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1994 J. Phys.: Condens. Matter 6 10607

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## Scaling theory for the optical properties of thick percolative metal–insulator films

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Received 9 May 1994, in final form 31 August 1994

**Abstract.** The 2D scaling approach for the optical properties of semicontinuous metal films is generalized to thick (3D) percolative films. Model calculations demonstrate that the scaling model is qualitatively different from any effective medium approach, and should be used whenever the inhomogeneity length scale is larger than the optical one.

The optical properties of composite materials are normally described by effective medium models, based on the static approximation (see [1] and references therein). It is now well known that these approximations are invalid over a wide range of the filling factor  $p$ , both above and below the percolation threshold  $p_c$  (the metal–insulator transition). The reason for this discrepancy is the divergence of the percolation correlation length  $\xi$ , which becomes larger than all other relevant length scales. In contrast, the optical properties of (2D) semicontinuous metal films, near the percolation threshold, are well described by a scaling based model [1, 2]. In this work we present the generalization of the 2D scaling model to thick percolative films, where ‘thick’ is defined below. This model should be applicable to metal–dielectric cermet, such as co-evaporated or co-sputtered Ag–SiO<sub>2</sub>, Au–Al<sub>2</sub>O<sub>3</sub>, and other composites.

The scaling model for the optical properties of 2D semicontinuous metal films consists of three main assumptions. (i) The optical response of a percolative film is determined by its fractal geometry. Therefore, the relevant length scale  $L(\omega)$  is much smaller than the optical wavelength  $\lambda = 2\pi c/\omega$  and is determined by the anomalous diffusion relation [3]  $L(\omega) \propto \omega^{-1/(2+\theta)}$ , where  $1/(2+\theta) \simeq 0.35$  in 2D and 0.27 in 3D. (ii) One cannot define an effective dielectric constant if  $L(\omega) < \xi$ . Instead, the optical responses of different areas of linear size  $L(\omega)$  should be averaged, using finite-size scaling and a wide distribution of the local (complex) dielectric constant. (iii) The contribution of DC insulating regions is not negligible in the AC case due to intercluster capacitance.

We start with the identification of the optical length scale  $L(\omega)$  and its meaning for thick (3D) films. Similar to the 2D case, the AC current correlation length is given by the distance an electron may diffuse over a time period  $1/\omega$ . At low frequencies (normally in the IR and FIR regimes) the intercluster and intracluster capacitive impedance is larger than the metallic impedance, hence this length scale is determined by the anomalous diffusion relation in large metallic clusters. In the 3D case, this length scale is also a lower bound

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for the AC current penetration depth. We emphasise that this length scale should not be confused with the skin depth, which measures the penetration depth of the electric field in a homogeneous medium. In some sense,  $L(\omega)$  is analogous to the anomalous skin effect, where the intrinsic properties (the mean free path) dominate the actual penetration depth. In this stage the term 'thick' becomes obvious: a film is thick and 3D if its thickness  $d$  is larger than  $L(\omega)$ . Note that this condition is much weaker than the usual definition for a 3D percolative film:  $d > \xi$ . Since the local AC current is uniform over a length scale  $L(\omega)$ , the local (complex) AC conductivity should be calculated over the same length scale. We thus divide the film into *cubes* of linear size  $L(\omega)$ , and calculate the complex AC conductivity of each cube. Similar to the 2D case, the average conductivity of the conducting and the insulating regions is determined by finite-size scaling, and a wide bimodal distribution function is used to describe all possible (complex) conductivities. In the 2D case the optical response (namely  $R$ ,  $T$  and  $A$ ) of each square of linear size  $L(\omega)$  is calculated separately, and the film response is given by averaging these different contributions. In the 3D case, each square is replaced by a pile of cubes (a column) of area  $L(\omega) \times L(\omega)$  and thickness  $d$ . The optical response of such a column depends on all the cubes of linear size  $L(\omega)$  and their order. The optical response of the entire film is the average of the contributions of all possible column configurations with their appropriate weights.

