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New optimal microstructures and restrictions on the attainable Hashin-Shtrikman bounds for multiphase composite materials

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We address the attainability of the Hashin–Shtrikman (HS) bounds for multiphase composite materials. We demonstrate that the HS bounds are not always attainable and give new restrictions on the attainable HS bounds in terms of the conductivities and volume fractions of the constituent phases. New optimal microstructures are also constructed to attain the HS bounds. Combined together, these results allow for precise characterization of the set of effective properties for a wide range of composite materials.

Keywords: composite materials; new optimal microstructures; Hashin–Shtrikman bounds

1. Introduction

A central problem in the study of composite materials is to characterize the effective properties of a composite based on the observed microstructure of the constituent phases. The effective properties, however, depend on the detailed microstructure, and finding the effective properties of a composite requires solutions to partial differential equations which are generally impractical for realistic microstructures. Since the seminal works of Hashin and Shtrikman (HS) [1], much of the attention in this area has focused on the study of optimal bounds on the effective properties with or without constraint on the volume fractions of the constituent phases [2]. A bound is optimal if it is microstructure-independent and attainable for some special microstructure within the prescribed, e.g. volume fraction, constraint. The problem of finding optimal bounds is rather complicated, as is evident from previous efforts in solving optimal bounds for various effective properties of two-phase composites [3–5].

We address the attainability of the HS bounds [1]. It has been well-known since the 1980's that the HS bounds are attainable for the case of two-phase composites [3–6]. Also, it has been known that the HS bounds are not always attainable for composites of three or more phases [7]. More recently, new bounds and new optimal microstructures are found by a number of authors [8–12] for multiphase composites in two dimensions. In particular, Gibiansky and Sigmund [9] constructed microstructures achieving the bulk modulus HS bounds for three-phase

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two-dimension elastic composites; Albin et al. [11] addressed the attainability of HS bounds for multiphase composites and obtained tight restrictions on the attainable HS bounds for multiphase two-dimension conductive composites. However, little is known about the attainability of the HS bounds for multiphase composites in three or higher dimensions, which will be the focus of this article.

This article is to report our progress on the attainability of the HS bounds for multiphase composites in any dimensions. We find a necessary condition and a sufficient condition for the attainable HS bounds. Specialized to two-dimension three-phase isotropic composites, our results recover the results on the attainable HS bounds in [13]; specialized to three-phase composites, our results imply a necessary and sufficient condition for the attainable HS bounds. For four and more phases composites, there is however a gap between the necessary condition and the sufficient condition. In two dimensions the improved bounds given by Nesi [8], Albin et al. [13] and Cherkaev [12] may be useful for clarifying the origin of this gap.

2. New restrictions on the attainable HS bounds

We consider $(N+1)$-phase $(N \geq 2)$ composites of isotropic phases of conductivities $k_0, k_1, \ldots, k_N$ $(0 < k_0 < k_1 < \ldots < k_N)$, volume fractions $\theta_0, \theta_1, \ldots, \theta_N \in (0, 1)$, and assume that the effective conductivity of the composite is isotropic and given by k_e. Denote by

$$\triangle c_i = \frac{nk_0}{k_i - k_0} \qquad (i = e, 1, \ldots, N), \tag{1}$$

where $n \geq 2$ is the dimension of the space. Then the lower HS bound can be expressed as [2,14]

$$k_e \geq k_0 + \frac{nk_0}{\triangle c_{\mathrm{HS}}} \Longleftrightarrow \triangle c_e \leq \triangle c_{\mathrm{HS}} := \frac{1}{\Gamma} - 1, \tag{2}$$

where

$$\Gamma = \sum_{i=1}^{N} \frac{\theta_i}{1 + \triangle c_i}.$$

Further, in [14] it has been shown that the lower HS bound is attainable, i.e. the inequalities in (2) hold as equalities if and only if a periodic solution to

$$\Delta\xi = p_0 \chi_{\Omega_0} + \cdots + p_N \chi_{\Omega_N} \text{ on } \quad Y, \tag{3}$$

satisfies the following overdetermined conditions ($\mathbf{I} \in IR^{n\times n}$ is the identity matrix):

$$\nabla\nabla\xi = \frac{p_i}{n}\mathbf{I} \text{ on } \quad \Omega_i, \ i = 1, \ldots, N. \tag{4}$$

