivity, while the local field distribution can be obtained by the inverse procedure starting from the effective admittances already calculated. Although

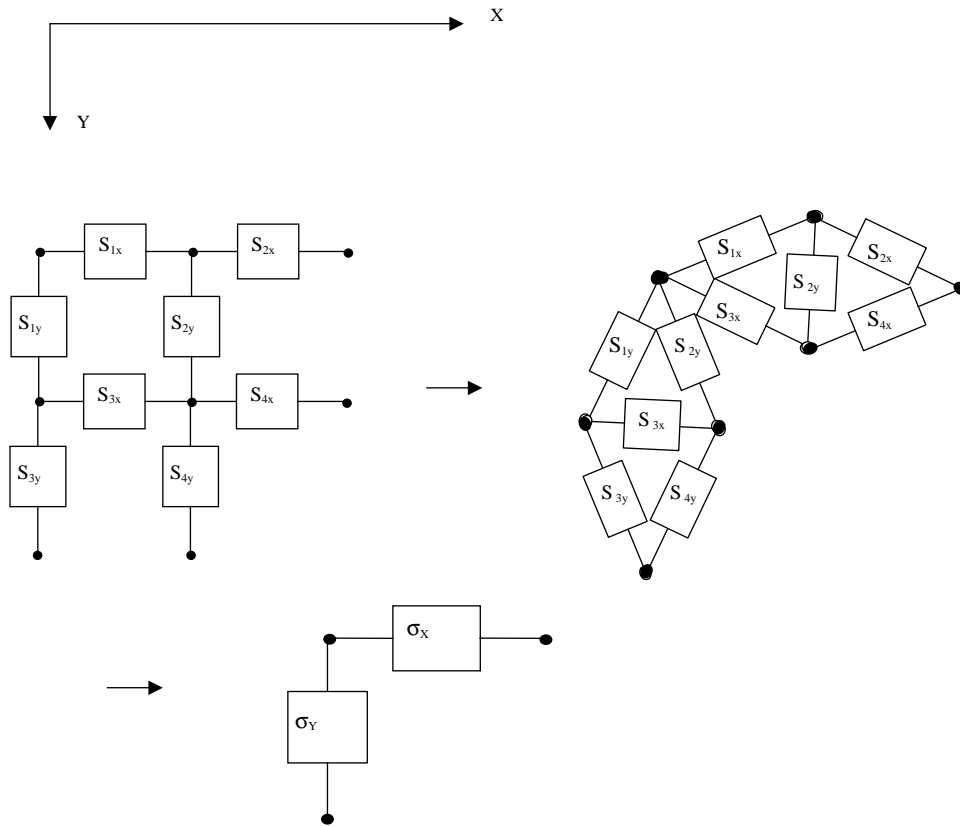


Figure 1. The real-space renormalization group for a square network.

this method is an approximation, it has been shown to give values of the effective conductivity near the percolation threshold very close to the exact ones for 2D composites [29] and critical exponents not far from the values known from percolation theory [12]. Furthermore, this method uses only a few matrices of size  $M \times M$  for sample sizes  $M$ , which considerably reduces the computational memory requirement in comparison to the other methods. Then we can easily reach sizes of  $1024 \times 1024$  with the same computer configuration as for the previous method (the EM). However, the first method (the EM) is also needed in the present work, since the ability of the RSRG method to determine the local field has not been checked before.

As discussed in the previous section, the localization properties of the optical waves in thin metal–dielectric composites seem to provide an interesting way to explain the anomalous absorption observed near the percolation threshold. One of the useful quantities for studying the localization properties in electronic systems is the inverse participation ratio (IPR) [23]. By analogy with the quantum counterpart, the local electric field in the Helmholtz equation plays the role of the electronic wave-function in the Schrödinger equation [7] and the IPR reads

$$\text{IPR} = \sum_i |E_i|^4 / \left( \sum_i |E_i|^2 \right)^2 \quad (5)$$

where  $E_i$  denotes the local electric field at site  $i$ . The IPR has been defined for the electronic wave-function in order to measure the spatial extent of the dominant eigenstates

and to characterize the electronic states in disordered materials [23]. Therefore, for the electromagnetic eigenmodes this quantity will behave as

