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Experimental validation on granular metallic films of an optical model based on the entropic analysis of their morphology

C. Andraud[a,*], J. Lafait[a], A. Beghdadi[b], J. Peiro[a]

[a]*Laboratoire d'Optique des Solides de l'Université P. Et M. Curie, Unité associée au CNRS D0781, Case 80-4, Place Jussieu, 75252 Paris Cedex 05, France*

[b]*L.P.M.T.M. Institut Galilée, Université Paris Nord, 93430 Villetaneuse, France*

Abstract

We have developed two models for the optical properties of heterogeneous media, based on the entropic analysis of their morphology. These models have been applied to actual granular metallic media close to the percolation. The models account well for the measured optical properties: the plasmon resonance and the infrared behavior are especially well accounted.

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1. Introduction

As defined by Shannon and Brillouin [1, 2], the information quantity is a negative entropy, the negentropy, characteristic of the disorder. We have developed this concept for the quantification of the disorder in binary images of heterogeneous media [3, 4]. It allows a new analysis of the disordered morphology less ambiguous than the fractal or multifractal analysis. This entropic analysis points out a length, the optimum length, characteristic of the disorder of the image, at which we choose to calculate the local optical properties. We have developed two optical models, following the approach proposed by Hilfer for porous media [5], based on the partition of the medium at the size of the optimum length. The first model uses a self-consistent calculation of the dielectric function, while the second makes a local calculation of the optical properties. Then, we have compared the prediction of our two models to the result of classical effective medium theories (Bruggeman, Maxwell Garnett) and to the optical measurements on gold granular thin films near the percolation threshold, obtained by thermal evaporation.

*Corresponding author. Tel.: 33 1 44 27 43 53; fax: 33 1 44 27 39 82; e-mail: andraud@moka.ccr.jussieu.fr.

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2. The configuration entropy

We briefly recall our definition of the configuration entropy. The entropic analysis is realized by exploring a binary image composed of black and white pixels with a sliding window of variable size l, and calculating in this window the discrete probability distribution of filling in black pixels $\{p_k\}$ (k being the number of black pixels). The entropy of the image is defined as the sum of all p_k weighted by their own information quantities:

$$H(l) = -\sum_{k=0}^{l^2} p_k(l) \ln(p_k(l)). \tag{1}$$

To compare the different entropies calculated for different size of analysis, one must [6] normalize this quantity with the theoretical maximum entropy. This theoretical maximum entropy is determined by using the Lagrange multiplier theory, which determines the probability distribution maximizing the entropy. This distribution is the equally probable distribution, and the corresponding entropy is equal, in the case of a window of size l to

$$H_{\max}^{\mathrm{Th}}(l) = \ln(l^2 + 1). \tag{2}$$

We thus define the normalized configuration entropy as

$$H^*(l) = \frac{-\sum_{k=0}^{l^2} p_k(l) \ln(p_k(l))}{\ln(l^2 + 1)}. \tag{3}$$

By varying the size of analysis one observes that the normalized configuration entropy presents a maximum for a length that we call the optimum length. This length depends on the black pixel fraction in the image. It presents a minimum around the percolation threshold.

The optimum length of entropy is characteristic of the disorder in the image. At this size, one can observe the maximum number of different configurations without any redundancy or lack of information, their distribution being closest to the flat histogram. Close to the percolation, this length is the size of the smallest element which, if added, make the medium pass from the nonpercolating to the percolating state. Under the point of view of optics, we consider that, it is the combination of all the local responses of blocks of this size in the image which will *finely* determine the global optical response of the medium.

3. Optical models

The optical models we developed are based on the model proposed by Hilfer for porous media [5, 7]. We are interested in giving an account of the optical properties of heterogeneous media, composed of metallic inclusions (black pixels), embedded in a dielectric matrix (white pixels). By contrast with the effective medium theories, which

make a partition between metallic and dielectric components, this model makes a partition of the whole medium at the size of the entropic optimum length and separates cells in percolated and nonpercolated classes.

In both classes, we calculate the effective dielectric function with the Maxwell Garnett model [8] modified by Cohen et al. [9] by determining its percolating state. One thus gets a set of effective dielectric functions, for all the percolated and non-percolated blocks.

In order to distinguish between two types of possible electromagnetic propagation in a heterogeneous medium, we then developed two different models on this basis. In the first model, we make a self consistent calculation of the effective dielectric function of the whole image by using a Bruggeman model [10] applied to n components, corresponding to the n blocks in the whole image. We then calculate the optical properties of the medium, by using Abelès formulae for thin films [11]. In this first model, the application of the effective medium treatment up to the size of the whole medium is equivalent to a coherent treatment, with the same value of the phases for all the local electromagnetic fields. In the second model, we make a local calculation of the optical properties (reflectance and transmittance), in each cell by using the local dielectric function and then applying Abelès formulae. A simple summation on all cells, directly gives the optical properties of the whole medium. In this second model, we explore the effect of an incoherent treatment directly applied from the size of the optimum length.