In (3) and (4), $Y = (0, 1)^n$ is the unit cell, $\Omega_i \subset Y$ with volume$(\Omega_i) = \theta_i$ $(i = 0, 1, \ldots, N)$, subdividing the unit cell Y, is the domain occupied by the ith phase, χ_{Ω_i}, equal to one on Ω_i and zero otherwise, is the characteristic function of Ω_i, and constants $p_0, \ldots, p_N$, satisfying $\sum_{i=0}^{N} \theta_i p_i = 0$, are given by

$$p_0 = 1, \quad p_i = \frac{\triangle c_i - \triangle c_{\mathrm{HS}}}{1 + \triangle c_i} \quad \text{if } i \neq 0. \tag{5}$$

Note that the particular unit cell $Y=(0,1)^n$ is immaterial since, by scaling, cutting, and patching a given size composite of an effective conductivity k_e, we can construct a periodic composite of the same effective conductivity k_e with any unit cell.

Equations (3) and (4) place strong restriction on the attaining microstructure, i.e. the domains $(\Omega_0, \ldots, \Omega_N)$. Moreover, it is not always possible to find such a microstructure for given volume fractions θ_i and constants $p_i\,(i=0,\ldots,N)$. Below we derive a necessary condition such that (3) and (4) admit a periodic solution. To this end, we first notice that (3) and (4) imply

$$\nabla\nabla\xi(\mathbf{x}+) = \frac{p_i}{n}\mathbf{I} + (p_0 - p_i)\mathbf{n}\otimes\mathbf{n} \quad \forall\,\mathbf{x}\in\partial\Omega_i\cap\partial\Omega_0, \tag{6}$$

where $\mathbf{n}$ is the unit normal on $\partial\Omega_0$, and $\mathbf{x}+$ denotes the boundary value approached from the exterior of Ω_i or the interior Ω_0. Let $\mathbf{m}\in I\!R^n$ be a unit vector and $u_{\mathbf{m}}=\mathbf{m}\cdot(\nabla\nabla\xi)\mathbf{m}$. By (3) and (6) we verify

$$\begin{cases} \Delta u_{\mathbf{m}} = 0 & \text{on } \Omega_0, \\ u_{\mathbf{m}} = p_i/n + (p_0 - p_i)(\mathbf{m}\cdot\mathbf{n})^2 & \text{on } \partial\Omega_i\cap\partial\Omega_0. \end{cases}$$

By the maximum principle applied to $u_{\mathbf{m}}$ on Ω_0, we have

$$u_{\mathbf{m}} \geq \lambda_{\min} := \min_{t\in[0,1],\ i\in\{1,..,N\}} \frac{p_i}{n} + (p_0 - p_i)t. \tag{7}$$

By (5) and the fact $\Delta c_{\mathrm{HS}} > \Delta c_N$, we have $p_0 - p_i > 0$, $p_1 > \cdots > p_N$, and hence $\lambda_{\min} = p_N/n < 0$. Further, noticing that det: $I\!R^{n\times n} \to I\!R$ is a null Lagrangian [2,15], by the divergence theorem we obtain that for any $\mathbf{M}\in I\!R^{n\times n}$,

$$\det(\mathbf{M}) = ⨍_Y \det(\nabla\nabla\xi + \mathbf{M}). \tag{8}$$

Here and subsequently, $⨍_V = 1/\mathrm{volume}(V)\int_V$ denotes the average value of the integrand over V. Replacing $\mathbf{M}$ by $-\lambda_{\min}\,\mathbf{I}$ in (8) and noticing that $\nabla\nabla\xi - \lambda_{\min}\,\mathbf{I}$ is positive semi-definite on Ω_0, we have