$$\text{IPR} = O(M^{-d}) \quad \text{for purely extended eigenmodes} \quad (6)$$

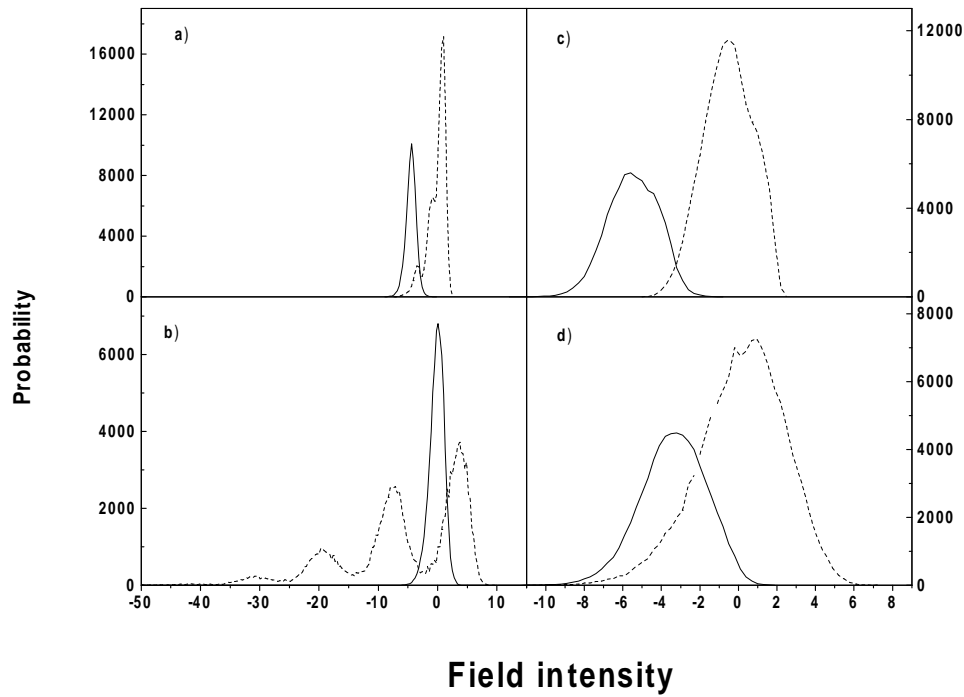
$$\text{IPR} = O(M^0) \quad \text{for strongly localized eigenmodes.} \quad (7)$$

Here  $d$  denotes the Euclidean dimension of the system ( $d = 2$  in this case) and  $M$  the size of the system. Thus in the case of purely extended eigenmodes, the field has the same strength over the whole surface of the film and the denominator will be  $M^4|E|^4$  while the numerator is  $M^2|E|^4$  (assuming the field to be constant) leading to a decay of the IPR as  $M^{-2}$ . In the case of strongly localized eigenmodes, the more significant field strengths are located in a limited area of average size  $M_c^2$  (where  $M_c$  is the localization length). It is then obvious that the IPR remains constant outside this area. Therefore we can estimate the degree of localization of the local field from the power-law decay exponent of the IPR which varies from 0 (for strongly localized eigenmodes) to  $-2$  (for purely extended eigenmodes). This exponent is determined by the slope of the variation of the IPR as a function of the system size on a log–log scale. It measures also the correlation fractal dimension ( $-D(2)$ ) of the local field [14, 30].

### 3. Results and discussion

In this section we consider a film of size  $512 \times 512$ . As discussed above, the RSRG method is a good approximation for the calculation of the effective conductivity and the critical exponents. Indeed, in a recent work by Brouers *et al* [29], the real conductivity calculated by this method was in good agreement with that of the Lobb and Frank method [28]. However, this agreement cannot be generalized to any other quantity. In particular, the value of the effective dc conductivity at the percolation threshold depends only on the appearance of an infinite metallic cluster channel in the sample, no matter how the bonds are distributed over the sample [11, 12]. The local field strength (which is the main quantity measuring the localization properties of the system) depends strongly on the local configuration of the bonds. Thus, some particular arrangements of the bonds yield a local field intensity ( $|E|^2$ ) of the order of  $R^{-2}$  via the RSRG method. Therefore, in the limit of vanishing losses the field intensity diverges which is unphysical (this divergence should be compensated by the dispersion of the current through the connected neighbouring bonds, and should then occur only for an isolated two-bond  $RL-C$  circuit). This overestimation of the local field can seriously limit the validity of the RSRG method for describing the localization properties of these films, particularly for vanishing losses.