The average conductivity of a good conductor-bad conductor mixture of linear size  $L \ll \xi$ , near the percolation threshold of the good conductor, is given by [4]

$$\sigma_{av}(L) = \sigma_m L^{-\mu/\nu} F[(\sigma_i/\sigma_m)L^{(\mu+s)/\nu}] \quad L \ll \xi \quad (1)$$

where  $\sigma_m$  and  $\sigma_i$  are the complex conductivities of the good and the bad conductors, respectively,  $\mu$  and  $s$  are their critical exponents, and  $\nu$  is the critical exponent of the percolation correlation length  $\xi = \xi_0(p - p_c)^{-\nu}$ . All the lengths are measured in units of the typical grain size  $a$ , thus both  $L$  and  $\xi$  are dimensionless. The scaling function  $F(z)$  has two limiting forms for  $|z| \ll 1$ , depending on the existence of a continuous path of the good conductor [4, 1]:

$$F(z) = \begin{cases} A_1 + A_2 z & p > p_c \\ A_3 z + A_4 z^2 & p < p_c. \end{cases} \quad (2a)$$

$$(2b)$$

The critical exponents appearing in (1) have universal values that depend only on the dimensionality of the system. In three dimensions  $\nu = 0.88$ ,  $\mu = 2.0$  and  $s = 0.73$ , for both site and bond percolation [5]. Continuum percolation may cause some changes in these values, but it is normally assumed that they are universal. The non-universal coefficients, such as  $\xi_0$ ,  $p_c$ ,  $L_0$  and  $A_{1-4}$  depend on the details of the microgeometry. Their values cannot be determined from the scaling theory of percolation and must be adjusted to the particular system in question (see [1]). For example,  $p_c$  may be measured by the DC resistance, and  $\xi_0$  is roughly the grain size.  $L_0$  and  $A_{1-4}$  are prefactors of order unity and are normally adjusted by fitting the experimental data. We note, however, that these prefactors are not free parameters of the model and could thus be measured independently.

On a length scale  $L < \xi$ , the sample appears highly inhomogeneous, hence the local AC conductivity has large fluctuations. In particular, *both metallic-like and dielectric-like conductivities are present*, depending on whether a (locally) conducting path exists or is absent. This may be described by a bimodal distribution function  $P(\sigma)$ , where (2a) and (2b) are the average conductivities of the metallic-like and the dielectric-like regions, respectively. For  $p \neq p_c$  the conductivity distribution function *also depends on*

$L$ , hence  $P = P(\sigma, L/\xi)$ . As long as  $L \ll \xi$ ,  $P(\sigma, L/\xi) \approx P(\sigma, 0)$ . For  $L \gg \xi$   $P(\sigma, L/\xi) \approx \delta(\sigma - \sigma_{avr}(\xi))$ , since the sample appears homogeneous on length scales larger than the percolation correlation length. The actual shape and width of  $P(\sigma)$  were numerically studied for the 2D DC case, and found to be universal [8]. For 3D  $P(\sigma)$  should still be universal, but could differ from the 2D function. The general conclusions of the following discussion do not depend on a particular choice of  $P(\sigma)$ . However, the actual values of  $R$ ,  $T$  and  $A$  do depend on this function, hence  $P(\sigma)$  for 3D is needed in order to fit experimental data. Alternatively, optical data might be used in order to estimate  $P(\sigma)$ .

