EFFECTIVE MEDIUM THEORY: MATHEMATICAL DETERMINATION OF THE PHYSICAL SOLUTION FOR THE DIELECTRIC CONSTANT

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The complex effective dielectric constant of an inhomogeneous medium, as calculated with the Effective Medium Theory (E.M.T.), is the solution of a quadratic equation and is therefore a multiple-valued function. In most cases, physical criteria alone are not sufficient to select the solution which has a physical meaning. By using the properties of complex functions and Riemann surfaces together with physical criteria, one can readily define a single solution with physical meaning. The method is applied to a cermet-like material with metallic Cr inclusions.

1. Introduction

During the last few years growing attention has been paid to the optical properties of inhomogeneous media, especially as selective coatings for photothermal conversion of solar energy [1,2]. Among the numerous theories allowing to calculate the effective dielectric constant of an inhomogeneous medium, the Effective Medium Theory (E.M.T.) proposed by Bruggeman [3] can be successfully used, especially for media with relatively high metallic inclusion density. The purpose of this paper is to clarify a mathematical problem raised by that theory which up to now has only been solved in some particular cases with the help of physical criteria.

In part 2, we emphasize that the complex effective dielectric constant of an inhomogeneous medium determined with E.M.T. is a multiple-valued function. Except in some particular cases, depending on the relative values of the dielectric constant of the matrix (ϵ_m) and the inclusion (ϵ_i) and of the filling factor (q), physical criteria are not sufficient to select the solution which has a physical meaning. We show in part 3 that, by using the properties of complex functions and Riemann surfaces, it is possible to obtain mathematical criteria leading, when coupled with physical ones, to a single solution with physical meaning in all cases. The method is eventually applied to a cermet-like material with metallic Cr inclusions, somewhat similar to black chromium coatings.

2. Effective medium theory

The theory can apply to a medium with n components, but, in order to simplify the formalism, we shall give the demonstration for two components only. It assumes that both components are in the form of homogeneous spheres or ellipsoids, the characteristic dimensions of which are small compared to the wavelength of the incident radiation. Both types of spheres or ellipsoids are embedded in an effective medium of complex dielectric constant ϵ_{av}, where the average influencing field $\boldsymbol{E}_{av}$ is defined as the spatial average of fields $\boldsymbol{E}_1$ and $\boldsymbol{E}_2$ inside the components:

$$\boldsymbol{E}_{av} = q\boldsymbol{E}_1 + (1-q)\boldsymbol{E}_2, \tag{1}$$

with

$$\boldsymbol{E}_{1(2)} = \frac{\epsilon_{av}}{g\epsilon_{1(2)} + (1-g)\epsilon_{av}} \boldsymbol{E}_{av}. \tag{2}$$

Eq. (2) results from the computation of the polarizability of an ellipsoid, with depolarization factor g along the applied field ($g = 1/3$ if the inclusion is spherical); q is the volumic filling factor of component 1.

If now ϵ_i and ϵ_m are the complex dielectric con-

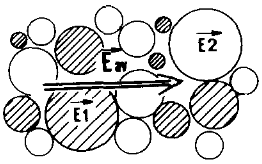

Fig. 1. Effective medium model.

stants of component 1 (inclusion) and component 2 (matrix) respectively, it follows from (1) and (2):

$$(1-g)\epsilon_{av}^2 + \epsilon_{av}[\epsilon_i(g-q) + \epsilon_m(g+q-1)] - g\epsilon_i\epsilon_m = 0. \quad (3)$$

Eq. (3) is quadratic versus ϵ_{av} and its solution is a multiple-valued function. If we put:

$$P = [-\epsilon_i(g-q) - \epsilon_m(g+q-1)]/2(1-g),$$

$$Q = P^2 + g\epsilon_i\epsilon_m/(1-g) = \beta_1 + i\beta_2, \quad (4)$$

the problem of the computation of ϵ_{av} is reduced to the computation of the complex square root of Q.

