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Influence of the Schulgasser inequality on effective moduli of two-phase isotropic composites

S. TOKARZEWSKI and J. J. TELEGA (WARSZAWA)

THE AIM of this paper is to study the effective transport coefficients λ_e of macroscopically isotropic two-phase composites for the case, where dielectric coefficients λ_1 and λ_2 of components are real. As an input we take: (i) N coefficients of the power expansion of $\lambda_e(x)$ at $x = 0$, where $x = (\lambda_2/\lambda_1) - 1$; (ii) the analytical property of $\lambda_e(x)$, namely $\lambda_e(-1) \geq 0$; (iii) the Schulgasser inequality $\lambda_e(x)\lambda_e(y) = (\lambda_1)^2$, $y = -x/(x+1)$. By starting from (i), (ii) and (iii), an infinite set of bounds on $\lambda_e(x)$ has been established and compared with the corresponding ones reported in literature. As an example of illustration of the obtained results, the regular arrays of spheres has been investigated numerically.

1. Introduction

THE EFFECTIVE TRANSPORT coefficients λ_e of composite materials may be evaluated by the method of bounds [5, 6, 7, 8, 12, 19, 20]. The bounds become increasingly narrow, when more information concerning the geometrical properties of the medium is available.

Milton has derived in the complex λ_e-plane an infinite set of narrowing bounds on λ_e. The calculation of his bounds requires the knowledge of successive terms of the power expansion of λ_e in $\lambda_2 - \lambda_1$. The coefficients of the expansion are geometrical in nature and their values are determined by the correlation functions of disordered geometry. Milton's approach is based on an analytic representation of the effective dielectric constant due to BERGMAN [4]. The problem of complex bounds was also discussed by FELDERHOF [12], who obtained the estimation of λ_e with the help of four characteristic geometrical functions introduced by BERGMAN [5]. Recently, interesting continued fraction representations for the set of complex bounds on λ_e were presented by BERGMAN [6] for three-, and by CLARK and MILTON [8] for two-dimensional systems.

The fundamental estimations of $\lambda_e(x)$ reported in literature [20] do not exploit the well known Schulgasser inequality $\lambda_e(x)\lambda_e(y) \geq (\lambda_1)^2$, $y = -x/(x+1)$ [22]. Direct links of this inequality with bounds for isotropic, inhomogeneous materials has been advocated by MILTON [20, p. 5297], see also [7, p. 927]. He suggested that some of the existing bounds on $\lambda_e(x)$ are not the best, cf. [20, p. 5297]. A simple case of incorporation of $\lambda_e(x)\lambda_e(y) \geq (\lambda_1)^2$ into the second order bounds on $\lambda_e(x)$ only, was studied in [6].

The main aim of this paper is to include the Schulgasser inequality $\lambda_e(x)\lambda_e(y) \geq (\lambda_1)^2$, $y = -x/(x+1)$ into an infinite set of fundamental real-valued bounds on

$\lambda_e(x)$ reported by MILTON [20]. This aim is achieved by applying Padé approximants and continued fractions to the formulation of a method of incorporation of Schulgasser inequality into lower and upper bounds on scalar, bulk transport coefficients of two-phase media, see Theorem 2.

2. Basic definitions and assumptions

This study is concerned with the effective dielectric constant Λ_e of a composite consisting of two isotropic components of dielectric moduli λ_1, λ_2 and volume fractions φ_1 and $\varphi_2 = 1 - \varphi_1$, respectively. The overall dielectric coefficient Λ_e is defined by the linear relationship between the volume-averaged electric field $\langle \mathbf{U} \rangle$ and volume-averaged displacement $\langle \mathbf{D} \rangle$:

$$\langle \mathbf{D} \rangle = \Lambda_e \langle \mathbf{U} \rangle. \tag{2.1}$$

The value $\langle \cdot \rangle$ is averaged over a representative volume or a basic cell. In general, Λ_e will be a second-order symmetric tensor, even when λ_1 and λ_2 are both scalars, and will depend on the microstructure of composite. Our consideration will be limited to one of the diagonal element of Λ_e, say λ_e, which has a well known Stieltjes integral representation [4, 9, 10]

$$G(x) = \frac{\lambda_e(x)}{\lambda_1} - 1 = x \int_0^1 \frac{d\gamma(u)}{1 + xu}, \tag{2.2}$$

where

$$x = h - 1, \qquad h = \frac{\lambda_1}{\lambda_2}. \tag{2.3}$$

Here $G(x)$ is defined for $x \in (-1, \infty)$, cf. [6, 12]. The spectrum $\gamma(u)$ appearing in (2.2) is a real, bounded and non-decreasing function determined for $0 \le u < \infty$. The representation (2.2) was introduced by BERGMAN [6] and referred to as characteristic, geometrical function.

Let us consider the power expansion of (2.2)

$$G(x) = \sum_{n=1}^{\infty} G_n x^n, \tag{2.4}$$

where

$$G_n = (-1)^{n-1} \int_0^{\infty} u^{n-1}\, d\gamma(u). \tag{2.5}$$

For composite materials the coefficients G_n $(n = 1, 2, \ldots,)$ are finite and series (2.4) is convergent for $|x| < 1$.

Now we are in a position to introduce the Padé approximants to Stieltjes series (2.4). To this end we consider the following rational functions

$$(2.6) \qquad [M+J/M] = \frac{L_{M+J}(x)}{P_M(x)} = \frac{a_1^{(J)}x + \cdots + a_{M+J}^{(J)}x^{M+J}}{1 + b_1^{(J)}x + \cdots +{}_M^{(J)} x^M}, \qquad J = 0, 1,$$

with the power expansion of $[M + J/M]$ at $x = 0$

$$(2.7) \qquad [M+J/M](x) = \sum_{n=1}^{\infty} G_{n,J}x^n, \qquad J = 0, 1.$$

The functions (2.6) are the subdiagonal ($J = 0$) and diagonal ($J = 1$) Padé approximants $[M + J/M]$ to the Stieltjes function (2.2), provided that

$$(2.8) \qquad G_{n,J} = G_n \qquad \text{for} \qquad n = 1, 2, \ldots, 2M + J, \qquad J = 0, 1.$$

Padé approximants (2.6) can also be expressed in the form of S-continued fractions

$$(2.9) \qquad [M+J/M](x) = \frac{g_1x}{1} \, {}_{+} \, \frac{g_2x}{1} \, {}_{+\cdots+} \, \frac{g_{2M+J}x}{1}, \qquad J = 0, 1,$$

equivalent to the following explicit expression, see [1, 26]

$$[M+J/M](x) = \cfrac{g_1x}{1 + \cfrac{g_2x}{\cdots \; 1 + \cfrac{g_{2M+J-1}x}{1 + g_{2M+1}x}}}.$$

The coefficients $g_1, \ldots, g_{2M+J}$ appearing in (2.9) are positive and uniquely determined by the $2M + J$ coefficients G_n ($n = 1, 2, ..., 2M + J$; $J = 0, 1$) of a Stieltjes series (2.4).