The local dielectric constant of an  $L \times L \times L$  cube of local AC conductivity  $\sigma$  is given by  $\epsilon = 1 + i4\pi\sigma/\omega$ . Following the 2D scaling model, the optical response of such a cube is given by the response of a thin (homogeneous) film having the same dielectric constant. This is fully determined by its characteristic matrix [6]

$$\mathbf{M} = \begin{pmatrix} \cos \beta & (i/g) \sin \beta \\ ig \sin \beta & \cos \beta \end{pmatrix} \quad (3)$$

where  $g = \sqrt{\epsilon/\mu}$  (with the convention of negative imaginary part, i.e., the time dependence is  $e^{i\omega t}$ ),  $\beta = kd$ ,  $k$  is the complex wavenumber and  $d$  the film thickness ( $d = L$  in this case). The characteristic matrix of a column of  $N$  cubes is given by

$$\tilde{\mathbf{M}} = \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix} = \prod_{i=1}^N \mathbf{M}_i \quad (4)$$

and the reflected and transmitted amplitudes are

$$\left. \begin{aligned} r &= [g_0(m_{11} + g_s m_{12}) - (m_{21} + g_s m_{22})] / [g_0(m_{11} + g_s m_{12}) + (m_{21} + g_s m_{22})] \\ t &= 2g_0 e^{2\pi i n_0 D/\lambda} / [g_0(m_{11} + g_s m_{12}) + (m_{21} + g_s m_{22})] \end{aligned} \right\} \quad (5)$$

Here  $g_0$ ,  $g_s$  represent the vacuum and the substrate refractive indices, respectively, and  $D = NL$  is the column (film) thickness.

The optical response of the entire film is calculated by averaging the optical contributions of all possible column configurations, with proper weights. Since no spacial correlations are considered, and since  $L \ll \lambda$ , it is possible to average intensities rather than amplitudes (similar to the 2D scaling model). This simplification should be valid as long as non-specular scattering is negligible. The optical reflectance is thus given by

$$R = \iint \dots \int dx_1 dx_2 \dots dx_N P(x_1)P(x_2) \dots P(x_N) |r(x_1, x_2, \dots, x_N)|^2 \quad (6)$$

with similar expressions for the transmittance  $T$  and the absorptance  $A$ . Here  $P(\sigma) \equiv P(\sigma, L/\xi)$  is the conductivity bimodal distribution function, and  $r(\sigma_1, \sigma_2, \dots, \sigma_N)$  is the reflected amplitude of a column of  $N$  cubes with complex conductivities  $\sigma_1, \sigma_2, \dots, \sigma_N$  respectively.

For a semi-infinite film, there is an infinite number of possible configurations. Even for finite thickness, the complexity of this procedure rapidly increases with increasing thickness. However, this restriction is practically unimportant since only a few layers close to the film surface are significant, while the contribution of deeper layers may be well approximated

by an effective medium approximation (EMA). This is especially true in the case of metal-insulator mixtures, where both reflectance and absorptance are non-negligible.

The optical reflectance, transmittance and absorptance of a 3D percolative Drude metal film are shown in figure 1, as calculated by the above method, for  $p = p_c$ ,  $d = nL(\omega)$  with  $n = 1, 2, 3, 4, 5$ , and  $\lambda = 10, 40, 70, 100 \mu\text{m}$ . The dielectric AC conductivity is  $\sigma_1 = i\epsilon_1\omega c_0 = i0.01\omega$ ,  $c_0$  being the intercluster capacitance per unit length, and all other parameters are similar to those of [2]:  $A_1 = A_2 = 1$ ,  $A_3 = A_4 = 0.75$ ,  $\xi_0 = 100 \text{ \AA}$  and  $L_0 = 4$ . For comparison, an effective medium type calculation is also shown. The term 'effective medium approximation' (EMA) stands here for *any* model that replaces the *inhomogeneous media* by an *effective homogeneous one, taking into account the geometry* and the dielectric constants of the constituents. We also assume that the effective dielectric constant does not depend on the film thickness. For example, in the Bruggemann EMA [9], the effective dielectric constant depends on the filling factor  $p$  and the depolarization factor  $L$ , which should reflect the actual shape of the grains ( $L = \frac{1}{3}$  for spheres). Since any EMA calculates the effective dielectric constant, or refractive index  $\tilde{n}_{\text{eff}} = n + ik$ , where  $n, k$  do not depend on the film thickness  $d$ , we have selected  $\tilde{n}_{\text{eff}}(\lambda)$  values so that the 10 and 40  $\mu\text{m}$  data intersect the scaling results at  $d = 2L(\omega)$ , and the 70 and 100  $\mu\text{m}$  data at  $d = 3L(\omega)$ . Therefore, our comparison with the EMA does not depend on any particular model for the effective dielectric constant. The optical transmittance in figure 1 rapidly decreases with increasing thickness, as expected for a homogeneous film. Indeed the scaling and the effective medium results are almost identical. In contrast, the optical reflectance and absorptance show pronounced discrepancies, where the EMA results have stronger thickness dependence. In particular, the EMA absorptance has much steeper curvatures with respect to the film thickness.