The complex multiple-valued function ϵ_{av} is defined in the complex area **C**. The variables are g, q, ϵ_i (complex) and ϵ_m (assumed to be real: non absorbing matrix). Physical considerations on these variables lead to restrict the domain of definition of ϵ_{av}

$$0 \leqslant q \leqslant 1; \qquad \mathrm{Im}\,(\epsilon_i) \geqslant 0,$$

$$0 \leqslant g \leqslant 1; \qquad \epsilon_m \text{ real} \geqslant 0. \quad (5)$$

Furthermore the function ϵ_{av} itself has to fulfil other conditions:

$$\mathrm{Im}\,(\epsilon_{av}) \geqslant 0, \qquad \epsilon_{av}(q=0) = \epsilon_m, \qquad \epsilon_{av}(q=1) = \epsilon_i. \quad (6)$$

The last two conditions correspond to the limits of homogeneous media consisting in the matrix material and the inclusion material.

On the other hand, in order to have a physical meaning, ϵ_{av} must be holomorphous. Now a multiple-valued function is not holomorphous. This problem of choosing a determination for ϵ_{av} can be solved by using the concept of Riemann surfaces.

3. Riemann surfaces

We consider the function complex square root of z, where z belongs to **C**. There are two distinct solutions to $w^2 = z$ (for $z \neq 0$):

$$w_1 = |z|^{1/2}\, e^{i\theta/2}, \qquad w_2 = |z|^{1/2}\, e^{i\theta/2 + i\pi} = -w_1,$$

with

$$\theta = \arg z, \qquad 0 \leqslant \theta \leqslant 2\pi.$$

Let S_1 and S_2 be two extended complex planes defined by:

$$S_1: \qquad 0 \leqslant \arg z < 2\pi,$$

$$S_2: \qquad 2\pi \leqslant \arg z < 4\pi.$$

We now imagine that S_1 and S_2 join together: the upper side of the positive real axis in S_1 is brought in coincidence with the lower side of the positive real axis in S_2 and reciprocally the lower side of S_1 positive real axis is brought in coincidence with the upper side of S_2 positive real axis. The resulting surface can be visualized by considering a continuous curve, belonging to that surface, winding twice around the origin (fig. 2).

This open connected set of complex planes attached to each other is known as a Riemann surface [4]. On that Riemann surface acting as a new domain of definition for w, the function w is holomorphous (i.e. continuous and differentiable): one can pass continuously from w_1 defined on S_1 to w_2 defined on S_2.

If we now go back to our mathematical problem summarized by eqs. (3) and (4), one can easily find

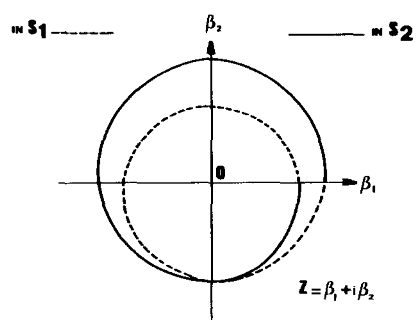

Fig. 2. A continuous curve on the suggested Riemann surface. $O\beta_1$, $O\beta_2$ define the classical complex plane **C**.

four sets of functions Σ_1, Σ_2 such as $\epsilon_{av} = \Sigma_1 + i\Sigma_2$ verifying eq. (3):

$$\Sigma_1 = P_1 + (\beta_2/|\beta_2|) Q_1, \qquad \Sigma_2 = P_2 + Q_2, \tag{7}$$

$$\Sigma_1 = P_1 + Q_1, \qquad \Sigma_2 = P_2 + (\beta_2/|\beta_2|) Q_2, \tag{8}$$

$$\Sigma_1 = P_1 - (\beta_2/|\beta_2|) Q_1, \qquad \Sigma_2 = P_2 - Q_2, \tag{9}$$

$$\Sigma_1 = P_1 - Q_1, \qquad \Sigma_2 = P_2 - (\beta_2/|\beta_2|) Q_2, \tag{10}$$

with

$$P_1 = -[\epsilon_1(g-q) + \epsilon_m(g-q-1)]/2(1-g),$$

$$P_2 = -\epsilon_2(g-q)/2(1-g), \tag{11}$$

$$(Q_1 + iQ_2)^2 = Q = \beta_1 + i\beta_2,$$

$$Q_1 = [\beta_1 + (\beta_1^2 + \beta_2^2)^{1/2}]^{1/2}/\sqrt{2},$$

$$Q_2 = [-\beta_1 + (\beta_1^2 + \beta_2^2)^{1/2}]^{1/2}/\sqrt{2}, \tag{12}$$

$$\beta_1 = \{1/4(1-g)^2\}\{(\epsilon_1^2 - \epsilon_2^2)(g-q)^2$$
$$+ \epsilon_m^2(g+q-1)^2 + 2\epsilon_1\epsilon_m[q(1-q) + g(1-g)]\}$$

$$\beta_2 = \{1/4(1-g)^2\}\{2\epsilon_1\epsilon_2(g-q)^2$$
$$+ 2\epsilon_m\epsilon_2[q(1-q) + g(1-g)]\} \tag{13}$$

$$\epsilon_i = \epsilon_1 + i\epsilon_2, \qquad \epsilon_m \text{ real}, \tag{14}$$