$$\det(-\lambda_{\min}\mathbf{I}) \geq \sum_{i=1}^{N} \theta_i ⨍_{\Omega_i} \det(\nabla\nabla\xi - \lambda_{\min}\mathbf{I}),$$

which, by (4), can be written as

$$\sum_{i=1}^{N} \theta_i \left(1 - \frac{p_i}{p_N}\right)^n \leq 1. \tag{9}$$

Inserting (5) and (2) into the above inequality, we obtain

$$\left[\theta_0 + \sum_{i=1}^{N-1} \theta_i \frac{\Delta c_i - \Delta c_N}{1+\Delta c_i}\right]^{-n} \sum_{i=1}^{N-1} \theta_i \left[\frac{\Delta c_i - \Delta c_N}{1+\Delta c_i}\right]^n \leq 1, \tag{10}$$

which is a necessary condition for the lower HS bound (2) to be attainable.

3. New optimal microstructures

For multiphase composites, the HS bounds are not always attainable for given conductivities and volume fractions of the constituent phases. To find sufficient conditions for the attainable HS bounds, we now construct a new class of microstructures such that the inequalities (2) hold as equalities. For two-phase composites, optimal microstructures include Hashin's construction of coated spheres [16], Milton's construction of coated ellipsoids [17], multi-rank laminates [12,18], Vigdergauz microstructure [19,20] and recently found periodic E-inclusions [21]. The interested reader is referred to the monograph [22] and references therein for a comprehensive description of various optimal microstructures and optimal conditions.

We remark that the construction of optimal microstructures follows from the optimal fields (3)–(4) and it is well-known optimal microstructures are non-unique even in the sense of gradient Young measure. By manipulating exactly solvable microstructures, such as multi-rank laminates, coated spheres, multi-coated spheres [23], coated ellipsoids, Vigdergauz microstructures, and periodic E-inclusions, we can find many optimal microstructures attaining the HS bounds. However, there appears no systematic method to characterize all of the attainable regimes apart from trial and error. For two-dimension and three-phase composites, Gibiansky and Sigmund [9] have systematically studied the attainable regimes by combinations of coated sphere and laminates; Albin et al. [11,12] has investigated in detail the attainable regimes by multi-rank laminates. Here our construction is valid for any dimensions and, to some extent, simple. Also, for three-phase composites, our construction is sufficient for all attainable HS bounds. Outside the attainable regime of HS bounds, it is unclear whether our construction is sufficient or not for all attainable regime. For two-dimension three-phase composites, Cherkaev [12] has systematically suggested optimal microstructures in regimes where the HS bounds are unattainable.

We now present the new optimal microstructures for multiphase composites, which consist of two basic building blocks: (1) two-phase periodic composites with periodic E-inclusions [24], and (2) three-phase coated spheres with the coating being two-phase composites of periodic E-inclusion microstructures. Below we describe their properties briefly; more details can be found in [24] and the author's forthcoming publications.

A periodic E-inclusion is a geometric shape which may be regarded as a generalization of ellipsoids. Its relevant optimality property for two-phase composites is described by a shape matrix $\mathbf{Q} \in I\!R^{n\times n}_{\mathrm{sym}}$ and a volume fraction $\theta^b \in (0,1)$. The shape matrix $\mathbf{Q}$ is further required to be symmetric, positive semi-definite and $\mathrm{Tr}(\mathbf{Q}) = 1$. We assume that the periodic E-inclusion is occupied by the phase with conductivity k_b whereas the matrix phase has conductivity k_a and volume fraction $\theta^a = 1 - \theta^b$. An extraordinary property of a periodic E-inclusion is that the effective conductivity tensor, denoted by $\mathbf{A}_e$, of such a periodic two-phase composite is given by the following closed-form formula [25]:

$$\frac{\mathbf{A}_e}{k_a} = \mathbf{I} + \frac{n\theta^b}{\triangle c_b}\mathbf{I} - \theta^a\theta^b\mathbf{Q}\left[\frac{\theta^a \triangle c_b}{n}\mathbf{Q} + \frac{\triangle c_b^2}{n^2}\mathbf{I}\right]^{-1}, \tag{11}$$

where, as in (1), $\Delta c_b = nk_a/(k_b - k_a)$. In particular, if $\mathbf{Q} = \mathbf{I}/n$, $\mathbf{A}_e = k_e\mathbf{I}$ and

$$k_e = k_a\left(1 + \frac{n}{\triangle c_e}\right), \qquad \triangle c_e = \frac{1 - \theta^b + \triangle c_b}{\theta^b}.$$

In terms of the effective property Δc_e, the volume fraction of the b-phase can be expressed as

$$\theta^b = \frac{1 + \triangle c_b}{1 + \triangle c_e}. \tag{12}$$

The second building block, illustrated in Figure 1, is a three-phase coated sphere: the core sphere has radius R_1 and conductivity k_β; the spherical shell between $r \in [R_1, R_2]$ is occupied by two-phase composites of conductivities k_α and k_γ and the microstructure of the two-phase composite is periodic E-inclusions with the γ-phase occupying the inclusions and the α-phase the matrix. The local shape matrix and local volume fraction of the periodic E-inclusions are given by

$$\mathbf{Q}(\mathbf{x}) = \frac{1}{n-1}[\mathbf{I} - \mathbf{e}_r \otimes \mathbf{e}_r], \quad \rho(r) = 1 - \frac{\eta}{r}, \tag{13}$$

where $\eta = \frac{(n-1)(\triangle c_\beta - \triangle c_\gamma)R_1}{n(1+\triangle c_\beta)}$, $r = |\mathbf{x}|$, $\mathbf{e}_r = \frac{\mathbf{x}}{r}$, and, as in (1), $\Delta c_\gamma = nk_\alpha/(k_\gamma - k_\alpha)$, $\Delta c_\beta = nk_\alpha/(k_\beta - k_\alpha)$. The volume fractions of the α, β, γ-phases within the coated sphere are denoted by $\theta^\alpha, \theta^\beta, \theta^\gamma$, respectively. Clearly,

$$\theta^\beta = \left(\frac{R_1}{R_2}\right)^n, \quad \theta^\alpha + \theta^\beta + \theta^\gamma = 1. \tag{14}$$

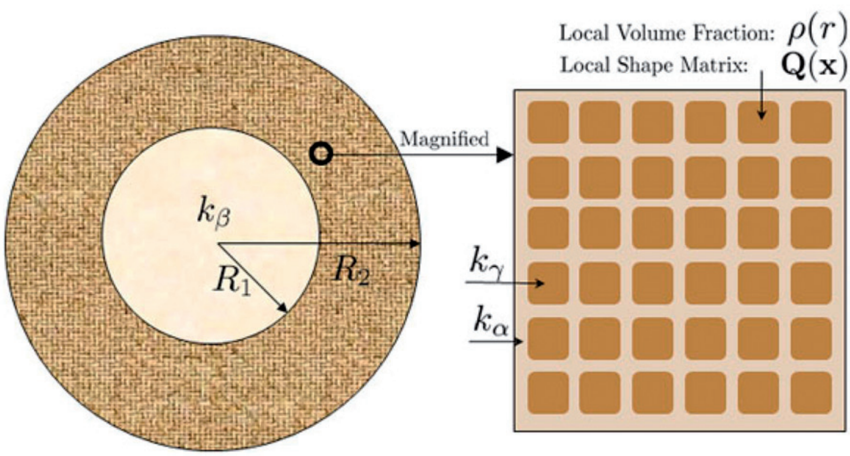

Figure 1. A three-phase coated sphere: the core sphere is occupied by phase-β; the shell is occupied by two-phase composites of phases α, γ. The microstructure of the composite is a periodic E-inclusion with local shape matrix $\mathbf{Q}(\mathbf{x})$ and local volume fraction $\rho(r)$.

