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# Bergman spectral representation of a simple expression for the dielectric response of a symmetric two-component composite

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**Abstract.** A simple expression for the effective dielectric constant of a symmetric two-component composite (aggregate topology) is presented and interpreted in terms of the Bergman spectral representation.

## 1. Introduction

The determination of effective material constants of composites has been a problem of essential technical and scientific interest for a long time. Outstanding contributions have been provided e.g. by Maxwell-Garnett [1], by Bruggeman [2], by Wiener [3], by Hashin and Shtrikman [4] and by Bergman [5]. To date, however, there still do not exist generally accepted mixing formulae, i.e. expressions e.g. for the dielectric constants of composites as functions of those of their components and their concentrations, that are able to describe reasonably well experimental results on certain not too special classes of composites.

In [6] the author proposed such expressions for different typical microstructures on the basis of simple interpolations between exactly known limits and applied them to the calculation of frequency dependent properties [7].

It was not proved, however, that these expressions obey the analytic properties and general requirements demanded by the Bergman spectral representation [5].

In this paper we concentrate on the sufficiently general case of the dielectric constant of a symmetric two-component composite. The expression obtained in [6] is first rederived. It is then proved that it has a Bergman spectral representation, the spectral function of which satisfies all known requirements. It is shown further that the easiest form of an interpolation of the spectral function between its exactly known limits at very low and very high concentrations results in the same expression as derived in [6]. In addition, possible generalizations and consequences for the DC conductivity and for percolating systems are discussed.

## 2. Dielectric constant of a symmetric two-component composite

As in [5] we define the effective dielectric constant of an inhomogeneous material by

$$\langle D \rangle = \varepsilon \langle E \rangle \quad (1)$$

where  $\langle \dots \rangle$  denotes the volume average of a given quantity.  $\langle E \rangle$  and  $\langle D \rangle$  are given by

$$\begin{aligned}\langle E \rangle &= (1 - c)\langle E_1 \rangle + c\langle E_2 \rangle \\ \langle D \rangle &= (1 - c)\varepsilon_1\langle E_1 \rangle + c\varepsilon_2\langle E_2 \rangle\end{aligned}\quad (2)$$

with  $\langle D_1 \rangle$ ,  $\langle E_2 \rangle$ ,  $\langle D_1 \rangle$  and  $\langle D_2 \rangle$  being the volume averages over the volumes  $V_1$  and  $V_2$  of the two types of constituent with dielectric constants  $\varepsilon_1$  and  $\varepsilon_2$ , and  $c = V_2/(V_1 + V_2)$ .

Hence, with  $\langle E \rangle \parallel \langle E_1 \rangle \parallel \langle E_2 \rangle$  we obtain

$$\varepsilon = \frac{(1 - c)\varepsilon_1 + c\varepsilon_2\langle E_2 \rangle/\langle E_1 \rangle}{(1 - c) + c\langle E_2 \rangle/\langle E_1 \rangle}\quad (3)$$

as an exact result. The mean field ratio  $\langle E_2 \rangle/\langle E_1 \rangle$  is a complicated and, in general, unknown expression, which depends on the special structure of a given sample.

This ratio can be calculated exactly in the limit of one single inclusion with the form of an ellipsoid, see, e.g. [8]. Representing the limits  $c \rightarrow 0$  and  $c \rightarrow 1$  of a symmetric composite by spherical inclusions of vanishing number and increasing distance we have

$$\langle E_2 \rangle/\langle E_1 \rangle = \frac{3\varepsilon_1}{2\varepsilon_1 + \varepsilon_2} \text{ for } c \rightarrow 0\quad (4)$$

and

$$\langle E_1 \rangle/\langle E_2 \rangle = \frac{3\varepsilon_2}{2\varepsilon_2 + \varepsilon_1} \text{ for } c \rightarrow 1.\quad (5)$$

The obvious generalizations to non-spherical inclusions with or without randomly distributed orientations are discussed, e.g. in [5], [6] and [9].

