Hashin-Shtrikman bounds and their attainability for multiphase composites

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Abstract

In this paper we consider the problem of characterizing the set of the effective tensors of multiphase composites, including those of conductive materials and elastic materials. We first present a novel derivation of the Hashin-Shtrikman (HS) bounds for multiphase composites and the associated attainment condition. The attainment condition asserts that the HS bound is attainable if and only if there exists a second gradient field that is constant in all but the matrix phases. By restricting and constructing such second gradient fields, we obtain a series of sufficient conditions such that the HS bounds are attainable or unattainable. These attainability and unattainability results appear new for a generic situation. For special situations, our attainability and unattainability results recover the results of [26, 32, 11, 1].

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1. Introduction

Since the seminal works of Hashin & Shtrikman [15, 17], finding optimal bounds on the effective properties, with or without restriction on the volume fractions, has become the central problem in the theory of composites [28]. The usual approach of finding optimal bounds consists of two steps: the first is to derive a microstructure-independent bound and the second is to study if this bound is attainable and if so, by what kind of microstructures. The optimal bounds can be categorized into two types according to the methods of derivation: the Hashin-Shtrikman (HS) bounds and the translation bounds [34, 24, 27]. The attaining microstructures include coated spheres and ellipsoids [14, 26], multi-coated spheres [25] and multi-rank laminations. By this approach, the G-closure problem [24, 36] for two-phase well-ordered conductive materials has been resolved [29, 12]. However, for multiphase composites, little is known about the attainability of the HS bounds.

In this paper we address the attainability of the HS bounds for general multiphase composites. We also present a new derivation of the HS bounds, which is motivated by the observation that the gradient field associated with an optimal microstructure is often the second gradient of a scalar potential [23]. Similar argument has been used by Silvestre [33]. The advantage of the new derivation is that it provides a necessary and sufficient attainment condition for the optimal microstructures and the associated gradient fields. In a periodic setting, the attainment condition is simply that the second gradient of the scalar potential is constant in all but the matrix phases, see (2.22). This attainment condition forms an overdetermined problem (2.22) for the microstructure and might seem too restrictive at the first sight. Nevertheless, using variational inequalities [22, 10] we can show the existence of these optimal periodic microstructures which we call *periodic E-inclusions* [23]. The results in Section 2 can then be roughly stated as a periodic microstructure attains the HS bound if, and only if the microstructure is a corresponding periodic E-inclusion (cf., Theorem 2.1). Therefore, from the attainability of the HS bound for one particular set of materials, we can infer the existence of a corresponding periodic E-inclusion, and hence the attainability of the HS bounds for many different sets of materials (cf., Corollary 2.2).

As far as the attainability of HS bounds is concerned, it suffices to study the existence of periodic E-inclusions. Gradient Young measures and quasiconvex functions have proven to be useful in describing, constructing and restricting microstructures [35, 5]. For an excellent introduction to these concepts, the reader is referred to the textbook of Evans [9]. Based on the gradient field of a periodic E-inclusion, we define a particular form of gradient Young measures as *sequential E-inclusions* (cf., (3.1)). From the basic relation between gradient Young measures and quasiconvex functions [20, 21], we can restrict sequential E-inclusions (and hence attainable HS bounds) by quasiconex functions. More restrictions on periodic E-inclusions can be found by the maximum principle. From these restrictions on sequential E-inclusions, we obtain sufficient conditions for unattainable HS bounds.

To construct optimal microstructures for multiphase composites, we may take elementary microstructures, e.g., simple laminates and coated spheres , as building blocks and construct multi-rank laminations and multi-coated spheres [26, 12, 11, 1]. This procedure is delicate, requiring tedious calculations. Taking the advantage of convexity properties of gradient Young measures (cf., Theorem 3.1), we focus on the optimal gradient fields and construct a class of optimal microstructures that can attain the HS bounds. From these optimal microstructures, we obtain sufficient conditions for attainable HS bounds.