Embedding the coated sphere inside an ambient medium of conductivity k_e, from (11) we see that the conductivity tensor on the entire space is given by

$$\mathbf{A}(\mathbf{x}) = \begin{cases} k_\beta \mathbf{I} & \text{if } r < R_1, \\ k_\nu \mathbf{e}_r \otimes \mathbf{e}_r + k_\tau[\mathbf{I} - \mathbf{e}_r \otimes \mathbf{e}_r] & \text{if } R_1 < r < R_2, \\ k_e \mathbf{I} & \text{if } r > R_2, \end{cases} \tag{15}$$

where $k_\nu = k_\alpha(1 + \frac{n\rho}{\triangle c_\gamma})$ and $k_\tau = k_\alpha\{1 + \frac{n\rho}{\frac{n(1-\rho)}{n-1} + \triangle c_\gamma}\}$. From symmetry, one may see that the coated sphere would be a neutral inclusion [26] if the ambient medium is chosen appropriately. Indeed, if

$$k_e = k_\alpha \frac{\triangle c_\gamma + n - (n-1)\sqrt[n]{\theta^\beta}\,\frac{\triangle c_\beta - \triangle c_\gamma}{1 + \triangle c_\beta}}{\triangle c_\gamma + \sqrt[n]{\theta^\beta}\,\frac{\triangle c_\beta - \triangle c_\gamma}{1 + \triangle c_\beta}}, \tag{16}$$

the solution to the following boundary value problem for a far applied electric field $\mathbf{e} \in I\!R^n$,

$$\begin{cases} \operatorname{div}[\mathbf{A}(\mathbf{x})\nabla\varphi] = 0 & \text{on } I\!R^n, \\ -\nabla\varphi = \mathbf{e} & \text{as } |\mathbf{x}| \to +\infty, \end{cases}$$

is given by $\varphi = -\mathbf{e}\cdot\nabla u$,

$$u(r) = \begin{cases} \frac{1}{2} a_\beta r^2 + d_1 & \text{if } r < R_1, \\ a_\gamma\Big[\frac{1}{2} r^2 + r R_1 \frac{\triangle c_\beta - \triangle c_\gamma}{1 + \triangle c_\beta}\Big] & \text{if } r \in [R_1, R_2], \\ \frac{1}{2} r^2 + d_2 & \text{if } r > R_2, \end{cases}$$

and satisfies that $\nabla\varphi = -\mathbf{e}$ if $r > R_2$, i.e. the electric field outside the coated sphere is undisturbed by the presence of the inhomogeneous coated sphere. In the above equation, $a_\gamma = \big[1 + \frac{R_1(\triangle c_\beta - \triangle c_\gamma)}{R_2 \triangle c_\gamma (1 + \triangle c_\beta)}\big]^{-1}$, $a_\beta = \frac{1 + 1/\triangle c_\gamma}{1 + 1/\triangle c_\beta} a_\gamma$, and constants d_1, d_2 are such that $u(r)$ is continuous at $r = R_1$ and R_2. Replacing the ambient medium by scaled copies of the coated sphere, we obtain a composite of the α, β, γ-phases and, from the neutrality of the coated spheres, find that the composite has an effective conductivity k_e. This construction is the well-known Hashin's construction of coated spheres [16].