Increasing the dielectric AC conductivity  $\sigma_1 = i0.1\omega$  yields similar results in general (figure 2), with the exception of the 10  $\mu\text{m}$  data: the optical reflectance is maximal at  $d = 4L(\omega)$ , and the absorptance has a shallow minimum. These are absent from the EMA curves if  $n < k$  (metallic-like). Selecting a dielectric-like effective dielectric constant,  $n > k$ , the above qualitative behaviour is recovered (figure 3). The reflectance is maximal at  $d = \lambda/4n$  due to constructive interference, and the absorptance is minimal at this thickness. Quantitatively, however, the EMA results are different from the scaling ones, where the interference effect in the latter case is much weaker. Indeed a distribution of local dielectric constants tend to wash out any interference effects. The existence, or absence, of interference fringes is thus a key feature in determining the physical significance of an effective dielectric constant.

In the above examples, the optical transmittance is fairly well described by the EMA, while the absorptance shows the largest discrepancies (note that the scaling 10  $\mu\text{m}$   $T$  in figures 2 and 3 is between the metallic-like and the dielectric-like EMA curves, but very close to both of them). It is interesting to understand why the scaling approach is more important for  $A$  than for  $T$ . In a homogeneous medium  $T$  rapidly decreases with increasing thickness due to the exponential term  $e^{-d/\delta}$  where  $\delta$  is the skin depth. The same trend is also expected in the scaling approach: the light is either absorbed or reflected when travelling through 'highly absorbing' or 'highly reflecting' regions, respectively, and the probability of a 'higher transparent' column rapidly decreases with increasing thickness. In contrast, the optical absorptance of an homogeneous film does not follow a monotonic thickness dependence: a very thin film is transparent and non-absorbing, while a thick film has high reflectance and low absorptance. At intermediate thicknesses both the reflectance and the transmittance are low, yielding large optical absorptance. For a Drude metal at low frequencies (mid- and far infrared) the absorptance reaches 50% in this regime [1]. The