We remark that the attainability and unattainability results in this paper apply broadly to various physical properties, and the individual phases and the composites are not necessarily isotropic, though some symmetries on the "softest" or "stiffest" phase are required for deriving the HS bounds. Further, the HS bounds in their classic form (A.1) are well understood, see e.g. [39, 26, 3, 4]. The dual bounds (2.18) are often referred to as the translation bounds. Mentions should be made of the works of Grabovsky [13] who, based on the translation method, has derived attainment conditions for two-phase composites which are closely related to ours, also see [1, 2] for two dimensional three-phase composites and [33] for cross-property bounds.

The paper is organized as follows. In Section 2 we derive the HS bounds for multiphase composites and establish the equivalence between the attainability of the HS bounds and the existence of a corresponding periodic E-inclusion. In Section 3 we focus on the optimal gradient fields, introduce the concept of sequential E-inclusions, find restrictions on sequential E-inclusions, and construct a class of sequential E-inclusions. In Section 4, we find a series of sufficient conditions for attainable HS bounds and unattainable HS bounds. Finally, in Section 5 we summarize our results and discuss the directions of generalization.

For future convenience, we introduce some notation. For two symmetric linear mappings $\mathbf{L}_1, \mathbf{L}_2 : \mathbb{R}^{m\times n} \to \mathbb{R}^{m\times n}$, we write $\mathbf{L}_1 \geq (\leq) \mathbf{L}_2$ if $\mathbf{L}_1 - \mathbf{L}_2$ is positive (negative) semi-definite and denote by $\mathcal{N}(\,\cdot\,)$ $(\mathcal{R}(\,\cdot\,))$ the null (range) space of a symmetric linear mapping $(\,\cdot\,)$. For any $\mathbf{F}_1, \mathbf{F}_2 \in \mathbb{R}^{m\times n}$, the inner product is defined as $\mathbf{F}_1 \cdot \mathbf{F}_2 = \mathrm{Tr}(\mathbf{F}_1\mathbf{F}_2^T)$. If $m = n$, we denote by $\mathbb{R}^{n\times n}_{sym}$ the symmetric matrices in $\mathbb{R}^{n\times n}$. We follow the conventions $1/\infty = 0$, $1/0 = \infty$ and interpret the inverse $\mathbf{L}^{-1}$ of a symmetric positive semi-definite linear mapping $\mathbf{L} : \mathbb{R}^{m\times n} \to \mathbb{R}^{m\times n}$ as

$$\mathbf{F}_2 \cdot \mathbf{L}^{-1}\mathbf{F}_2 = \sup_{\mathbf{F}_1 \in \mathbb{R}^{m\times n}} \{2\mathbf{F}_2 \cdot \mathbf{F}_1 - \mathbf{F}_1 \cdot \mathbf{L}\mathbf{F}_1\} \qquad \forall\, \mathbf{F}_2 \in \mathbb{R}^{m\times n}. \tag{1.1}$$

Further, let $Y = (0,1)^n$ be a unit cell. For a function $f : \mathbb{R}^n \to \mathbb{R}$, f being periodic on Y means $f(\mathbf{x}+\mathbf{r}) = f(\mathbf{x})\ \forall\, \mathbf{x} \in \mathbb{R}^n$ and $\mathbf{r} \in \mathbb{Z}^n$. Denote by $L^2_{per}(Y) = \{f \mid f : \mathbb{R}^n \to \mathbb{R}$ is periodic on Y and $\fint_Y |f|^2 < +\infty\}$, and $W^{k,p}_{per}(Y, \mathbb{R}^m)$ the set

$$\{\mathbf{u} \mid \mathbf{u} : \mathbb{R}^n \to \mathbb{R}^m \text{ is periodic on Y} \quad \text{and} \quad \int_Y [\sum_{|\alpha|\leq k} |D^\alpha \mathbf{u}|^p < +\infty\}.$$