From (3)–(5) the effective dielectric constant  $\varepsilon$  together with its derivative  $\varepsilon' = d\varepsilon/dc$  are exactly known in the limits  $c = 0$  and  $c = 1$ :

$$\varepsilon|_{c=0} = \varepsilon_1 \quad \varepsilon|_{c=1} = \varepsilon_2\quad (6)$$

and

$$\begin{aligned}\varepsilon'|_{c=0} &= \frac{3\varepsilon_1}{2\varepsilon_1 + \varepsilon_2}(\varepsilon_2 - \varepsilon_1) \\ \varepsilon'|_{c=1} &= \frac{3\varepsilon_2}{2\varepsilon_2 + \varepsilon_1}(\varepsilon_2 - \varepsilon_1).\end{aligned}\quad (7)$$

One possible simple interpolation between these boundary values can be obtained from the (3, 2) Padé approximation

$$\varepsilon = \frac{\alpha + \beta c + \gamma c^2}{1 + \delta c}\quad (8)$$

where the constants  $\alpha$ ,  $\beta$ ,  $\gamma$  and  $\delta$  have to be chosen in such a way that (6) and (7) are fulfilled.

The result is

$$\varepsilon = \frac{\varepsilon_1 + 4c\varepsilon_1(\varepsilon_2 - \varepsilon_1)/(2\varepsilon_1 + \varepsilon_2) + 2c^2(\varepsilon_2 - \varepsilon_1)^2/(2\varepsilon_1 + \varepsilon_2)}{1 + c(\varepsilon_2 - \varepsilon_1)/(2\varepsilon_1 + \varepsilon_2)}\quad (9)$$

which is exactly the expression given in [6].

We remark that the equations (4)–(7) and, hence, (9) contain all the exact knowledge of first order in  $c$  or  $(1 - c)$ , respectively and no assumptions about higher-order terms. An exact knowledge of e.g. the next order  $c^2$  or  $(1 - c)^2$ , respectively, would require the performance of an infinite number of calculations for two inclusions at all possible distances. Therefore, any expression 'improved' compared to (9) for the effective dielectric constant  $\varepsilon$  is necessarily approximative or includes phenomenological parameters.

### 3. Bergman spectral representation of the dielectric constant

Introducing the variables  $f = 1 - \varepsilon/\varepsilon_1$  and  $s = (1 - \varepsilon_2/\varepsilon_1)^{-1}$  it was proved by Bergman [5] that for any given sample  $f(s)$  has the spectral representation

$$f(s) = \frac{A}{s} + \int_0^1 \frac{g(s') ds'}{s - s'} \quad (10)$$

with a positive spectral function  $g(s)$ , which obeys, in the case of isotropic or cubic rotational symmetry, the equations

$$\int_0^1 g(s) ds = c - A \quad (11)$$

$$\int_0^1 s g(s) ds = \frac{1}{3} c(1 - c) \quad (12)$$

and the inequality

$$\int_0^1 \frac{g(s) ds}{1 - s} \leq 1 - A \quad \text{with} \quad A \leq \frac{2c}{3 - c} \quad (13)$$

(see also [10]–[12]).

Equation (9) expressed in the variables  $f$  and  $s$  gives

$$f(s) = \frac{2c^2}{1 + c} \frac{1}{s} + \frac{c(1 - c)}{1 + c} \frac{1}{s - (1 + c)/3} \quad (14)$$

(14) is of the form (10) with

$$A = (2c^2)/(1 + c) \quad (15)$$

$$g(s) = \frac{c(1 - c)}{1 + c} \delta\left(s - \frac{1 + c}{3}\right) \quad (16)$$

The equations (11) and (12) and the inequalities (13) are also fulfilled by (14). Hence, (9) has a correct Bergman spectral representation.

### 4. Rederivation of the dielectric constant by interpolating the Bergman spectral function

It is possible to obtain (9) alternatively by a simple interpolation of the spectral function  $g(s)$  between its values in the limits  $c \rightarrow 0$  and  $c \rightarrow 1$ ; as follows.

The spectral function  $g(s)$  is closely related to the internal surface mode spectrum of the sample, see e.g. [5], [11] and [12]. This mode spectrum is determined by the AC

dielectric function  $\varepsilon(\omega)$ , which is given by the same expression as the dielectric constant  $\varepsilon$  if retardation effects are neglected.