For future convenience, we express the volume fractions of β, γ-phases in terms of the effective property Δc_e. By (16) we find

$$\theta^\beta = \Big[\frac{(\triangle c_e - \triangle c_\gamma)(1 + \triangle c_\beta)}{(1 + \triangle c_e)(\triangle c_\beta - \triangle c_\gamma)}\Big]^n, \tag{17}$$

and, by the second of (13), the volume fractions θ^α, θ^γ satisfy

$$\frac{\theta^\alpha}{\theta^\alpha + \theta^\gamma} = \frac{n}{R_2^n - R_1^n} \int_{R_1}^{R_2} r^{n-1}(1 - \rho(r))\mathrm{d}r,$$

which, together with (14), implies

$$\theta^\gamma = 1 - \frac{\triangle c_\beta - \triangle c_\gamma}{1 + \triangle c_\beta}\sqrt[n]{\theta^\beta} - \frac{1 + \triangle c_\gamma}{1 + \triangle c_\beta}\theta^\beta. \tag{18}$$

We now construct optimal microstructures attaining the lower HS bounds (2) for $(N+1)$-phase composites. Since $\Delta c_{\rm HS} > \Delta c_N$, we have $\Delta c_{\rm HS} \in [\Delta c_{m+1}, \Delta c_m)$ for some $m \in \{N-1, \ldots, 1, 0\}$. ($\Delta c_m = +\infty$ if $m=0$) That is, by (2),

$$\frac{1}{1+\Delta c_m} < \Gamma \leq \frac{1}{1+\Delta c_{m+1}}. \tag{19}$$

As illustrated in Figure 2, we consider an $(N+1)$-phase composite divided into N parts, each of which has the same effective conductivity specified by $\Delta c_e = \Delta c_{\rm HS}$. The microstructures of various parts are as follows: the ith part ($i=1,\ldots,m$) consists of Hashin's construction of the three-phase coated spheres of the 0, i, N-phases as described above; the jth part ($j=m+1,\ldots,N$) consists of a two-phase composite of 0, j-phases whose microstructure is a periodic E-inclusion with the shape matrix $\mathbf{Q}=\mathbf{I}/n$. The volume fractions of the ith part, denoted by θ_i' ($i=1,\ldots,N$), are given by

$$\begin{aligned} \theta_i' &= \frac{\theta_i}{\theta_i^\beta} && \text{if } i = 1,\ldots,m, \\ \theta_j' &= \frac{\theta_j}{\theta_j^b} && \text{if } j = m+1,\ldots,N-1, \\ \theta_N' &= \frac{1}{\theta_N^b}\left[\theta_N - \sum_{i=1}^{m} \theta_i^\gamma \theta_i'\right], \end{aligned} \tag{20}$$

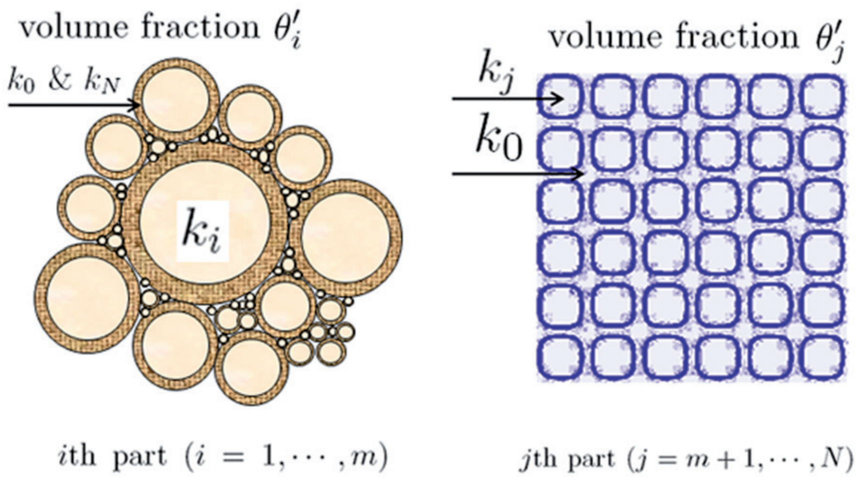

Figure 2. The microstructure of the overall composite: the ith-part ($i=1,\ldots,m$) consists of the Hashin's construction of the three-phase coated spheres of the 0, i, N-phases as described above; the jth-part ($j=m+1,\ldots,N$) consists of a two-phase composite of 0, j-phases whose microstructure is a periodic E-inclusion with the shape matrix $\mathbf{Q}=\mathbf{I}/n$. Their volume fractions are denoted by θ_i' ($i=1,\ldots,N$), $\theta_i' \geq 0$, and $\sum_{i=1}^N \theta_i' = 1$.