2. Hashin-Shtrikman bounds and their attainment conditions

Let Ω_i $(i = 0, \cdots, N)$ with $|\partial\Omega_i| = 0$ be a measurable disjoint subdivision of the unit cell $Y = (0,1)^n$ and $\theta_i = |\Omega_i|/|Y| \neq 0$ be their volume fractions. Without loss of generality, we assume $\Omega_1, \cdots, \Omega_N$ are closed and Ω_0 is open in Y, and refer to $\mathcal{O} = (\Omega_1, \cdots, \Omega_N)$ as the microstructure of the composite. Consider a periodic $(N+1)$-phase composite

$$\mathbf{L}(\mathbf{x}, \mathcal{O}) = \mathbf{L}_i \qquad \text{if } \mathbf{x} \in \Omega_i \qquad (i = 0, 1, \cdots, N), \tag{2.1}$$

where $\mathbf{L}_i : \mathbb{R}^{m\times n} \to \mathbb{R}^{m\times n}$ $(i = 0, \cdots, N)$ is either a positive definite symmetric tensor or an elasticity tensor with the usual symmetries. These tensors describe the materials properties of the constituent phases which include but are not limited to conductive and elastic properties.

From the homogenization theory [37, 18, 8], the effective tensor $\mathbf{L}^e(\mathcal{O})$ of the periodic composite (2.1) is given by

$$\begin{aligned}\mathbf{F} \cdot \mathbf{L}^e(\mathcal{O})\mathbf{F} &= \min_{\mathbf{v} \in W^{1,2}_{per}(Y,\mathbb{R}^m)} \fint_Y (\nabla\mathbf{v} + \mathbf{F}) \cdot \mathbf{L}(\mathbf{x}, \mathcal{O})(\nabla\mathbf{v} + \mathbf{F}) \\ &= \min_{f \in L^2_{per}(Y)} \min_{\nabla\cdot\mathbf{v} = f} \fint_Y (\nabla\mathbf{v} + \mathbf{F}) \cdot \mathbf{L}(\mathbf{x}, \mathcal{O})(\nabla\mathbf{v} + \mathbf{F}) \qquad \forall\, \mathbf{F} \in \mathbb{R}^{m\times n}. \end{aligned} \tag{2.2}$$

Here and subsequently, $\fint_V \cdot = \frac{1}{\text{volume}(V)}\int_V \cdot$ denotes the average value of the integrand in region V. A minimizer of the right hand side, which is unique within an additive constant and denoted by $\mathbf{u} \in W^{1,2}_{per}(Y, \mathbb{R}^m)$, solves the following equation

$$\begin{cases} \operatorname{div}\big[\mathbf{L}(\mathbf{x}, \mathcal{O})(\nabla \mathbf{u} + \mathbf{F})\big] = 0 & \text{on } Y, \\ \text{periodic boundary conditions} & \text{on } \partial Y. \end{cases} \tag{2.3}$$

A problem of critical importance is to calculate the effective properties of a composite based on the observed microstructure $\mathcal{O}$. The effective tensor $\mathbf{L}^e(\mathcal{O})$, however, depends on the detailed microstructure of the composite in a way that is difficult to characterize. Therefore, it is often more useful to find sharp bounds on the effective tensor in terms of simple features of the microstructure, e.g., volume fractions, than to calculate the exact effective tensor. Such bounds include the well-known Voigt and Reuss bounds [38, 31]

$$\mathbf{H}_\Theta \le \mathbf{L}^e \le \mathbf{L}_\Theta, \tag{2.4}$$

where $\mathbf{L}_\Theta = \sum_{i=0}^N \theta_i \mathbf{L}_i$ ($\mathbf{H}_\Theta = [\sum_{i=0}^N \theta_i \mathbf{L}_i^{-1}]^{-1}$) is the arithmetic (harmonic) mean. Tighter bounds are obtained by Hashin & Shtrikman [15, 16].