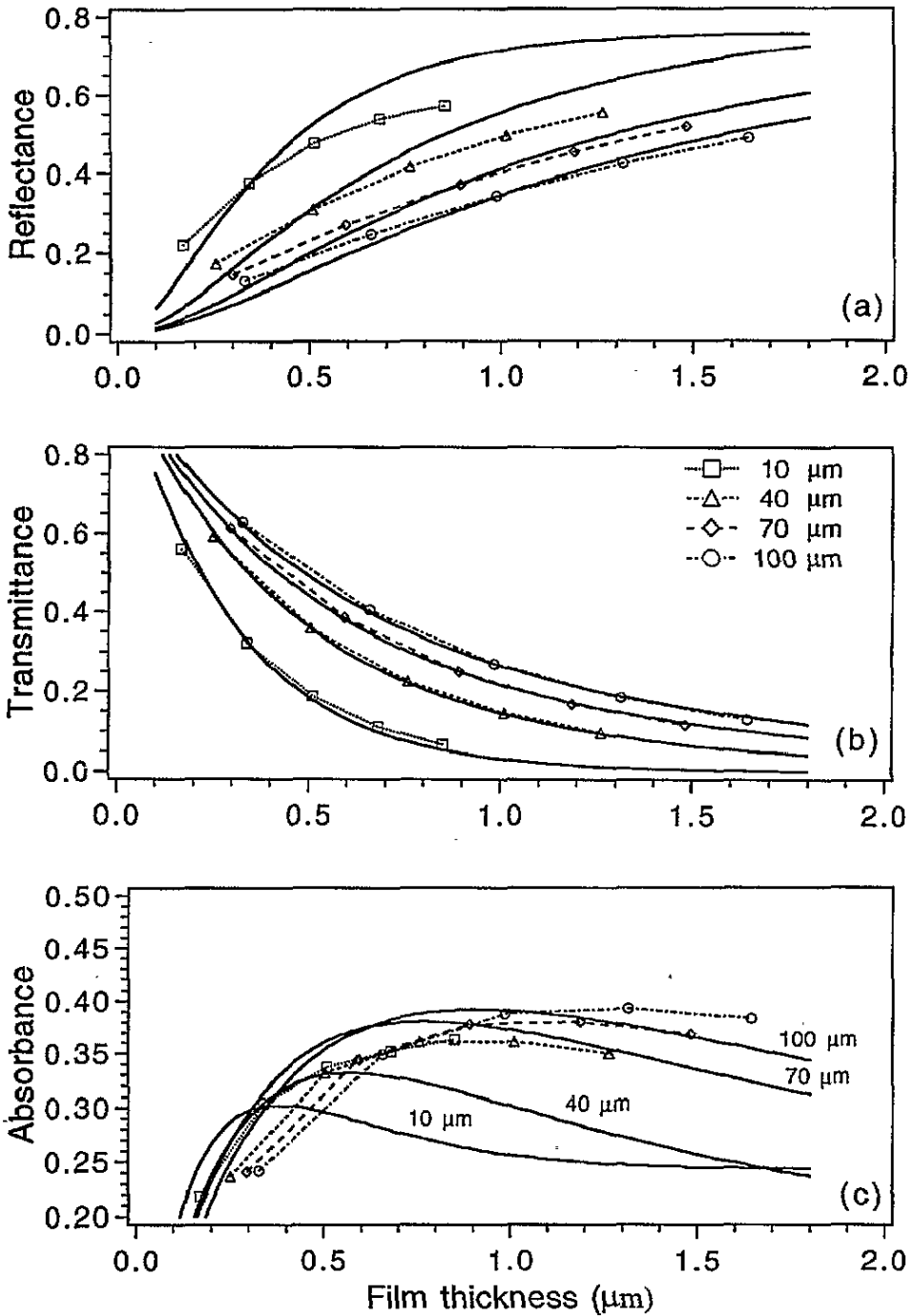


Figure 1. The reflectance, transmittance and absorbance of a metal-insulator film at  $p_c$  as calculated by the 3D scaling model and by an effective dielectric constant, versus the film thickness. The scaling model is applied for one to five layers at the wavelengths 10, 40, 70, and 100  $\mu\text{m}$ . The complex effective refractive indices are  $0.66 + i2.84$ ,  $1.22 + i4.54$ ,  $1.76 + i5.08$  and  $2.04 + i5.54$ , respectively.