After this preparation, we can recall the infinite set of fundamental bounds on $\lambda_e(x)$ derived by MILTON in [20]. By expanding his estimations $U_{N,0}(\varrho)$ and $V_{N,0}(\varrho)$ ($\varrho = x/(x+2)$; $x = \lambda_2/\lambda_1 - 1$) [20, p. 5296] into S-continued fractions dependent on x, we obtain:

THEOREM 1. *For two-phase inhomogeneous media, the S-continued fractions* (2.9) *generated by power expansion* (2.4) *obey the following inequalities:*

(i) *If $x \geq 0$ then*

$$(2.10) \qquad V_{N,0}(x) \geq (-1)^N \frac{\lambda_e}{\lambda_1} \geq (-1)^N N_{N,0}(x).$$

(ii) *If $-1 \leq x \leq 0$, then*

$$(2.11) \qquad V_{N,0}(x) \leq \frac{\lambda_e}{\lambda_1} \leq U_{N,0}(x),$$

where C_{N+1} is given by the following recurrence formula

$$C_1 = 1, \qquad C_p = \frac{g_p}{1 - C_{p+1}}, \qquad p = 1, 2, ...N, \tag{2.12}$$

while $U_{N,0}(x)$ and $V_{N,0}(x)$ take the following S-continued fraction forms

$$\begin{aligned} U_{N,0}(x) &= 1 + \frac{g_1 x}{1} \, \frac{g_2 x}{+ \, 1} + \cdots + \frac{g_N x}{1}, \\ V_{N,0}(x) &= 1 + \frac{g_1 x}{1} \, \frac{g_N x}{+ \, 1} + \cdots + \frac{C_{N+1} x}{1}. \end{aligned} \tag{2.13}$$

Here $U_{N,0}(x)$ is a Padé approximant given by (2.9) *to power series* (2.4)*, N denotes the number of known coefficients of a power series expansion* (2.4)*, while $x = (\lambda_2/\lambda_1) - 1$.*

For macroscopically isotropic composites the well known Schulgasser inequality holds [22]:

$$\frac{\lambda_e(x)}{\lambda_1} \frac{\lambda_e(y)}{\lambda_1} \geq 1, \qquad \text{if} \qquad y = -\frac{x}{x+1} \qquad \text{and} \qquad x > -1. \tag{2.14}$$

The main purpose of this paper is to incorporate the relation (2.14) into S-fraction bounds (2.10)–(2.11).

3. Schulgasser inequality $\lambda_e(x)\lambda_e(y) \geq (\lambda_1)^2$

Let us consider the following class of S-continued fractions

$$\psi_{N+1}(x, q_{N+1}) = 1 + \frac{g_1 x}{1} \, \frac{g_2 x}{+ \, 1} + \cdots + \frac{g_N x}{1} \, \frac{q_{N+1} x}{+ \, 1}. \tag{3.1}$$

Here $g_j > 0$ ($j = 1, 2, ..., N$) are uniquely determined by N terms of a power expansion of λ_e/λ_1, while q_{N+1} is a free parameter belonging to the interval

$$R_{N+1,0} = \left\{ q_{N+1} \mid q_{N+1} \geq 0 \right\}. \tag{3.2}$$

Now we will seek the interval $R_{N+1,1}(x)$ of admissible values of q_{N+1} defined by

$$R_{N+1,1}(x) = \left\{ q_{N+1} \mid \psi_{N+1}(x, q_{N+1}) \psi_{N+1}(y, q_{N+1}) \geq 1 \right\}, \tag{3.3}$$

where $y = -x/(x+1)$. It is obvious that q_{N+1} determined by (3.3) satisfy the Schulgasser relation (2.14)

$$\psi_{N+1}(x, q_{N+1}) \psi_{N+1}(y, q_{N+1}) \geq 1. \tag{3.4}$$

Of interest is the equality

$$\psi_{N+1}(x, q_{N+1})\psi_{N+1}(y, q_{N+1}) = 1, \qquad y = -x/(x+1), \tag{3.5}$$

i.e.:

$$\left(1 + \frac{g_1 x}{1} {}_{+} \cdots {}_{+} \frac{g_N x}{1} {}_{+} \frac{q_{N+1} x}{1}\right)\left(1 + \frac{g_1 y}{1} {}_{+} \cdots {}_{+} \frac{g_{N+1} y}{1} {}_{+} \frac{q_{N+1} y}{1}\right) = 1. \tag{3.6}$$

The recurrence formula for S-continued fractions reported in [2, Chap. 4.2] yields

$$\left(1 + \frac{g_1 z}{1} {}_{+} \cdots {}_{+} \frac{g_N z}{1} {}_{+} \frac{q_{N+1} z}{1}\right) = \frac{A_N(z) + A_{N-1}(z) q_{N+1}}{B_N(z) + B_{N-1}(z) q_{N+1}}, \tag{3.7}$$

where $A_N(z)$ and $B_N(z)$ are polynomials determined by

$$A_{-1} = 1, \quad A_0 = 1, \quad A_j(z) = A_{j-1}(z) + z g_j A_{j-2}(z), \quad j = 1, 2, ..., N, \tag{3.8}$$

$$B_{-1} = 0, \quad B_0 = 1, \quad B_j(z) = B_{j-1}(z) + z g_j B_{j-2}(z), \quad j = 1, 2, ..., N. \tag{3.9}$$

