LOCALIZATION AND ABSORPTION OF LIGHT
IN 2D COMPOSITE METAL-DIELECTRIC FILMS
AT THE PERCOLATION THRESHOLD

L. Zekri
U.S.T.O., Departement de Physique, L.E.P.M.,
B.P.1505 El M’Naouar, Oran, Algeria
and
The Abdus Salam International Centre for Theoretical Physics, Trieste, Italy,

R. Bouamrane
U.S.T.O., Departement de Physique, L.E.P.M.,
B.P.1505 El M’Naouar, Oran, Algeria,

N. Zekri
U.S.T.O., Departement de Physique, L.E.P.M.,
B.P.1505 El M’Naouar, Oran, Algeria
and
The Abdus Salam International Centre for Theoretical Physics, Trieste, Italy

and

F. Brouers
Universite de Liege, Institut de Physique, Etude Physique des Materiaux,
Sart Tilman 4000, Liege Belgium.

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1Regular Associate of the ICTP.
Abstract

We study in this paper the localization of light and the dielectric properties of thin metal-dielectric composites at the percolation threshold and around a resonant frequency where the conductivities of the two components are of the same order. In particular, the effect of the loss in metallic components are examined. To this end, such systems are modelized as random $L - C$ networks, and the local field distribution as well as the effective conductivity are determined by using two different methods for comparison: an exact resolution of Kirchoff equations, and a real space renormalization group method. The latter method is found to give the general behavior of the effective conductivity but fails to determine the local field distribution. It is also found that the localization still persists for vanishing losses. This result seems to be in agreement with the anomalous absorption observed experimentally for such systems.
Introduction

Thin metal-dielectric films have been shown experimentally to exhibit an anomalous high absorption in the visible, near infrared and microwave regimes and near 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 percolation threshold where

$$\epsilon_{eff} = \sqrt{\epsilon_m \epsilon_d}$$  \hspace{1cm} (1)

The indices $m, d$ and $eff$ stand respectively for the metal, dielectric and effective medium. From this equation, both real and imaginary parts of the dielectric constant of the metallic component contribute to the optical absorption of the system. Furthermore, a vanishing metallic absorption naturally should lead to a vanishing absorption of the whole system since a superconductor-dielectric mixture should be non dissipative. Therefore, this description of the effective properties of such medium may not be sufficient to explain the behavior experimentally observed for such systems [1-4].

Indeed, since the light has a wave behavior, the backscattering and the interference effects can strongly affect its propagation through a disordered system [7, 8, 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 both absorption and disorder are present, can be a good tool for explaining the anomalous behavior observed in such thin composite films, particularly at the classical percolation threshold which is a transition point of the effective $dc$ conductivity of the system from non-conductor to conductor due to the appearance of a continuous path of the conducting region through the sample. The classical percolation threshold in 2D disordered bonds corresponds to a concentration of the metallic bonds $p_c = 0.5$ [11] (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 some virtually connected percolating clusters [12]).

On the other hand, it has recently been found numerically in such films, giant local field fluctuations [13] at the percolation threshold and for frequencies close to a resonant one $\omega_{res}$ where the conductivities of the two components are of the same order ($|\sigma_m| = |\sigma_d|$). High local field fluctuations have also been found both in fractal 2D films [14], 3D rough surfaces [15] and non-linear Raman scattering [16]. In both systems the
electromagnetic modes were found to be localized, apparently, due to such fluctuations. Furthermore, Brouers et al. [13] 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 deduce, then, that the local transmission has also a log-normal distribution in such films. Therefore, if we use the analogy between the electric field in Helmholtz equation and the electronic wave-function in Schrödinger equation [7], the local transmission is equivalent to the electronic conductance at zero temperature [17] where a log-normal distribution is a signature of localization [18, 19].

This is the aim of the present paper where we study the localization and absorption properties of such films modelized by a square $RL – C$ network. The local field and the effective conductivity are calculated by using two different methods for comparison with the results of [13]: an exact resolution of the Kirchoff equations for such network which we call Exact Method (EM) from now on, and a Real Space Renormalization Group (RSRG) self-similar scheme [20]. The degree of localization is measured by means the inverse participation ratio (IPR) [21] applied to the electric field while the absorption is deduced from the real part of the effective conductivity. We compare in a first step the field distribution obtained by the two methods and then examine the effect of the loss in the metallic component on the localization as well as the absorption at the percolation threshold.

Method of the calculations

The conductivity of metallic and dielectric grains is related to their dielectric constant by [4]

$$\sigma_{m,d} = \frac{-i\omega \epsilon_{m,d}}{4\pi}$$

(2)

where $d$ is the film thickness, $\omega$ the field frequency and $\epsilon_{m,d}$ the dielectric constants respectively of the metal and dielectric components assumed to be homogeneous spheres. Here the film thickness and the size of the components must be smaller than the light wavelength in order to neglect the magnetic field variation. The dielectric constant of the insulator is real while the metallic one is complex and, from Eq.(2), its imaginary part (absorption) is related to the real part of the conductivity.