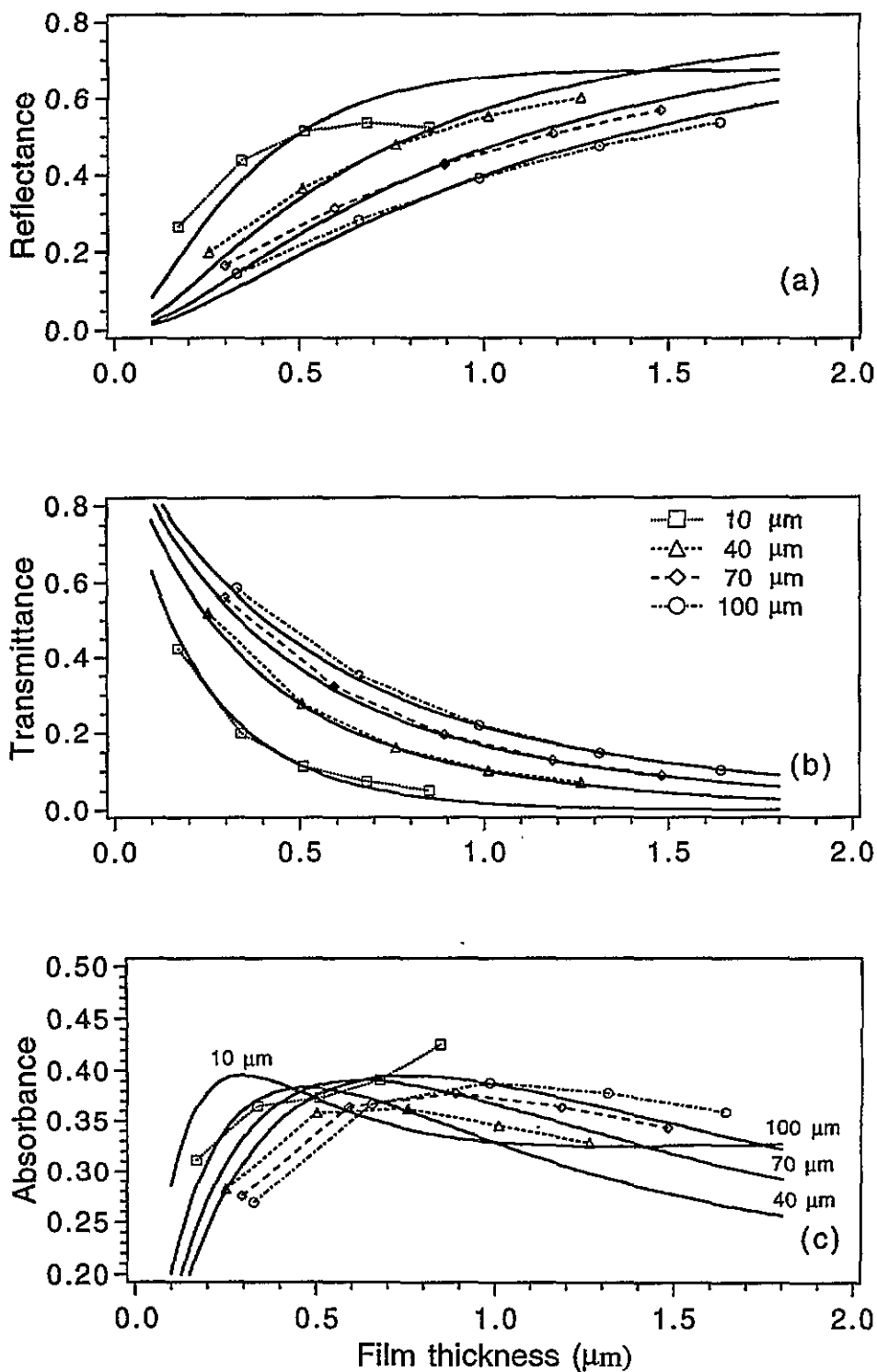


Figure 2. As figure 1 for  $\sigma_i = i0.1\omega$ . The effective refractive indices are  $1.16 + i3.08$ ,  $1.72 + i4.88$ ,  $2.04 + i5.58$  and  $2.26 + i6.04$  for  $\lambda = 10, 40, 70$  and  $100 \mu\text{m}$  respectively.