On the basis of (3.7), relation (3.6) takes the form

$$\frac{A_N(x) + x A_{N-1}(x) q_{N+1}}{B_N(x) + x B_{N-1}(x) q_{N+1}} \frac{A_N(y) + y A_{N-1}(y) q_{N+1}}{B_N(y) + y B_{N-1}(y) q_{N+1}} = 1. \tag{3.10}$$

Here $y = -x/(x+1)$. Simple rearrangements of (3.10) yield

$$\alpha_{N+1}(x) q_{N+1}^2 + \beta_{N+1}(x) q_{N+1} + \delta_{N+1}(x) = 0, \tag{3.11}$$

where

$$\alpha_{N+1}(x) = xy\left[A_{N-1}(x)A_{N-1}(y) - B_{N-1}(x)B_{N-1}(y)\right], \tag{3.12}$$

$$\begin{aligned}\beta_{N+1}(x) = {} & x\left[A_{N-1}(x)A_N(y) - B_{N-1}(x)B_N(y)\right] \\ & + y\left[A_N(x)A_{N-1}(y) - B_N(x)B_{N-1}(y)\right],\end{aligned} \tag{3.13}$$

$$\delta_{N+1}(x) = A_N(x)A_N(y) - B_N(x)B_N(y). \tag{3.14}$$

The solutions of (3.11) are given by

$$\begin{aligned} q'_{N+1}(x) &= -\frac{\beta_{N+1}(x)}{2\alpha_{N+1}(x)}\left[1 + \sqrt{1 - \frac{4\alpha_{N+1}(x)\delta_{N+1}(x)}{\beta_{N+1}^2(x)}}\right], \\ q''_{N+1}(x) &= -\frac{\beta_{N+1}(x)}{2\alpha_{N+1}(x)}\left[1 - \sqrt{1 - \frac{4\alpha_{N+1}(x)\delta_{N+1}(x)}{\beta_{N+1}^2(x)}}\right]. \end{aligned} \tag{3.15}$$

On account of (3.3) and (3.15) we have

(i) if $\alpha_{N+1}(x) \le 0$, then

$$R_{N+1,1}(x) = \left\{q_{N+1} \mid q''_{N+1}(x) \le q_{N+1} \le q'_{N+1}(x)\right\}, \tag{3.16}$$

(ii) if $\alpha_{N+1}(x) \ge 0$, then

$$R_{N+1,1}(x) = \left\{q_{N+1} \mid q_{N+1}\tau \le q''_{N+1} \vee q_{N+1}\tau \ge q'_{N+1}\right\}. \tag{3.17}$$

According to definition (3.3), a class of bounds given by

$$\psi_{N+1}(x, q_{N+1}), \qquad q_{N+1} \in R_{N+1,1}(x) \tag{3.18}$$

satisfies the Schulgasser inequality (2.14).

4. Inequality $\lambda_e(x)/\lambda_1 \ge \Lambda(x)$

Let us assume now that for fixed $x = (\lambda_2/\lambda_1) - 1$, the lower bound $\Lambda(x)$ on the effective modulus $\lambda_e(x)/\lambda_1$ is known,

$$\lambda_e(x)/\lambda_1 \ge \Lambda(x). \tag{4.1}$$

By using (3.1) we can write

$$\psi_{N+1}(x, q_{N+1}) \ge \Lambda(x). \tag{4.2}$$

Of interest is the equality, cf. $(2.10)_2$ and (2.14),

$$\psi_{N+1}(x, C_{N+1}) = \left(1 + \frac{g_1 x}{1} {}_{+ \cdots +} \frac{g_N x}{1} {}_{+} \frac{C_{N+1} x}{1}\right) = \Lambda(x). \tag{4.3}$$

By applying recurrence formulae (3.8)–(3.9) to continued fraction (4.3), we obtain

$$\Lambda(x) = \frac{A_N(x) + x A_{N-1}(x) C_{N+1}}{B_N(x) + x B_{N-1}(x) C_{N+1}}. \tag{4.4}$$

Hence

$$C_{N+1}(x) = \frac{\Lambda(x) B_N(x) - A_N(x)}{x[A_{N-1}(x) - \Lambda(x) B_{N-1}(x)]}. \tag{4.5}$$

Now we are in a position to introduce the interval $R_{N+1,2}(x)$ of admissible values of q_{N+1} given by

$$R_{N+1,2}(x) = \left\{q_{N+1} \mid \psi_{N+1}(x, q_{N+1}) \ge \Lambda(x)\right\}. \tag{4.6}$$

On account of (4.5) and (4.6), $R_{N+1,2}(x)$ takes a form

$$R_{N+1,2}(x) = \Big\{q_{N+1} \mid q_{N+1} \le C_{N+1}(x)\Big\}. \tag{4.7}$$

Note that, according to (4.6) and (4.7), a class of bounds determined by

$$\psi_{N+1}(x, q_{N+1}), \qquad q_{N+1} \in R_{N+1,2}(x) \tag{4.8}$$

satisfies the inequality (4.2).

5. Bounds exploiting Schulgasser inequality

Let us introduce an interval $R_{N+1}(x)$

$$R_{N+1}(x) = R_{N+1,0} \cap R_{N+1,1}(x) \cap R_{N+1,2}(x), \tag{5.1}$$

where $R_{N+1,0}$, $R_{N+1,1}(x)$ and $R_{N+2,2}(x)$ are defined by (3.2), (3.16)–(3.17) and (4.7), respectively. Note that the class of functions

$$\psi_{N+1}(x, q_{N+1}), \qquad q_{N+1} \in R_{N+1}(x) \tag{5.2}$$

satisfy the inequalities (2.14) and (4.1). For $x \to -1^+$ the lower estimation of $\lambda_e(x)$ is well known, cf. [4, 5, 6, 23]

$$\Lambda(-1^+) = 0. \tag{5.3}$$

For such a case it is convenient to introduce the notation

$$\lim_{x \to -1^+} Q(x) \equiv Q(-1^+) \equiv Q(-1), \tag{5.4}$$

consequently used in the sequel. Now we are ready to formulate the theorem solving the problem of incorporation of the Schulgasser inequality (2.14) into bounds (2.10)–(2.12).