When the light frequency is large compared to the relaxation frequency, composite metal dielectric films can be modelled by 2D resistor networks [4, 5, 22]. The effective properties of these networks have already been extensively studied during the
last two decades [5,6,23-25]. In the $RL - C$ picture, and if the frequency is smaller than
the plasmon frequency $\omega_p$, capacitors $C$ stand for the dielectric grains with a conductivity $\sigma_C = -iC\omega$ and a concentration $1 - p$ while inductances $L$ represent the metallic grains with a conductivity $\sigma_L = (-iL\omega + R)^{-1}$ ($R$ being the loss), and a concentration $p$ de-
posited or evaporated over the substrate. Therefore we can take in this case, without loss
of generality, $L = C = \omega_{\text{res}} = 1$ (the resonant frequency $\omega_{\text{res}}$ corresponds to the frequency
where metallic and dielectric conductivities have the same magnitude for small losses, i.e.,
$C\omega_{\text{res}} = 1/L\omega_{\text{res}}$). $L$ and $C$ are constants near the resonant frequency, this assumption
can be generalized to any frequency $\omega$ (smaller than the plasmon frequency) normalized
to $\omega_{\text{res}}$. The metallic conductivity for small losses becomes

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

while the dielectric conductivity is

$$
\sigma_C = -i\omega
$$

The first method (EM), used for the calculation of the local field dist-
ribution and the effective conductivity, consists in solving exactly Kirchhoff equations for the corre-
sponding 2D square resistor network. This implies the use of $M^2 \times M^2$ matrices (where
$M$ is the size of the square lattice), which are impossible to handle numerically for large
samples (for memory and computational time consuming reasons). However, we take
advantage of the sparse configuration of such matrices and their organization in blocks
in their diagonal region. The diagonalization and inversion of such matrices, then, is
obtained simply from the diagonalization and inversion of the constituent blocks. This
reduces considerably the memory and the computational time consuming. We note that
this method provides exactly the same results on the effective conductivity as the Frank
and Lobb method [25] but calculates also the local field distribution through the lattice
which cannot be done by the other method. However, the time consuming remains large
with this method (in particular when averaging over a large number of configurations),
and the maximum size we reach by this method is 256 x 256 (which is sufficient for our
statistical treatment).

This is one of the reasons for using also the RSRG method which is much less
computational time consuming. This method, extensively described in previous works
(see [13, 20, 26]), consists in a transformation of the 2D square lattice into Weatstone
bridges in $x$ and $y$ directions (see Fig.1). Each bridge is transformed into an equivalent
admittance, and after a number of steps the lattice is reduced into two equivalent ad-
mittances following these directions. It is then easy to calculate by 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 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 [3, 27] and critical exponents not far away from the known values of the percolation theory [11]. Furthermore, this method uses only few matrices of $M \times M$ for sample sizes $M \times M$ which reduces considerably the computational memory in comparison with the other methods. We can, then, easily reach sizes of 1024x1024 with the same computer configuration as for the previous method. However, the first method (EM) is also needed in the present work since the validity of RSRG in calculating the local field has not been checked before.

As discussed in the previous section, it seems that the localization properties of the optical waves in thin metal-dielectric composites is an interesting way to explain the anomalous absorption observed near the percolation threshold. This classical threshold corresponds to the appearance of an infinite metallic channel which, in 2D disordered systems, is reached for an equal probability of the two components [11]. One of the useful quantities to study the localization in electronic systems is the inverse participation ratio (IPR) [21]. By analogy with the quantum counterpart, the local electric field in Helmholtz equation plays the role of the electronic wave function in the Shrödinger equation [7] and the IPR becomes

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

where $E_i$ denotes the local electric field at site $i$. The IPR has been defined for the electronic waves in order to measure the spatial extent of the dominant eigenstates and to characterize the electronic states in disordered materials [21]. Therefore, for the electromagnetic eigenmodes this quantity will behave as

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

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

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 a significant strength over the whole surface of the film and the denominator will be $M^4 |E|^4$ while the numerator behaves as $M^2 |E|^4$ (by assuming the field 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 light 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 the system size in log-log scale. It also measures the correlation fractal dimension ($-D_2$) of the local field [13, 27]. From now on we will call this exponent the slope of the IPR.

**Results and Discussion**

In this section we consider a film of size 256x256. As discussed above, the RSRG method is a good approximation for the calculation of the effective conductivity and the critical exponents. However, this agreement cannot be generalized to any other quantity. In particular, the value of the effective conductivity at the percolation threshold is due only to the appearance of an infinite metallic cluster channel in the sample no matter how the bonds are distributed over the sample [28, 11]. However, the distribution of bonds can affect sensitively the local field distribution which is the main quantity to measure the localization properties of light in this system. Thus, some particular arrangements of the bonds yield a local field intensity ($|E|^2$) of the order of $R^{-2}$ by the RSRG method. Therefore, in the limit of vanishing losses the field intensity will diverge which is unphysical. This divergence can limit the validity of the RSRG method for vanishing losses.