where constants θ_i^β, θ_i^γ and θ_j^b are given by the right-hand sides of (17), (18) and (12) with γ replaced by N, β replaced by i, Δc_e replaced by Δc_{HS}, and b replaced by j:

$$\begin{aligned} \theta_i^\beta &= \Big[\frac{1-(1+\triangle c_N)\Gamma}{\frac{\triangle c_i - \triangle c_N}{1+\triangle c_i}}\Big]^n, \\ \theta_i^\gamma &= (1+\triangle c_N)\Gamma - \frac{1+\triangle c_N}{1+\triangle c_i}\theta_i^\beta, \\ \theta_j^b &= (1+\triangle c_j)\Gamma. \end{aligned} \tag{21}$$

Within the ith part ($i=1,\ldots,m$), the volume fractions of the 0, i, N-phases are such that this part has an effective conductivity specified by Δc_{HS}; so do the volume fractions of the 0, j-phases within the jth-part ($j=m+1,\ldots,N$). From (12), (17) and (18), we see that the volume fraction of the ith phase within the overall composite, denoted by $\hat{\theta}_i$, is given by

$$\begin{aligned} \hat{\theta}_i &= \theta'_i\theta_i^\beta = \theta_i \qquad (i=1,\ldots,m), \\ \hat{\theta}_j &= \theta'_j\theta_j^b = \theta_j \qquad (j=m+1,\ldots,N-1), \\ \hat{\theta}_N &= \sum_{i=1}^m \theta'_i\theta_i^\gamma + \theta'_N\theta_N^b = \theta_N. \end{aligned}$$

Therefore, the volume fraction of each phase in the constructed composite is consistent with the prescribed volume fraction. Further, the conductivity of the constructed composite achieves the lower HS bound since each part of it has the conductivity specified by Δc_{HS}. Of course, the above construction does not make any sense unless the volume fractions θ'_i satisfy the obvious constraints: $\theta'_i \geq 0$ ($i=1,\ldots,N$) and $\sum_{i=1}^N \theta'_i = 1$. Indeed, by (21) and direct calculations we verify that

$$\sum_{i=1}^N \theta'_i = \sum_{q=m+1}^{N-1} \frac{\theta_j}{\theta_j^b} + \frac{1}{\theta_N^b}\left[\theta_N + \sum_{p=1}^m \frac{\theta_i}{\theta_i^\beta}(\theta_N^b - \theta_i^\gamma)\right] = 1,$$

and $\theta'_i \geq 0$ $\forall i=1,\ldots,N-1$. Requiring $\theta'_N = 1 - \theta'_1 - \cdots - \theta'_{N-1} \geq 0$ gives rise to

$$\Big[\theta_0 + \sum_{i=1}^{N-1}\theta_i\frac{\Delta c_i - \Delta c_N}{1+\Delta c_i}\Big]^{-n}\sum_{i=1}^m \theta_i\Big[\frac{\Delta c_i - \Delta c_N}{1+\Delta c_i}\Big]^n + \Big[\sum_{i=1}^N \frac{\theta_i}{1+\Delta c_i}\Big]^{-1}\sum_{j=m+1}^{N-1}\frac{\theta_j}{1+\Delta c_j} \leq 1, \tag{22}$$

which, together with (19), forms a sufficient condition such that the lower HS bound (2) is attainable (by the constructed composite). We remark that (22) is trivially satisfied if $m=0$, and hence the lower HS bound (2) is attainable if $\Delta c_{\mathrm{HS}} \geq \Delta c_1$. This has been shown in [23,27].