THEOREM 2. *For macroscopically isotropic two-phase inhomogeneous media, the S-continued fractions* (2.9) *generated by power expansion* (2.4) *obey the following inequalities:*

(i) *If* $x \ge 0$ $(x = (\lambda_2/\lambda_1) - 1)$, *then*

$$\begin{gathered}
(-1)^N \psi_{N+1}(x, E_{N+1}) \ge (-1)^N \frac{\lambda_e(x)}{\lambda_1} \ge (-1)^N \psi_N(x), \\
\psi_N(x) = 1 + \frac{g_1 x}{1} \, \frac{g_2 x}{1} \, \frac{}{+ \cdots +} \, \frac{g_N x}{1}, \\
\psi_{N+1}(x, E_{N+1}) = 1 + \frac{g_1 x}{1} \, \frac{g_2 x}{1} \, \frac{}{+ \cdots +} \, \frac{g_N x}{1} \, \frac{E_{N+1} x}{1}.
\end{gathered} \tag{5.5}$$

(ii) *If* $-1 \le x \le 0$ $(x = (\lambda_2/\lambda_1) - 1)$, *then*

$$\psi_N(x) \ge \frac{\lambda_e(x)}{\lambda_1} \ge \psi_{N+2}(x, E_{N+1}, H_{N+2}),$$

$$\psi_N(x) = 1 + \frac{g_1 x}{1} \underset{+\cdots+}{} \frac{g_N x}{1}, \tag{5.6}$$

$$\psi_{N+2}(x, E_{N+1}, H_{N+2}) = 1 + \frac{g_1 x}{1} \underset{+\cdots+}{} \frac{g_N x}{1} \underset{+}{} \frac{E_{N+1} x}{1} \underset{+}{} \frac{H_{N+2} x}{1}.$$

Here the coefficients H_{N+2} *and* E_{N+1} *are given by*

$$H_{N+2} = \frac{A_N(-1) - E_{N+1} A_{N-1}(-1)}{A_N(-1)}, \quad E_{N+1} = \min\{D_{N+1}, C_{N+1}\}, \tag{5.7}$$

$$D_{N+1} = \max\left\{q'_{N+1}(-1), q''_{N+1}(-1)\right\}, \quad C_{N+1} = \frac{A_N(-1)}{A_{N-1}(-1)}, \tag{5.8}$$

where $q'_{N+1}(-1)$, $q''_{N+1}(-1)$ *are determined by* (3.15). *Relation* $(5.8)_2$ *is a consequence of* (4.5) *and* (5.3), *while* N *appearing in* (5.5)–(5.8) *denotes the number of known coefficients of power series* (2.4).

P r o o f. It follows from Appendix A that $\alpha_{N+1}(-1) \le 0$ and $\delta_{N+1}(-1) \ge 0$. Thus the roots of (3.11) q'_{N+1} and q''_{N+1} have opposite signs, cf. (3.15). On account of (5.1), (5.7) and (5.8), we get

$$R_{N+1}(-1) = \left\{\tau \mid 0 \le \tau \le E_{N+1}\right\}. \tag{5.9}$$

Hence the class of bounds (5.2) takes a form

$$\psi_{N+1}(x,\tau) = 1 + \frac{g_1 x}{1} \underset{+\cdots+}{} \frac{g_N x}{1} \underset{+}{} \frac{\tau x}{1}, \qquad 0 \le \tau \le E_{N+1}. \tag{5.10}$$

The first derivative of $\psi_{N+1}(x,\tau)$ with respect to τ satisfies

$$\frac{\partial \psi_{N+1}(x,\tau)}{\partial \tau} > 0, \quad \text{for} \quad x \in (0,\infty), \quad 0 \le \tau \le E_{N+1} \quad \text{and} \quad N = 0, 2, \dots,$$
$$\frac{\partial \psi_{N+1}(x,\tau)}{\partial \tau} < 0, \quad \text{for} \quad x \in (0,\infty), \quad 0 \le \tau \le E_{N+1} \quad \text{and} \quad N = 1, 3, \dots. \tag{5.11}$$

Hence the continued fraction $\psi_{N+1}(x,\tau)$ $(x \in (0,\infty))$ defined by (5.10) assumes its extremal values for

$$\tau = 0 \qquad \text{and} \qquad \tau = E_{N+1}. \tag{5.12}$$

By substituting (5.12) into (5.10) we obtain the formula (5.5).

If $-1 \le x \le 0$, the inequalities (5.6) result from the relations:

$$0 < g_{N+1} \le E_{N+1}, \quad \lambda_e/\lambda_1 \ge \psi_{N+2}(x, C_{N+2}) \ge \psi_{N+2}(x, E_{N+1}, H_{N+2}), \tag{5.13}$$

where

$$(5.14)\qquad \psi_{N+2}(x, C_{N+2}) = 1 + \frac{g_1 x}{1} {}_{+\cdots+} \frac{g_N x}{1} {}_{+} \frac{g_{N+1} x}{1} {}_{+} \frac{C_{N+2} x}{1},$$

$$(5.15)\qquad \psi_{N+2}(x, E_{N+1}, H_{N+2}) = 1 + \frac{g_1 x}{1} {}_{+\cdots+} \frac{g_N x}{1} {}_{+} \frac{E_{N+1} x}{1} {}_{+} \frac{H_{N+2} x}{1}. \quad \square$$

Note that for $D_{N+1} \geq C_{N+1}$, the bounds determined by Th. 2 reduce to the existing ones defined by Th. 1, since the parameters C_{N+1} given by (2.12) and $(5.8)_2$ coincide, while $H_{N+2} = 0$. Hence the estimations (5.5)–(5.6) obtained in the present paper can not be worse than the previous bounds (2.10)–(2.11) reported in literature [20]. Moreover, for some cases they have to be better. In the next section we demonstrate the analytical form of a low order bounds on $\lambda_e(x)/\lambda_1$ given by (5.5) and (5.6).

6. Low order bounds on λ_e

To illustrate Th. 2 we will evaluate bounds on an effective dielectric constant $\lambda_e(x)$ for the cases, where (i) no coefficients ($N = 0$), (ii) one coefficient ($N = 1$) and (iii) two coefficients ($N = 2$) of the power expansion of $\lambda_e(x)/\lambda_1$ are available.