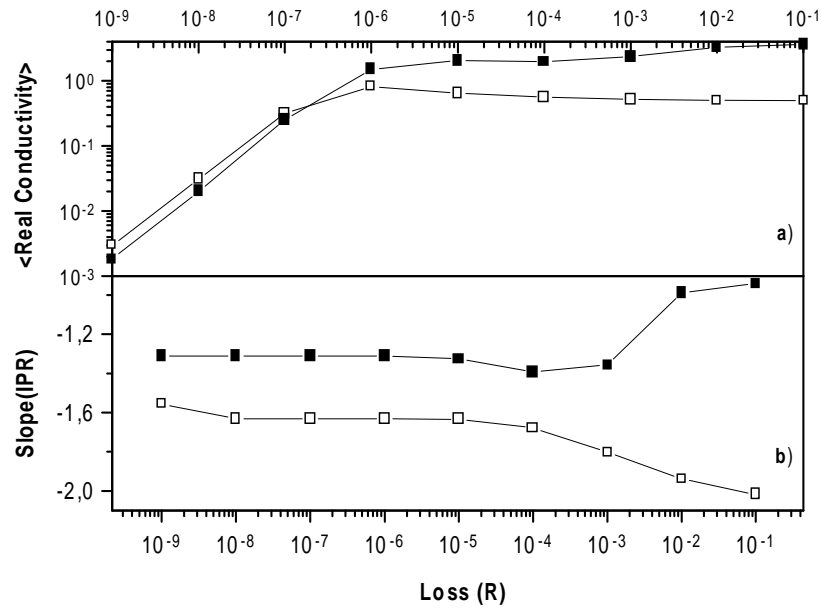
In figures 2 we compare the distributions of the logarithm of the local field intensity obtained by the RSRG method to those from the EM for sample sizes  $512 \times 512$ , for two different losses and two different frequencies, in order to check the validity of the RSRG method in calculating the local field. The RSRG distributions seem to be wider with much larger field strengths than the EM distributions (supporting the previous discussion on the overestimation of the field by the RSRG method) which seem to be perfectly log–normal for any loss and frequency. Furthermore, other peaks of small field strengths appear in the distributions obtained by the RSRG method, particularly for small losses and at the characteristic frequency ( $\omega = 1$ ), while for larger losses and different frequencies these peaks seem to move to larger field strengths and overlap with the main one, contributing to its broadening. This behaviour strongly affects the IPR calculations since the broadening of the field distribution means an increase of the degree of localization [19, 20]. However, the additional peaks appearing for small losses in the RSRG method should contribute only slightly to the IPR



**Figure 2.** The distribution of the local field intensity  $\log(|E|^2)$  for  $\omega = 1$ : (a)  $R = 10^{-1}$ , (b)  $R = 10^{-6}$ ; and  $\omega = 1/8$ : (c)  $R = 10^{-1}$ , (d)  $R = 10^{-6}$ . Solid curves correspond to the EM calculations and dashed curves to the RSRG method.

since they are many orders of magnitude smaller than the main peak. Therefore, although the distributions of the field for the RSRG method are clearly different from the exact ones (from the EM), it seems that the results from the renormalization group method for  $\omega = 1$  fit better the localization properties of these films in comparison with those for other frequencies. Therefore, we will restrict ourselves to the characteristic frequency ( $\omega = 1$ ) for the rest of the paper.

In figures 3, by using both the RSRG method and the EM, we show the real part of the conductivity averaged over 100 samples of size  $512 \times 512$  (figure 3(a)) and the slope of the IPR (figure 3(b)) as functions of the loss parameter  $R$ . In figure 3(a) the conductivity tends to vanish as a power law for small losses with an exponent close to 1 and a non-monotonic behaviour due to strong statistical fluctuations for  $R < 10^{-4}$ , while it tends to saturate at  $1/2$  for larger losses (where the conductivity seems to be self-averaged). This behaviour for vanishing losses supports our previous discussion on the dissipation of the film in this limit. It disagrees then with the effective-medium predictions (equation (1)) where the real conductivity should saturate at 1 for  $R \rightarrow 0$ . This discrepancy can be explained by the correlation length which should become greater than the sample size or by its divergence for vanishing losses where the effective-medium relation (1) is not valid. Although a slight difference is shown between the two methods in figure 3(a), they follow qualitatively the same behaviour for small losses, confirming the validity of the RSRG method for the calculation of the conductivity. However, the IPR shows a delocalization (for the RSRG method) as the loss increases; this is opposite to the case for the EM which yields the inverse behaviour (see figure 3(b)). Indeed, for the same configuration, an increase of the loss corresponds to