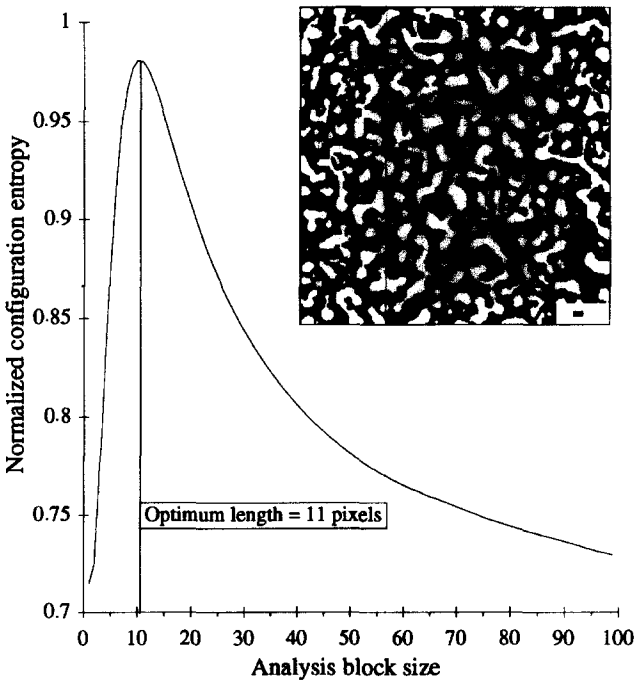

Fig. 1. Image of granular gold film and its associated normalized entropy.

4. Application to real materials

We show now the application of the normalized configuration entropy, and of the two optical models, presented above, to a real image of a thin discontinuous gold film close to the percolation. The deposition of the film is realized by thermal evaporation under ultra high vacuum. The mass thickness is equal to $d = 61$ Å. The image was binarized, giving a black pixel concentration $p = 32\%$. From these values, we deduce the "real" thickness of the film, $d/p = 190$ Å. The entropic analysis gives an optimum length equal to 18 pixels, corresponding to 100 nm on the sample (white mark) (Fig. 1).

We compare the predictions of our models to the predictions of classical E.M.T. (Bruggeman, Maxwell Garnett) and to the optical measurements on the discontinuous film. The image and the experimental optical properties both point out the fact that the sample is slightly below the percolation threshold. The decrease of the reflectance R and the increase of the transmittance T with wavelength are indeed characteristic of a dielectric behavior of the whole medium (Fig. 2(a) and (b)).

The Maxwell Garnett theory gives a relatively good account of the position and amplitude of the metallic grain resonance observed around 600 nm, but does not predict correctly the optical properties in the near infrared. On the contrary, the Bruggeman model accounts well for the optical properties in the near infrared, but

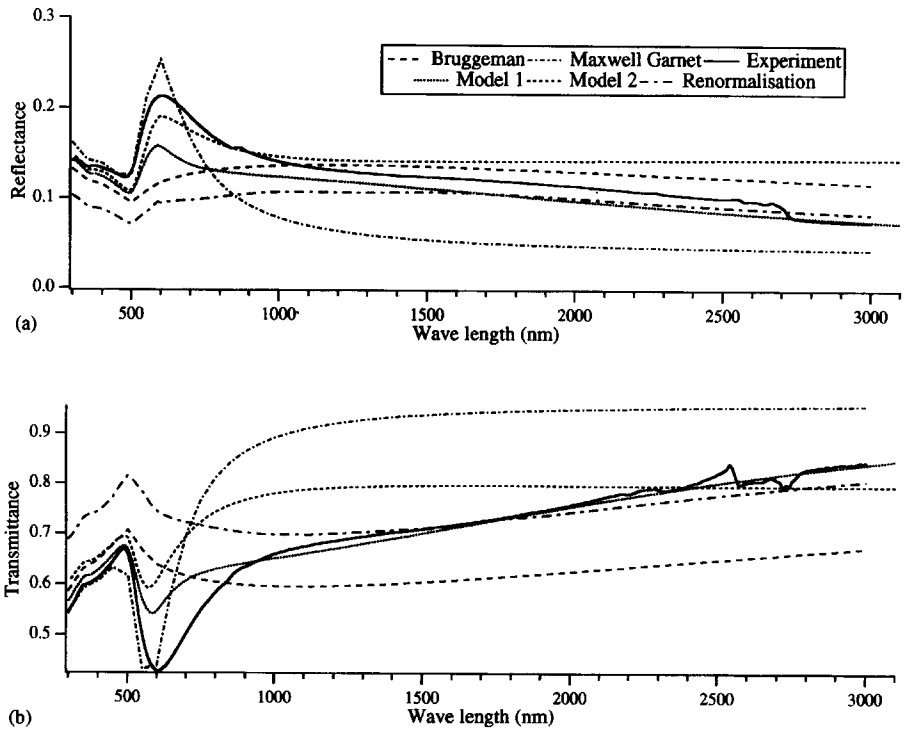

Fig. 2. Reflectance (a) and transmittance (b) of the granular gold film.

never reproduces the grain resonance. This result confirms that the Bruggeman model predicts a too broad and dumped grain resonance. Our first model accounts well for the properties in the near infrared, and gives the best prediction for the grain resonance, although its amplitude is weaker and its width is narrower than the experimental resonance. This result confirms that this model takes into account the geometry of the medium better, the average concentration and the concentration distribution, which notably govern the resonance. Our second model is not so efficient as the first model in the near infrared, but it is somewhat better for the reflectance near the resonance. Its relative failure as compared to the first model should come from the noncoherent calculation performed from a too small size.

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