(i) The recurrence formulae (3.8) and (3.9) give:

$$(6.1)\qquad A_{-2} = 0, \quad A_{-1} = 1, \quad A_0 = 1, \quad B_{-2} = 0, \quad B_{-1} = 0, \quad B_0 = 1.$$

Then relations (3.12)–(3.14) yield

$$(6.2)\qquad \alpha_1(x) = xy, \qquad \beta_1(x) = x + y, \qquad \delta_1(x) = 0.$$

Hence from (3.15), (4.5) we get

$$(6.3)\qquad q_1' = -\frac{x+y}{xy}, \qquad q_1'' = 0, \qquad C_1 = -\frac{1}{x}, y = -x/(x+1).$$

For $x = -1^+$ the equations (6.3) reduce to

$$(6.4)\qquad q_1' = 1, \qquad q_1'' = 0, \qquad C_1 = 1.$$

From (5.7) and (5.8), it follows that

$$(6.5)\qquad D_1 = 1, \qquad E_1 = 1.$$

Hence, on the basis of Th. 2 the bounds on λ_e are given by

$$(6.6)\qquad 1 \geq \frac{\lambda_e}{\lambda_1} \geq 1 + x, \quad \text{if} \quad -1 \leq x \leq 0; \qquad 1 + x \geq \frac{\lambda_e}{\lambda_1} \geq 1, \quad \text{if} \quad x \geq 0.$$

(ii) $N = 1$. Then

$$A_{-2} = 0, \qquad A_{-1} = 1, \qquad A_0 = 1, \qquad B_{-1} = 0, \qquad B_0 = 1, \tag{6.7}$$

$$\begin{gathered} \alpha_2(x) = 0, \qquad \beta_2(x) = 2g_1xy, \\ \delta_2(x) = g_1x + g_1y + g_1^2xy, \qquad y = -x/(x+1), \end{gathered} \tag{6.8}$$

$$q_2' = -\infty, \qquad q_2'' = -\frac{g_1x + g_1y + g_1^2xy}{2g_1xy}, \qquad C_2 = \frac{-(1+g_1x)}{x}. \tag{6.9}$$

For $x = -1^+$ we have

$$\begin{gathered} q_2' = -\infty, \qquad q_2'' = \frac{1-g_1}{2}, \qquad C_2 = 1 - q_1, \\ D_2 = \frac{1-g_1}{2}, \qquad E_2 = \frac{1-g_1}{2}, \qquad H_3 = \frac{1}{2}. \end{gathered} \tag{6.10}$$

From (5.5), (5.6) and (6.10) we readily obtain

$$1 + \frac{g_1x}{1}{}_{+}\frac{(1-g_1)x/2}{1} \le \frac{\lambda_e}{\lambda_1} \le 1 + g_1x. \tag{6.11}$$

(iii) $N = 2$. Now we have

$$\alpha_3(x) = xy[(1+g_1x)(1+g_1y) - 1], \qquad y = -x/(x+1), \tag{6.12}$$

$$\beta_3(x) = xg_1[x + y + (g_1+g_2)xy] + yg_2[x + y + (g_1+g_2)xy], \qquad y = -x/(x+1), \tag{6.13}$$

$$\delta_3(x) = g_1x(1+g_2y) + g_1y(1+g_2x) + g_2g_2xy, \qquad y = -x/(x+1). \tag{6.14}$$

Thus for $x = -1^+$

$$q_3' = \frac{1-g_1-g_2}{1-g_1}, \qquad q_3'' = 0, \qquad C_3 = \frac{1-g_1-g_2}{1-g_1}. \tag{6.15}$$

Hence

$$\begin{gathered} 1 + \frac{g_1x}{1}{}_{+}\frac{g_2x}{1}{}_{+}\frac{\dfrac{(1-g_1-g_2)x}{1-g_1}}{1} \ge \frac{\lambda_e}{\lambda_1} \ge 1 + \frac{g_1x}{1}{}_{+}\frac{g_2x}{1}, \quad \text{if} \quad -1 \le x \le 0, \\ 1 + \frac{g_1x}{1}{}_{+}\frac{g_2x}{1}{}_{+}\frac{\dfrac{(1-g_1-g_2)x}{1-g_1}}{1} \le \frac{\lambda_e}{\lambda_1} \le 1 + \frac{g_1x}{1}{}_{+}\frac{g_2x}{1}, \quad \text{if} \quad x \ge 0. \end{gathered} \tag{6.16}$$

It is interesting to compare the low order bounds existing in literature (Th. 1) with the bounds incorporating the Schulgasser inequality (Th. 2). The basic bounds (6.6) are the same, the estimations (6.11) are more restrictive than the well known Wiener bounds [27] (Fig. 1), while the inequalities (6.16) coincide with Hashin - Shtrikman bounds reported in [14].

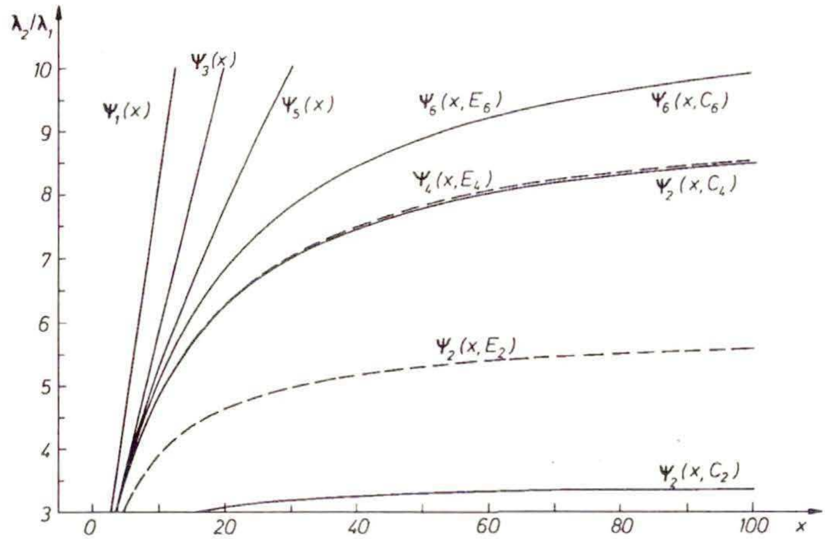

FIG. 1. Existing (—) and improved (– – –) bounds on the effective dielectric constant of a face-centered lattice of spheres for volume fraction $\varphi_2 = 0.71$. Upper bounds $\Psi_N(x)$ $(N = 1, 3, 5)$ coincide, while lower ones $\Psi_{N+1}(x, C_{N+1})$ and $\Psi_{N+1}(x, E_{N+1})$ differ significantly for $N = 1$ and slightly for $N = 3, 5$.