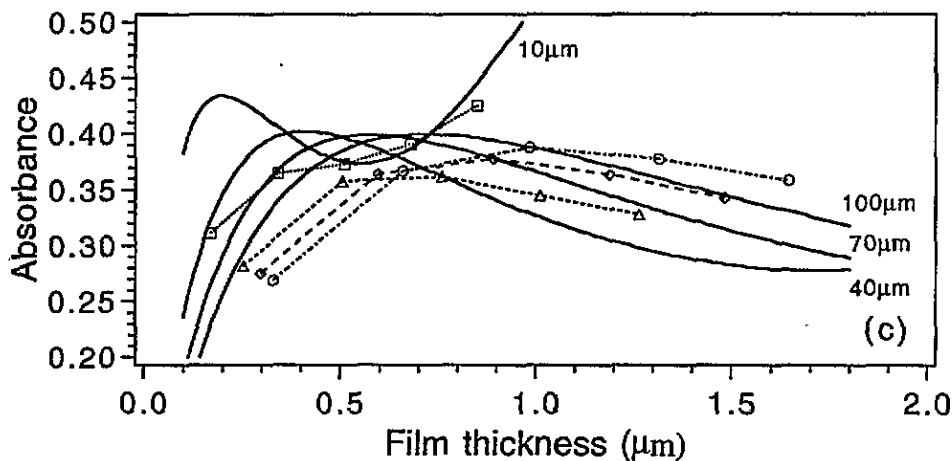
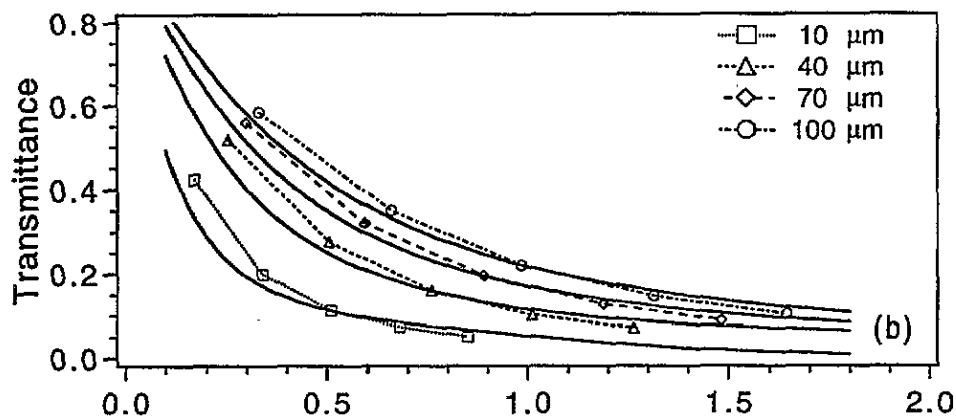
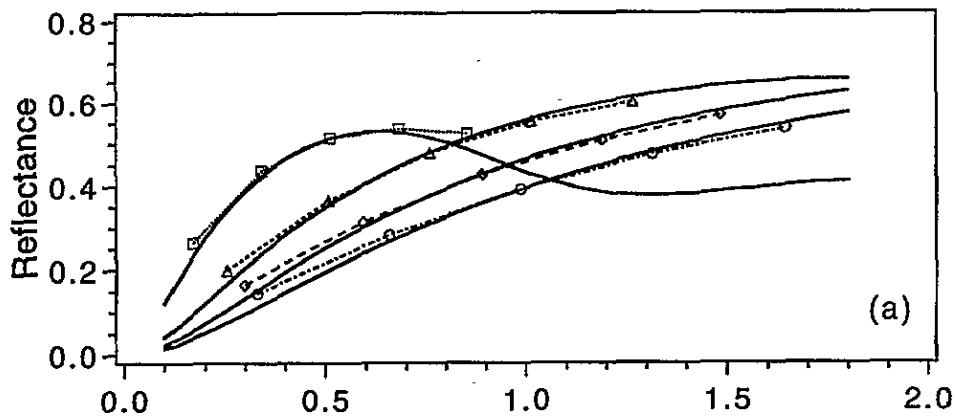
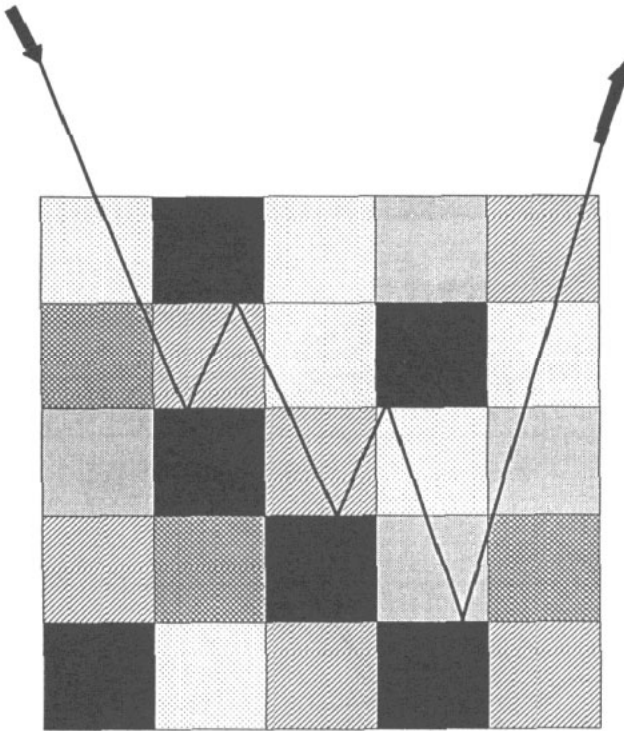