For spherical inclusions in the limits  $c \rightarrow 0$  and  $c \rightarrow 1$  these modes are the Fröhlich modes at  $2\varepsilon_1(\omega) + \varepsilon_2(\omega) = 0$  corresponding to  $s = \frac{1}{3}$ , and at  $2\varepsilon_2(\omega) + \varepsilon_1(\omega) = 0$  corresponding to  $s = \frac{2}{3}$ , respectively. Hence,  $g(s) \sim c\delta(s - \frac{1}{3})$  and  $g(s) \sim (1 - c)\delta(s - \frac{2}{3})$  in these limits. The prefactors  $c$  and  $(1 - c)$  ensure that the mode strengths vanish for  $c \rightarrow 0$  and  $c \rightarrow 1$ , respectively.

The easiest interpolation between these limits without further information or assumptions is given by

$$g(s) \sim c(1 - c)\delta\left(s - \frac{1 + c}{3}\right). \quad (17)$$

From (12) we then get

$$g(s) = \frac{c(1 - c)}{1 + c}\delta\left(s - \frac{1 + c}{3}\right) \quad (18)$$

and from (11)

$$A = \frac{2c^2}{1 + c}. \quad (19)$$

The results (19) and (18) are identical to (15) and (16) obtained from expression (9), the Padé approximation used for its derivation being equivalent to a linear interpolation of the spectral function  $g(s)$  between the limits  $c \rightarrow 0$  and  $c \rightarrow 1$ .

## 5. Discussion

The dielectric constant calculated from (9) lies for all  $0 \leq c \leq 1$  between the limits obtained by Hashin and Shtrikman [4] for isotropic composites, see [6]. Its value is larger than those obtained from e.g. the Bruggeman [2], Looyenga [13] or Lichtenecker [14] theories and is in better agreement with results of numerical simulations [15], see figure 1. The remaining difference may be due to the fact that multipole fields are not taken into account by (9). This interpretation is supported by the fact that the results of [15] violate the upper Hashin-Shtrikman limit (based on the dipole approximation) for e.g.  $c = 0.3$  and  $\varepsilon_2 < 30$ .

The interpolation scheme presented here results in a  $\delta$ -like spectral function corresponding to only one surface mode inside a given sample for any  $c$ . In reality there occurs, of course, a spectrum of many modes corresponding to the special geometry and, hence, a continuous spectrum for infinite samples or after ensemble averaging. It would be easy to generalize the expression (18) to the case of a spectrum with finite half-width (terminated at  $s = 0$  and  $s = 1$ ) and to introduce some bending of its centre of gravity from the straight line between  $c = 0$  and  $c = 1$ . In this way the agreement with experimental results and/or with computer simulations could be improved. Such a procedure requires, however, the introduction of purely phenomenological parameters, which cannot be justified theoretically without drastic approximations and, possibly, harmful consequences. In the case of the Bruggeman theory [2], for example, the exact boundary conditions (7) are violated near the frequencies of the Fröhlich modes, see [7], and also [16] for the discussion of a related problem.

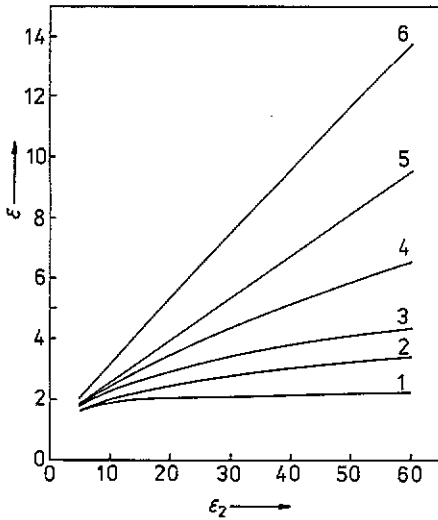


Figure 1. Comparison of the results of the Maxwell-Garnett (1), Lichtenecker (2), Bruggeman (3), Looyenga (4) and the present (5) expressions for the effective dielectric function compared with the computer simulation results of Stölzle *et al* (6) for  $c = 0.3$  and  $\epsilon_1 = 1$  as functions of the second component  $\epsilon_2$ . The data of Stölzle [15] are fitted by their analytical expression  $\epsilon^a = (1 - c)\epsilon_1^a + c\epsilon_2^a$  with  $a(c) = 1.6c + 0.265$