The new variables β_1 and β_2 (functions of ϵ_1, ϵ_2, ϵ_m, g, q) are the coordinates of a point in the complex plane **C** in which the functions (7) to (10) are defined.

None of these four functions is holomorphous on **C**, because of the discontinuity occurring at the branch points located at $\beta_2 = 0$. But if one considers a Riemann surface composed of two sheets S_1 and S_2 connected along the positive part of the real axis ($\beta_2 = 0$) as indicated previously, one can define two determinations (i.e. holomorphous functions) on this new domain of definition:

$$\beta_1 < 0: \quad \Sigma_1 = P_1 + (\beta_2/|\beta_2|)Q_1, \quad \Sigma_2 = P_2 + Q_2,$$

$$\beta_1 > 0: \quad \Sigma_1 = P_1 + Q_1, \quad \Sigma_2 = P_2 + (\beta_2/|\beta_2|)Q_2, \tag{15}$$

$$\beta_1 < 0: \quad \Sigma_1 = P_1 - (\beta_2/|\beta_2|)Q_1, \quad \Sigma_2 = P_2 - Q_2,$$

$$\beta_1 > 0: \quad \Sigma_1 = P_1 - Q_1, \quad \Sigma_2 = P_2 - (\beta_2/|\beta_2|)Q_2. \tag{16}$$

Now, the question is: which one of the two determinations (15) and (16) has a physical meaning.

4. Choice of the physical solution

Taking into account conditions (5) on q, g, ϵ_2, ϵ_m the variations of β_1 and β_2 are limited, i.e. the domain of definition of the two determinations is now restricted to part of the Riemann surface. On this restricted domain, the physical determination has to fulfil conditions (6).

It is easy to verify that determination (16) does not satisfy the condition $\mathrm{Im}(\epsilon_{av}) \geqslant 0$ for $\beta_1 < 0$, i.e. in the domain of definition. Thus, determination (16) cannot be the physical solution. On the contrary, it can be verified that determination (15) fulfils the three conditions (6) for $\beta_1 < 0$ and for $\beta_1 > 0$, $\beta_2 > 0$, but none of them for $\beta_1 > 0$, $\beta_2 < 0$. A short calculation shows that the part $\beta_1 > 0$, $\beta_2 < 0$ of the domain of definition is in fact out of the limits defined by (5). Therefore, one can say that determination (15) is the physical solution to equation (3). Considering the last restriction ($\beta_1 > 0$, $\beta_2 < 0$ excluded) of its domain of definition, one can write this determination under a single form:

$$\epsilon_{av} = \epsilon_{1av} + \epsilon_{2av},$$

$$\epsilon_{1av} = P_1 + (\beta_2/|\beta_2|)Q_1, \qquad \epsilon_{2av} = P_2 + Q_2, \tag{17}$$

β_2, P_1, Q_1, P_2, Q_2 being defined in (11), (12), (13). This is the only physical solution for the complex effective dielectric constant of an inhomogeneous medium (with non absorbing matrix) obtained with the E.M.T. theory.

5. Practical use and application

We shall now examine a case of practical use: $g = 1/3$ (spherical particles). The branch points corresponding to a change in the sign of β_2 exist only if the equation $\beta_2 = 0$ (cf. eqs. (13)) has roots, i.e. if:

$$9q^2(\epsilon_i - \epsilon_m) + 3q(3\epsilon_m - 2\epsilon_1) + \epsilon_1 + 2\epsilon_m = 0$$

has real solutions; this occurs when: $\epsilon_1 \leqslant 17\epsilon_m/16$. These roots are:

$$q_{1(2)} = \{3(2\epsilon_1 - 3\epsilon_m)$$

$$(\pm)\ [\epsilon_m(17\epsilon_m - 16\epsilon_1)]^{1/2}\}/18(\epsilon_1 - \epsilon_m).$$

For $\epsilon_1 < 17\epsilon_m/16$, β_2 is always negative, there is no branch point; for $\epsilon_1 \geqslant 17\epsilon_m/16$ there are two branch points for $q = q_1$ and $q = q_2$. None, one or two of these points can be located in the domain $0 \leqslant q \leqslant 1$.