The necessary condition (10) and the sufficient condition (19), (22) are explicit in terms of conductivities and volume fractions of the constituent phases. In particular, for three-phase composites ($N=2$), we find the necessary condition (10) guarantees (19), (22) for some $m \in \{N-1,\ldots,0\}$, and hence is sufficient as well. For $N>2$, the attainability of the lower HS bounds can be easily studied for specified conductivities and volume fractions and examples are shown in Figure 3. In these examples,

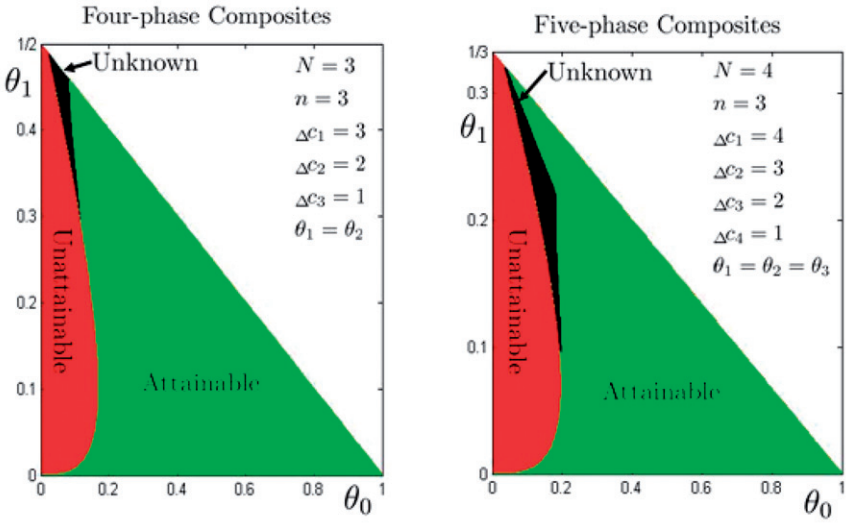

Figure 3. The attainability of the lower HS bounds. Various parameters are specified on the top-right of each panel; all intermediate phases have the same volume fraction. The (θ_0, θ_1)-plane are divided into three regions where the attainability of the lower HS bound (2) is true, false, and unknown, respectively.

we assume the volume fractions of all intermediate phase are equal, i.e. $\theta_1 = \ldots = \theta_{N-1}$ and material properties $\Delta c_1, \ldots, \Delta c_N$ are given on the top-right of each panel. Vary (θ_0, θ_1). The region where (10) violated implies that the lower HS bound is unattainable and is labeled as "Unattainable", while the region where (19) and (22) satisfied for some $m \in \{0, \ldots, N-1\}$ means that the lower HS bound is attainable and is labeled as "Attainable". The attainability of the lower HS bound is unknown for the remaining region, labeled as "Unknown".

4. Summary and conclusion

The problem of the exact description of the set of effective conductivities of multiphase composite materials is addressed by deriving necessary conditions and sufficient conditions for the best known bounds – the HS's bounds – to be attainable. The necessary condition is obtained by using a null Lagrangian; the sufficient condition is achieved by constructing new optimal microstructures. Specialized to three-phase composites, these conditions yield a necessary and sufficient condition. For more general situations, parametric studies may be easily performed and examples are provided.

We remark that similar necessary conditions and sufficient conditions can be derived in parallel for the attainability of the upper HS bounds. Combined together, these attainability conditions allow for the exact description of the set of effective conductivities of multiphase composite materials when the conductivities and volume fractions of the constituent phases are such that the lower and the upper HS bounds are both attainable. If the lower or the upper HS bound is unattainable, the exact description of the set of effective conductivities demands better bounds than the 50-year-old HS bounds. Finally, the conditions presented here apparently apply to physical properties, such as permittivity of dielectric materials, permeability of para/dia-magnetic materials, and bulk modulus of elastic materials. The constructed microstructure also allows for closed-form solutions to the governing equations of composite materials where the inclusion phases are nonlinear, which will be useful for a wide range of materials modeling and optimization.

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