**Figure 3.** (a) The real part of the effective conductivity in a log–log plot and (b) the slope of the IPR in a semi-log plot as functions of the loss parameter  $R$  for  $\omega = 1$ . Open squares correspond to the RSRG method and solid squares to the EM. The conductivity is averaged over 50 samples and the IPR is calculated for only one configuration.

an increase of the absorption as shown in figure 3(a), which enhances the localization [10]. This confirms the failure of the renormalization group method in describing the localization properties of the system, due to the overestimated field strengths and fluctuations obtained by this method which broaden the distributions shown in figure 2. On the other hand, the IPR exponent obtained by the EM seems to saturate at the value  $-1.3$  in the region of small losses (see figure 3(b)) indicating that the eigenmodes are not purely extended even for vanishing losses (from (6), an exponent  $-2$  corresponds to a constant field through the film while the exponent  $-1.3$  corresponds to clusters of large field strengths appearing in different regions of the whole film). Note that in the limit  $R = 0$  the localization is due only to the disorder in the constituent conductivities (Anderson-like weak localization). The disorder here does not come from the strength of the conductivity (since the conductivities of the two components have the same strength at zero loss) but from its phase which takes randomly two values:  $+\pi/2$  for the metal from (3) and  $-\pi/2$  for the dielectric component from (4) for vanishing losses, while for larger losses, in addition to the dissipation, this Anderson localization is enhanced as the degree of disorder is enhanced by the difference between the conductivities of the two components ( $||\sigma_m| - |\sigma_d||$ ) which becomes finite. Therefore, even for vanishing losses, where the system becomes non-dissipative, the field energy remains confined in regions of the film where it resonates between the superconductor  $L$  and the dielectric  $C$ .

As shown for the conductivity, the IPR in the region of loss between  $10^{-3}$  and  $10^{-6}$  shows also a non-monotonic behaviour, from figure 3(b). This is probably a signature of a phase transition from localized to extended eigenmodes. It seems that this phase transition can be characterized by the statistical properties of the conductivity and the electric field. Making an analogy with the conductance fluctuations in quantum systems is then possible.



#### 4. Conclusions

In this article we have studied, by using the RSRG method and the EM, the localization and absorption properties of the electromagnetic field in a thin semicontinuous metal–dielectric film for a characteristic frequency  $\omega_{res} = 1$  at the classical percolation threshold. It seems that the results for the real part of the effective conductivity within the RSRG method agree qualitatively with the exact calculations (by the EM) and yields the expected power-law behaviour for vanishing loss which is a scaling effect. However, this RSRG method fails to provide the right behaviour of the IPR, due to the large field strengths and fluctuations obtained in comparison with the EM results. On the other hand, the IPR saturates at  $-1.3$  for vanishing losses, indicating localized clusters of large field strengths through the whole system. Therefore, we can explain the anomalous absorption observed by a confinement of the electric field in small regions of the film at the percolation threshold, which corresponds to a resonating oscillation between the components of the system. We found also in figure 3(a) that the effective conductivity strongly fluctuates in the region of the loss between  $10^{-3}$  and  $10^{-6}$  while it is self-averaged for larger losses. This motivated an extensive study of the statistical properties of the effective conductivity for these losses and for different metallic concentrations. This investigation is described in the next paper. It is also important to examine the effect of local arrangement of the metal (existing in realistic films) on these properties. In this case even the percolation concept should change.