Two of these cases are illustrated on figs. 3a and 3b where the real part of the effective dielectric constant ϵ_{1av} of a cermet with metallic Cr inclusions in an oxide matrix ($\epsilon_m = 2.3$) has been plotted versus q at two wavelengths. The full lines correspond to the determination defined in eq. (17) (the correct one) and the dashed lines to the one defined in eq. (16). At $\lambda = 0.47\ \mu$m ($\epsilon_1 = 0.84$, $\epsilon_2 = 8.8$) (fig. 3a) the two curves are well separated, the branch points are located out of the domain of variation of q, it is therefore easy to choose the physical solution with the help of physical criteria (at $q = 0$ and $q = 1$) alone. At $\lambda = 3\ \mu$m (fig. 3b) ($\epsilon_1 = -27.4$, $\epsilon_2 = 60.8$) the curves are crossing, there are two branch points in the domain $0 \leqslant q \leqslant 1$. The full lines only correspond to the physical solution of Bruggeman's equation (3) and give the correct effective dielectric constant of the inhomogeneous medium under consideration. If one of the solutions (8) to (10) has been used, this would have led to discontinuities at the branch points.

The problem of the discontinuity which may be found for the average dielectric constant if calculated with functions (8) to (10) has not been pointed out by many authors. The optical properties of most of the cermets to which the Bruggeman's theory was applied essentially covered the visible or, at least, the very near infrared range. Considering the curves of fig. 3a, and extending these results to cermets of other materials, one can roughly say that, in most cases, there are no crossing points in the visible range and therefore the problem was not raised. The advantage of our treatment is to give the correct expression valid at all wavelengths directly.

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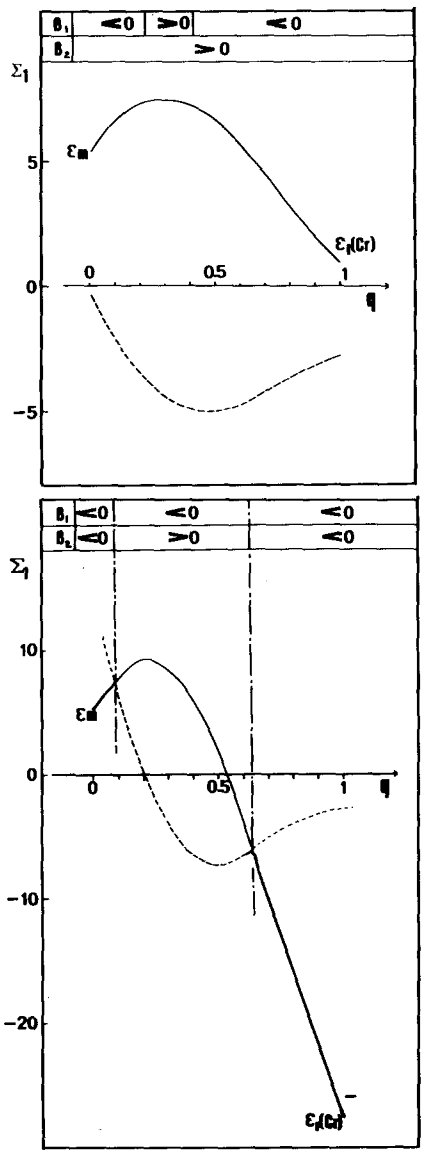

Fig. 3. Real part of the average dielectric constant of a cermet ($\epsilon_m = 2.3$) with metallic Cr inclusions; versus filling factor in Cr. The two curves correspond to the two determinations. The full line corresponds to the physical solution of Bruggeman's equation. (a): $\lambda = 0.47\ \mu$m; ϵ_i(Cr) = 0.84 + i8.8. (b): $\lambda = 3\ \mu$m; ϵ_i(Cr) = −27.4 + i60.8.

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