In Figs.2 we compare the distributions of the local field intensity obtained by the RSRG method to those of the EM for sample sizes 256x256, for two different losses and two different frequencies in order to check the validity of RSRG in calculating the local field. The RSRG distributions seem to be wider, with very large field strengths, than for the EM distributions (supporting the previous discussion on the divergence of the field) which seem to be perfectly log-normal for any loss and frequency. Furthermore, other peaks of small field strengths appear in the distribution obtained by RSRG particularly for small losses and at the resonant frequency $\omega = 1$, while for higher losses and different frequencies, these peaks move to larger field strengths and overlap with the main one contributing to the broadening of the distribution. This behavior affects strongly the IPR calculations since the broadening of the distribution means an increase of the localization [18, 19]. However, the additional peaks appearing for small losses in the RSRG method should contribute only slightly to the IPR since they are many orders smaller than the main peak. Therefore, although the distributions of field for RSRG are clearly different...
from those of the EM, it seems that the results for $\omega = 1$ provide the best fit for the localization properties of these films. Therefore, we will restrict ourselves to the resonant frequency ($\omega = 1$) from now on.

In Figs.3, we show, by using both RSRG and EM methods, the real part of the conductivity averaged over 50 samples of size 256x256 (Fig.3a) as well as the slope of the IPR (Fig.3b) as a function of the loss parameter $R$ for $\omega = 1$. It seems that the conductivity tends to vanish as a power-law for small losses with an exponent close to 1 while it tends to saturate for larger losses with strong fluctuations in the region of the loss between $10^{-3}$ and $10^{-6}$ (while for smaller or larger losses the conductivity seems to be self averaged). The behavior for small losses is in agreement with the theoretical predictions. Indeed, for vanishing losses both dielectric and metallic components of the film are non-dissipative and then the whole system becomes non-dissipative. Furthermore, from Eqs. (1-4), the real part of the effective conductivity should behave as

$$\text{Real}(\sigma_{\text{eff}}) = \frac{1}{2} \frac{R}{\omega}$$

which seems to be well fitted in Fig.3a. Although a difference is shown between the two methods, they follow qualitatively the same behavior for small losses. However, as expected from Figs.2, the IPR shows a delocalization (for the RSRG method) for increasing loss while the EM method yields the inverse situation which is the expected behavior (for the same configuration, an increase of the loss corresponds to an increase of the absorption which means a localization [19]). Therefore, this confirms the failure of this renormalization group method in describing the localization properties of the system, due to the large field strengths obtained by this method which broaden the distributions shown in Fig.2. On the other hand, the IPR slope obtained by the EM method seems to saturate at the value $-1.3$ in the region of small losses (see Fig.3b) indicating that the eigenmodes remain localized even for vanishing losses. Indeed, this localization is due only to the disorder in the conductivity (Anderson like localization). The disorder here does not come from the strength of the conductivity (since the conductivities of the two components have the same strength) but from its phase which takes randomly two values: $+\frac{\pi}{2}$ and $-\frac{\pi}{2}$ for vanishing losses. We also see from this Figure, in the region of loss between $10^{-3}$ and $10^{-6}$, a non monotonic behavior of the IPR (EM). This is due to the strong fluctuations observed in this region. Therefore, this region should show a different statistical behavior.
Conclusion

In this article we have studied, by using the RSRG method and also the EM method, the localization and absorption properties of the electromagnetic field in a thin semicontinuous metal-dielectric film for a characteristic frequency $\omega_{\text{res}} = 1$ at the classical percolation threshold. It seems that the real part of the effective conductivity for RSRG agrees qualitatively with the exact calculations (EM) and give the expected power-law behavior for vanishing loss. While this method (RSRG) fails in giving the right behavior for the IPR, due to the large field strengths provided in comparison with the EM method. On the other hand, it seems that the IPR saturates at $-1.3$ for vanishing losses in agreement with the anomalous experimental results observed for such films. We can explain the anomalous absorption observed by a confinement of the light at the percolation threshold. We also found that the conductivity strongly fluctuates in the region of the loss between $10^{-3}$ and $10^{-6}$ while it is self averaging for larger or smaller losses. An extensive study of the statistical properties of the conductivity is then needed. Indeed, may be in that region the logarithm of the conductivity is self averaged in this region. Furthermore, it should be interesting to study these effective properties around the classical threshold where interesting features can occur. It is also important to examine these effects for a partially ordered sample since realistic films show a local arrangement. All these interesting features will be the subject of a forthcoming investigation.

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Figure Captions

Fig.1 The real space renormalization group for a square network.

Fig.2 The distribution of the local electric 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 RSRG method.

Fig.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 a function of the loss parameter $R$ for $\omega = 1$. Open squares correspond to the RSRG method and solid squares to the EM method. The conductivity is averaged over 50 samples and the IPR is calculated for only one configuration.
FIGURE 1
FIGURE 2

Field intensity
FIGURE 3