7. Even number of terms of a power expansion of λ_e

In this section we will compare the known (2.10)–(2.11) and obtained (5.5)–(5.6) bounds calculated from an even number $(N = 0, 2, 4, ...)$ of coefficients of power series (2.4). To this end we prove that for $x \to -1^+$, thus $y = -x/(x+1) \to \infty$ $(N = 0, 2, ...)$, the expressions (3.15) reduce via (3.12)–(3.14) to

(7.1)
$$\lim_{x \to -1} 2\alpha_{N+1}(x) \neq 0,$$

(7.2)
$$q'_{N+1} = \lim_{x \to -1^+} -\frac{\beta_{N+1}(x)}{2\alpha_{N+1}(x)} \left(1 + \sqrt{1 - \frac{4\alpha_{N+1}(x)\delta_{N+1}(x)}{\beta^2_{N+1}(x)}}\right) = \frac{A_N(-1)}{A_{N-1}(-1)},$$
$$q''_{N+1} = \lim_{x \to -1^+} -\frac{\beta_{N+1}(x)}{2\alpha_{N+1}(x)} \left(1 - \sqrt{1 - \frac{4\alpha_{N+1}(x)\delta_{N+1}(x)}{\beta^2_{N+1}(x)}}\right) = 0.$$

P r o o f. The recurrence formulae (3.8) and (3.9) for S-continued fractions [2] and the Schulgasser inequality (3.4) yields

(7.3)
$$\frac{A_N(x)A_N(y)}{B_N(x)B_N(y)} \geq 1,$$
$$A_N(x)A_N(y) > 0, \qquad B_N(x)B_N(y) > 0,$$
$$\lim_{x \to -1^+} \frac{A_N(y)}{A_{N-1}(y)} \leq \infty, \qquad \lim_{x \to -1^+} \frac{B_N(y)}{A_N(y)} \leq \infty.$$

For even N, on the basis of (3.12), (3.14) and (7.3), we have

$$\alpha_{N+1}(x) = xy\delta_N(x) = xyB_{N-1}(x)B_{N-1}(y)\left(\frac{A_{N-1}(x)A_{N-1}(y)}{B_{N-1}(x)B_{N-1}(y)} - 1\right) \neq 0, \tag{7.4}$$

$$\lim_{x\to-1^+}\frac{\beta_{N+1}(x)}{2\alpha_{N+1}(x)} = \frac{1}{y}\,\frac{A_N(y))}{A_{N-1}(y)}\,\frac{\left(A_{N-1}(x) - B_{N-1}(x)\dfrac{B_N(y)}{A_N(y)}\right)}{\left(A_{N-1}(x) - B_{N-1}(x)\dfrac{B_{N-1}(y)}{A_{N-1}(y)}\right)} \tag{7.5}$$

$$+\frac{1}{x}\,\frac{\left(A_N(x) - B_N(x)\dfrac{B_{N-1}(y)}{A_{N-1}(y)}\right)}{\left(A_{N-1}(x) - B_{N-1}(x)\dfrac{B_{N-1}(y)}{A_{N-1}(y)}\right)} = -\frac{A_N(-1)}{A_{N-1}(-1)},$$

$$\lim_{x\to-1^+}\frac{\alpha_{N+1}(x)}{\beta_{N+1}(x)}\,\frac{\delta_{N+1}(x)}{\beta_{N+1}(x)} = \lim_{x\to-1^+}\frac{\alpha_{N+1}(x)}{\beta_{N+1}(x)}\lim_{x\to-1^+}\frac{\delta_{N+1}(x)}{\beta_{N+1}(x)}$$
$$= \frac{A_N(-1)}{A_{N-1}(-1)}\lim_{x\to-1^+}\frac{\delta_{N+1}(x)}{\beta_{N+1}(x)},$$

$$\lim_{x\to-1^+}\frac{\delta_{N+1}(x)}{\beta_{N+1}(x)} = \lim_{x\to-1^+} \tag{7.6}$$

$$\frac{\left(A_N(x) - B_N(x)\dfrac{B_N(y)}{A_N(y)}\right)}{x\left(A_{N-1}(x) - B_{N-1}(x)\dfrac{B_N(y)}{A_N(y)}\right) + \dfrac{yA_{N-1}(y)}{A_N(y)}\left(A_N(x) - B_N(x)\dfrac{B_{N-1}(y)}{A_{N-1}(y)}\right)} = 0.$$

From (7.4)–(7.6), follow the relations (7.1) and (7.2). □

For $A(-1) = 0$ and even N ($N = 0, 2, \ldots$), the relation (4.5) coincides with $(7.2)_1$. Hence inequalities (5.5) and (5.6) agree with (2.10) and (2.11). Consequently for even N, the S-continued fraction method based on the Schulgasser inequality (2.14) does not provide better bounds than the approaches neglecting this inequality. Therefore an improvement of the existing bounds on $\lambda_e(x)$ can be expected for odd N ($N = 1, 3, ..$) of coefficients of power expansion of $\lambda_e(x)$ only.

8. Regular arrays of spheres

Now we are prepared to apply Th. 2 to regular lattices of spheres embedded in an infinite matrix. By λ_e, λ_2 and λ_1 we denote the dielectric constants of the composite, spheres and matrix, respectively. The first three coefficients of the power expansion of $(\lambda_e/\lambda_1) - 1$ are as follows [4], cf. (2.2), (2.4):

$$\frac{\lambda_e}{\lambda_1} - 1 = \varphi_2 x - \frac{1}{3}\varphi_1\varphi_2 x^2 + O(x^3), \tag{8.1}$$

where, as previously, $x = (\lambda_2/\lambda_1) - 1$. Here φ_2, φ_1 denote volume fractions of the spheres and matrix. On the basis of (2.6), S-continued fractions (2.9) associated with (8.1) are expressed by

$$(8.2) \qquad [0/0] = 0, \qquad [1/0] = \frac{\varphi_2 x}{1}, \qquad [1/1] = \frac{\varphi_2 x}{1} {}_{+} \frac{(\varphi_1/3)x}{1},$$

where

$$(8.3) \qquad g_1 = \varphi_2, \qquad g_2 = \varphi_1.$$

Hence from (6.6), (6.11) and (6.16) we have:

(i) for $N = 0$

$$(8.4) \qquad \begin{aligned} 1 \geq \lambda_e/\lambda_1 \geq 1 + x, \qquad \text{if} \quad \lambda_2 \leq \lambda_1, \\ 1 \leq \lambda_e/\lambda_1 \leq 1 + x, \qquad \text{if} \quad \lambda_2 \geq \lambda_1; \end{aligned}$$

(ii) for $N = 1$

$$(8.5) \qquad 1 + \frac{\varphi_2 x}{1} {}_{+} \frac{\varphi_1 x/2}{1} \leq \frac{\lambda_e}{\lambda_1} \leq 1 + \frac{\varphi_2 x}{1};$$

(iii) for $N = 2$

$$(8.6) \qquad \begin{aligned} 1 + \frac{\varphi_2 x}{1} {}_{+} \frac{\varphi_1 x/3}{1} \geq \frac{\lambda_e}{\lambda_1} \geq 1 + \frac{\varphi_2 x}{1} {}_{+} \frac{\varphi_1 x/3}{1} {}_{+} \frac{2x/3}{1}, \qquad \text{if} \quad \lambda_2 \leq \lambda_1, \\ 1 + \frac{\varphi_2 x}{1} {}_{+} \frac{\varphi_1 x/3}{1} \leq \frac{\lambda_e}{\lambda_1} \geq 1 + \frac{\varphi_2 x}{1} {}_{+} \frac{\varphi_1 x/3}{1} {}_{+} \frac{2x/3}{1}, \qquad \text{if} \quad \lambda_2 \geq \lambda_1. \end{aligned}$$

According to the results of Sec. 7 valid for even N, the bounds (8.4) and (8.6) agree with the existing bounds following from Th. 1, where (8.6) are Hashin–Shtrikman bounds. Of interest is the case (8.5). For $N = 1$, from Th. 1 follow the well known Wiener bounds [27]

$$(8.7) \qquad 1 + \frac{\varphi_2 x}{1} {}_{+} \frac{\varphi_1 x}{1} \leq \frac{\lambda_e}{\lambda_1} \leq 1 + \frac{\varphi_2 x}{1}.$$

By comparing (8.5) with (8.7) we conclude that incorporation of the Schulgasser inequality (Th. 2) improves lower bound of WIENER [27], while the upper one remains the same (Fig. 1). To determine bounds more exactly, further terms of the power expansion of $\lambda_e(x)/\lambda_1$ are required. For simple, body-centered and face-centered, cubic lattices of spheres, MCPHEDRAN and MILTON [16] evaluated the coefficients of a power series expansion of $\lambda_e(\alpha)/\lambda_1$, $\alpha = x/(x+2)$ at $\alpha = 0$, and gathered them in tables as discrete functions of φ_2. In [25] we derive a simple formula relating the terms of a power series of $\lambda_e(x)/\lambda_1$ to the terms of

Table 1. Low order coefficients G_n, g_n, C_{N+1}, E_{N+1}, H_{N+2} for evaluation of S-continued fraction bounds for the effective conductivity of regular arrays of spheres.

Arrays of spheres		$n=1$	$n=2$	$n=3$	$n=4$	$n=5$	$n=6$	$n=7$
$\varphi_2 = 0.52$	G_n	0.52	0.0832	0.0248	0.0102	0.0050	0.0028	
Simple	g_n	0.52	0.1600	0.1380	0.2420	0.1727	0.2579	
cubic	C_n	1.00	0.4800	0.6667	0.7930	0.6949	0.7514	0.6568
	E_n	1.00	0.2400	0.6667	0.7427	0.6949	0.7473	0.6568
	H_n		0.0000	0.5000	0.0000	0.0634	0.0000	0.0055
$\varphi_2 = 0.67$	G_n	0.67	0.0737	0.0155	0.0053	0.0025	0.0015	
Body-	g_n	0.67	0.1100	0.1009	0.2761	0.2020	0.2566	
centered	C_n	1.00	0.3300	0.6667	0.8486	0.6747	0.7006	0.6337
	E_n	1.00	0.1650	0.6667	0.8082	0.6747	0.6960	0.6337
	H_n		0.0000	0.5000	0.0000	0.0476	0.0000	0.0066
$\varphi_2 = 0.71$	G_n	0.71	0.0686	0.0147	0.0058	0.0030	0.0018	
Face-	g_n	0.71	0.0967	0.1171	0.3342	0.1221	0.3168	
centered	C_n	1.00	0.2900	0.6667	0.8244	0.5947	0.7947	0.6013
	E_n	1.00	0.1450	0.6667	0.7794	0.5947	0.7889	0.6013
	H_n		0.0000	0.5000	0.0000	0.0546	0.0000	0.0074

the power expansion of $\lambda_e(\alpha)/\varepsilon_1$, $\alpha = x/(x+2)$. From the coefficients given in [16, Tabs. 6, 7, 8] we have calculated, by using the algorithm proposed by us in [25], the coefficients G_n of power series (2.4). The coefficients g_n, C_{N+1} and E_{N+1} gathered in Table 1 are evaluated by means of the numerical procedure proposed in [25]. Note that for even n (odd N), the coefficients E_{N+1} ($n = N+1$) are smaller than C_{N+1}, while for odd n (even N) they take the same values. For face-centered cubic arrays of spheres (fcc) the existing bounds and the improved ones are presented in Tables 2 and 3.

Table 2. Existing $\{\psi_N(x), \psi_{N+1}(x, C_{N+1})$, Th. 1$\}$ and improved $\{\psi_N(x), \psi_{N+2}(x, E_{N+1}, H_{N+2})$ Th. 2$\}$ low order bounds on $\lambda_e(x)/\lambda_1$ for the fcc lattice of spheres.