Figure 3. As figure 2, where dielectric-like effective refractive indices are selected:  $3.72 + i1.68$ ,  $5.24 + i2.0$ ,  $5.84 + i2.2$  and  $6.24 + i2.36$  for  $\lambda = 10, 40, 70$  and  $100\ \mu\text{m}$  respectively.



enhance absorptance in this case is due to multiple reflectance inside the film, hence its strong thickness dependence. Since coherent effects are suppressed in the scaling approach, the absorptance is also expected to be modified.

For oblique incidence the 3D scaling model becomes much more complicated since the transfer matrix method cannot be used: the incident and the reflected beams have different paths. Therefore, one should explicitly calculate the path of each multiply reflected beam, and average over all possible configurations for each path. An illustration of a possible path of an s polarized wave is shown in figure 4, where the light beam is partially trapped (localized) between randomly dispersed highly reflecting regions. Note that such a path is not possible at normal incidence, since the beam travels along *the same* path after each internal reflection. It is thus expected that the absorptance of an oblique s polarized wave should be enhanced in comparison with the normal incidence absorptance.



**Figure 4.** A schematic drawing of cubes of linear size  $L(\omega)$ , and a possible path of an s polarized oblique beam, illustrating partial light localization.

In conclusion, we have generalized the scaling model for the optical properties of semicontinuous metal films to thick films, and demonstrated some typical results of this model. It is concluded that the transmittance of thick percolative films may be reasonably described by an effective dielectric constant, in contrast with the optical absorptance and reflectance. Experimentally, the 3D scaling approach could be confirmed by the following observations: (i) the effective dielectric constant is thickness dependent; (ii) if the film is not too thick, its effective dielectric constant depends on the substrate as well as on its

thickness [7] (i.e. the same film on different substrates yields different  $\epsilon_{\text{eff}}$ ); (iii) interference effects are very weak, showing the intrinsic inhomogeneity and (iv) the optical absorptance of an oblique s polarized wave is higher than the normal incidence absorption. Finally, the optical response may be fitted by this model, where the weak thickness dependence allows interpolation for arbitrary film thicknesses.

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