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#### References

- [1] Gadenne P, Beghadi A and Lafait J 1988 *Opt. Commun.* **65** 17
- [2] Yagil Y, Yosefin M, Bergman D J, Deutscher G and Gadenne P 1991 *Phys. Rev. B* **43** 11 342
- [3] Yagil Y, Gadenne P, Julien C and Deutscher G 1992 *Phys. Rev. B* **46** 2503
- [4] Sarychev A K, Bergman D J and Yagil Y 1995 *Phys. Rev. B* **51** 5366
- [5] Brouers F, Clerc J P, Giraud G, Laugier J M and Randriamantany Z A 1993 *Phys. Rev. B* **47** 666
- [6] Bruggeman D A 1935 *Ann. Phys., Lpz.* **24** 636  
Landauer R 1978 *Electrical Transport and Optical Properties of Inhomogeneous Media (AIP Conf. Proc. 40)* ed J C Garland and D B Tanner (New York: AIP) pp 2–43  
Niklasson G A and Granqvist C G 1984 *J. Appl. Phys.* **55** 3382
- [7] John S 1988 *Comment. Condens. Matter Phys.* **14** 193
- [8] Lee P A and Ramakrishnan T V 1985 *Rev. Mod. Phys.* **57** 287
- [9] Genack A Z 1987 *Phys. Rev. Lett.* **58** 2043  
Genack A Z and Garcia N 1991 *Phys. Rev. Lett.* **66** 2064
- [10] Gupta A K and Jayannavar A M 1995 *Phys. Rev. B* **52** 1456  
Yosefin M 1994 *Europhys. Lett.* **25** 675
- [11] Stauffer D and Aharony A 1994 *Percolation Theory* 2nd edn (London: Taylor and Francis)  
Efros A L and Shklovskii B I 1976 *Phys. Status Solidi* **76** 475
- [12] Bergman D J and Stroud D 1992 *Solid State Physics* vol 46 (New York: Academic) p 147
- [13] Fritzsche H and Pollak M (ed) 1990 *Hopping and Related Phenomena* (Singapore: World Scientific)  
Pollak M and Shklovskii B 1991 *Hopping Transport in Solids* (Amsterdam: Elsevier–North-Holland)  
Chen I-G and Johnson W B 1992 *J. Mater. Sci.* **27** 5497  
Gupta A K and Sen A K 1998 *Phys. Rev. B* **57** 3375
- [14] Brouers F, Blacher S and Sarychev A K 1995 *Fractals in the Natural and Applied Sciences* (London: Chapman and Hall) p 237
- [15] Stockman M I, Pandey L N, Muratov L S and Georges T F 1994 *Phys. Rev. Lett.* **72** 2486

- [16] Markel V A, Shalaev V M, Stechel E B, Kim W and Armstrong R L 1996 *Phys. Rev. B* **53** 2425
- [17] Shalaev V M, Poliakov E Y and Markel V A 1996 *Phys. Rev. B* **53** 2437
- [18] Landauer R 1970 *Phil. Mag.* **21** 263
- [19] Chase K S and Mackinnon A 1987 *J. Phys. C: Solid State Phys.* **20** 6189  
Sheng P and Zhang Z 1991 *J. Phys.: Condens. Matter* **3** 4257  
Markos P and Kramer B 1993 *Ann. Phys., Lpz.* **2** 339
- [20] Senouci K, Zekri N and Ouasti R 1996 *Physica A* **234** 23
- [21] Sarychev A K 1977 *Zh. Eksp. Teor. Fiz.* **72** 1001  
Bernasconi J 1978 *Phys. Rev. B* **18** 2185
- [22] Brouers F, Blacher S and Sarychev A K 1998 *Phys. Rev. B* **58** 15 897
- [23] Bell R J and Dean P 1970 *Discuss. Faraday Soc.* **50** 55  
Kramer B and Weaire D 1979 *Amorphous Semiconductors* ed M H Brodsky (Berlin: Springer) p 9  
Wegner F J 1980 *Z. Phys. B* **36** 209
- [24] Zeng X C, Hui P M and Stroud D 1989 *Phys. Rev. B* **39** 1063
- [25] Curtin W A and Ashcroft N W 1985 *Phys. Rev. B* **31** 3287
- [26] Clerc J P, Giraud G and Luck J M 1990 *Adv. Phys.* **39** 191
- [27] Zekri L 1998 *MSc Thesis* USTO, Oran, Algeria
- [28] Frank D J and Lobb C J 1988 *Phys. Rev. B* **37** 302
- [29] Brouers F, Sarychev A K, Blacher S and Lothaire O 1997 *Physica A* **241** 146
- [30] Mato C and Caro A 1989 *J. Phys.: Condens. Matter* **1** 901