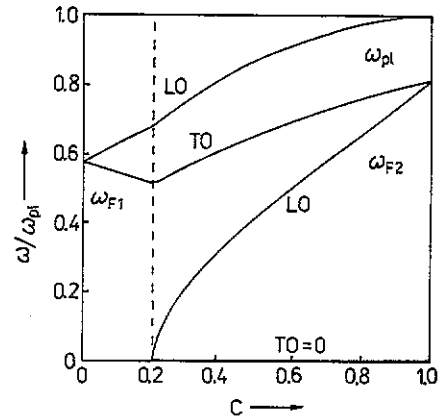


Figure 2. Frequency dependence of the TO and LO modes (maxima of  $\text{Im } \epsilon$  and  $\text{Im}(-\frac{1}{\epsilon})$  respectively) of a percolating metal-insulator mixture ( $\epsilon_1 = 1, \epsilon_2 = 1 - \omega_{pl}^2/\omega(\omega + i\gamma)$ ,  $\gamma/\omega_{pl} = 0.01$  and  $c_0 = 0.2$ ).  $\omega_{F1}$  and  $\omega_{F2}$  are the Fröhlich modes for  $c \rightarrow 0$  and  $c \rightarrow 1$ .

The constant  $A$  in the Bergman spectral representation is connected with the DC conductivity of a composite build-up from a conducting component  $\epsilon_2$  and a non-conducting component  $\epsilon_1$  by [10]–[12]

$$A = \frac{\sigma}{\sigma_2} \tag{20}$$

which, in the case of brine-saturated rocks, obeys Archie's empirical law ([17], see also [18] for discussion)  $\sigma/\sigma_2 = ac^m$  with  $a$  being of the order of unity and  $m = 1.3, \dots, 4$ .

If we write  $A = c\bar{A}$  then  $\bar{A}$  can be interpreted as the relative fraction of component 2 that contributes to the DC conductivity, i.e. isolated regions, closed conducting paths etc decrease  $\bar{A}$  [12]. In our case (9) we have

$$A = \frac{2c^2}{1+c} \xrightarrow{c \rightarrow 0} 2c^2 \tag{21}$$

corresponding to  $a = 2$  and  $m = 2$ , and

$$\bar{A} = \frac{2c}{1+c} \tag{22}$$

i.e. the relative fraction of the component 2 contributing to DC conductivity is enhanced for all  $c$  compared with a simple linear behaviour  $\bar{A} = c$ .

(9) as a smooth interpolation between the limits  $c \rightarrow 0$  and  $c \rightarrow 1$  naturally does not result in a percolation threshold  $c_0$  with  $\sigma \equiv 0$  for  $c \leq c_0$ . Systems with percolation require

$$\frac{\sigma}{\sigma_2} = \alpha(c - c_0)^\gamma \theta(c - c_0) \tag{23}$$

where  $\theta(c - c_0) = 1$  for  $c \geq c_0$  and  $\theta(c - c_0) = 0$  otherwise.

Introducing such a form of  $A = \sigma/\sigma_2$  together with the one-mode approximation  $g(s) = \beta\delta(s - s_0)$ , the parameters  $\alpha, \beta, \gamma$  and  $s_0$  can be obtained from (11) and (12) under the additional requirements  $s_0 \rightarrow \frac{1}{3}$  for  $c \rightarrow 0$  and  $s_0 \rightarrow \frac{2}{3}$  for  $c \rightarrow 1$ .

This yields

$$\begin{aligned}\alpha &= \frac{1}{(1 - c_0)^\gamma} \\ \beta &= c - \frac{(c - c_0)^\gamma}{(1 - c_0)^\gamma} \theta(c - c_0) \quad s_0 = \frac{1}{3}(1 - c)c \frac{1}{\beta} \\ \gamma &= \frac{3}{2}(1 - c_0)\end{aligned}\tag{24}$$

resulting in a dielectric constant of a composite with a percolation threshold, which can be used e.g. for the calculation of frequency-dependent properties. An example for the frequency dependence of the TO and LO modes of a metal-insulator composite is given in figure 2.

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