φ_2	N	x	$\psi_N(x)$	$\psi_{N+2}(x, E_{N+1}, H_{N+2})$	$\psi_{N+1}(x, C_{N+1})$
	1	−0.5	0.6450	0.607011	0.584795
0.71	3	−0.5	0.6258	0.624909	0.624863
	5	−0.5	0.6255	0.625497	0.625497
	1	−0.7	0.5030	0.411030	0.376512
0.71	3	−0.7	0.4634	0.457736	0.457466
	5	−0.7	0.4621	0.461837	0.461835
	1	−0.9	0.3610	0.162217	0.135318
0.71	3	−0.9	0.2921	0.252278	0.250850
	5	−0.9	0.2872	0.282345	0.282319

Table 3. Existing $\{\psi_N(x), \psi_{N+1}(x, C_{N+1})$, Th. 1$\}$ and improved $\{\psi_N(x), \psi_{N+1}(x, E_{N+1})$ Th. 2$\}$ low order bounds on $\lambda_e(x)/\lambda_1$ for the fcc lattice of spheres.

φ_2	N	x	$\psi_N(x)$	$\psi_{N+1}(x, E_{N+1})$	$\psi_{N+1}(x, C_{N+1})$
	1	50.0	36.500	5.303030	3.290323
0.71	3	50.0	21.817	7.806020	7.768516
	5	50.0	13.861	8.872180	8.870695
	1	70.0	50.700	5.457399	3.333333
0.71	3	70.0	29.629	8.206098	8.163556
	5	70.0	17.539	9.442256	9.440478
	1	90.0	64.900	5.548043	3.357934
0.71	3	90.0	37.427	8.449407	8.403644
	5	90.0	21.133	9.796655	9.794679

9. Concluding remarks

By starting from: (i) N coefficients of the power expansion of $\lambda_e(x)$ at $x = 0$, (ii) – the analytical property $\lambda_e(-1) > 0$, and (iii) – the Schulgasser inequality (2.14), an infinite set of upper and lower bounds on the effective transport coefficient $\lambda_e(x)$ of two-phase, isotropic composites have been established (Theorem 2) and investigated in detail.

With respect to the corresponding estimations reported in literature (Th. 1), the improvement has been obtained for the case of lower bounds on $\lambda_e(x)$ constructed from an odd number N of coefficients of a power expansion of $\lambda_e(x)$, cf. Fig. 1, Tables 2 and 3. For even N the incorporation of the Schulgasser inequality (2.14) does not provide better bounds in comparison to the approaches neglecting this inequality [7, 8, 22].

As an example of illustration of Theorem 2, the existing and improved bounds on the effective dielectric constant for regular, face-centered arrays of spheres have been evaluated and depicted in Fig. 1, Tabs. 2 and 3. A significant improvement has been observed for $N = 1$. For $N = 2$ the difference between the bounds reported in the literature [20] and in the present paper is relatively small, while for $N = 3$ it is negligible (Fig. 1). Note that the above conclusion is valid for a special geometry of two-phase composite, namely a regular array of spheres. For such a composite and for $n = 4, 6$, from Table 1 we have $E_n/C_n \simeq 1$. In the case of other geometrical structures, when the ratio E_n/C_n satisfies for instance $E_n/C_n < 0.5$ (Tab. 1), it is possible to get much better improvement.

Appendix A

In this Appendix we demonstrate the lemma indispensable for incorporating the Schulgasser inequality (2.14) into the bounds on λ_e.

LEMMA A.1. If a Stieltjes function

$$\frac{\lambda_e(x)}{\lambda_1} = 1 + x\int_0^1 \frac{d\gamma(u)}{1+xu} \tag{A.1}$$

satisfies the relations

$$\frac{\lambda_e(x)}{\lambda_1}\frac{\lambda_e(y)}{\lambda_1} \geq 1, \qquad y = -x/(1+x), \qquad x \in (-1,\infty), \tag{A.2}$$

then Padé approximants $A_N(x)/B_N(x)$ to $\lambda_e(x)/\lambda_1$ obey the inequalities

$$\frac{A_N(x)}{B_N(x))}\frac{A_N(y)}{B_N(y)} \geq 1 \quad (N = 0,1,2...), \quad y = -x/(1+x), \quad x \in (-1,\infty). \tag{A.3}$$

Here $A_N(x)$ and $B_N(x)$ are polynomials determined by recurrence formulae (3.8) – (3.9).

P r o o f. The analytical properties of $A_N(x)/B_N(x)$ $(N = 0,1,2...)$ yield:

$$\text{if} \quad \lim_{x\to -1^+} \frac{A_N(x)}{B_N(x)}\frac{A_N(y)}{B_N(y)} = 1 \quad \text{then} \quad \frac{A_N(x)}{B_N(x)}\frac{A_N(y)}{B_N(y)} \geq 1 \quad \text{in } x \in (-1,\infty), \tag{A.4}$$

where $y = -x/(x+1)$. Hence of interest is the inequality (A.3) taken for $x \to -1^+$. On the basis of Theorem 1 we have:

(i) if N is odd, then

$$\frac{A_N(-1^+)}{B_N(-1^+)} \geq \frac{\lambda_e(-1^+)}{\lambda_1}, \qquad \text{and} \qquad \frac{A_N(\infty)}{B_N(\infty)} \geq \frac{\lambda_e(\infty)}{\lambda_1}, \quad \text{if} \quad x \geq 0. \tag{A.5}$$

(ii) If N is even, then

$$\begin{aligned} &\frac{A_N(-1^+)}{B_N(-1^+)} \geq \frac{\lambda_e(-1^+)}{\lambda_1}, \qquad && \text{if} \quad -1 \leq x \leq 0, \\ &\frac{A_N(\infty)}{B_N(\infty)} \leq \frac{\lambda_e(\infty)}{\lambda_1}, && \text{if} \quad x \geq 0. \end{aligned} \tag{A.6}$$

According to Th. 1 and Th. 15.2 reported in [1], Padé approximants $A_N(-1^+)/B_N(-1^+)$ and $A_N(\infty)/B_N(\infty)$ $(N = 0,2,...)$ are the best bounds for Stieltjes function $\lambda_e(-1^+)/\lambda_1$ and $\lambda_e(\infty)/\lambda_1$ with respect to a given number of coefficients of a power expansion of $\lambda_e(x)/\lambda_1$ at $x = 0$. Hence the relations

$$\frac{A_N(-1^+)}{B_N(-1^+)}\frac{A_N(\infty)}{B_N(\infty)} \geq 1, \qquad N = (0,2,...) \tag{A.7}$$

have to be satisfied. From (A.4) – (A.7) one can easily derive the inequality (A.3). □

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