Below we present a novel derivation of the HS bounds. Let $\mathbf{I}$ be the identity matrix in $\mathbb{R}^{n\times n}$ and, for simplicity, assume that $m = n$,

$$\begin{cases} \text{either } \mathbf{L}_i \ge \mathbf{L}_c \text{ or } \mathbf{L}_i \le \mathbf{L}_c \ \ \forall\, i = 0, \cdots, N, \\ \mathcal{R}(\mathbf{L}_0 - \mathbf{L}_c) \subset \{x\mathbf{I} :\ x \in \mathbb{R}\},\ \ \mathcal{R}(\mathbf{L}_i - \mathbf{L}_c) \supset \mathbb{R}^{n\times n}_{sym} \ \ \forall\, i = 1, 2, \cdots, N, \end{cases} \tag{2.5}$$

where, as the original Hashin and Shtrikman's derivation, a comparison tensor $\mathbf{L}_c$

$$\begin{cases} (\mathbf{L}_c)_{piqj} = \mu_1^c \delta_{ij}\delta_{pq} + \mu_2^c \delta_{pj}\delta_{iq} + \lambda^c \delta_{ip}\delta_{jq}, \\ \mu_1^c > 0, \qquad k_c = \lambda^c + \mu_1^c + \mu_2^c > 0 \end{cases} \tag{2.6}$$

has been chosen. Note that the first line in (2.5) facilitates the following algebraic estimate (2.10) whereas the second implies that for some $a \in \mathbb{R}$, $(\mathbf{L}_0 - \mathbf{L}_c)\mathbf{F} = a\mathbf{I} \Leftrightarrow \mathrm{Tr}(\mathbf{F}) = a\mathbf{I}\cdot(\mathbf{L}_0 - \mathbf{L}_c)^{-1}\mathbf{I}$ and that $\mathbf{L}_i - \mathbf{L}_c$ is invertible on $\mathbb{R}^{n\times n}_{sym}$ for $i = 1, \cdots, N$. The usefulness of these conditions will be clear later. We further denote by

$$\triangle c_i = \mathbf{I}\cdot(\mathbf{L}_i - \mathbf{L}_c)^{-1}\mathbf{I}, \quad \gamma = \sum_{i=0}^N \frac{\theta_i}{1/k_c + \triangle c_i}, \quad \triangle c_* = \frac{1}{\gamma} - \frac{1}{k_c}. \tag{2.7}$$

For the lower HS bound, we assume that

$$\mathbf{L}_i \ge \mathbf{L}_c \qquad \forall\, i = 0, 1, \cdots, N. \tag{2.8}$$

Then the integral on the right hand side (r.h.s.) of (2.2) is bounded from below as

$$\int_Y \big\{(\nabla\mathbf{v} + \mathbf{F})\cdot[\mathbf{L}(\mathbf{x}, \mathcal{O}) - \mathbf{L}_c](\nabla\mathbf{v} + \mathbf{F}) + (\nabla\mathbf{v} + \mathbf{F})\cdot\mathbf{L}_c(\nabla\mathbf{v} + \mathbf{F})\big\}$$
$$\ge \sum_{i=0}^N \int_{\Omega_i} \frac{1}{\triangle c_i}(f + \mathrm{Tr}\mathbf{F})^2 + k_c \int_Y f^2 + \mathbf{F}\cdot\mathbf{L}_c\mathbf{F}, \tag{2.9}$$

where, for the first term on the left hand side (l.h.s.) of (2.9), we have used the algebraic inequality

$$(X+\mathbf{F})\cdot(\mathbf{L}_i-\mathbf{L}_c)(X+\mathbf{F}) \geq \frac{1}{\triangle c_i}[\mathrm{Tr}(X+\mathbf{F})]^2 \qquad \forall\, X\in\mathbb{R}^{n\times n}, \tag{2.10}$$

and for the second term we have used

$$\int_Y \nabla\mathbf{v}\cdot\mathbf{L}_c\nabla\mathbf{v} \geq k_c\int_Y [\mathrm{Tr}(\nabla\mathbf{v})]^2 = k_c\int_Y f^2. \tag{2.11}$$

The above inequality can be conveniently shown by Fourier analysis. Further, we can easily show that inequality (2.10) holds as an equality if and only if

$$(\mathbf{L}_i-\mathbf{L}_c)(X+\mathbf{F}) = \frac{\mathrm{Tr}(X+\mathbf{F})}{\triangle c_i}\mathbf{I} \qquad \forall\, i=0,\cdots,N, \tag{2.12}$$

and inequality (2.11) holds as an equality if and only if there is a scalar potential $\xi\in W^{2,2}_{per}(Y)$ such that

$$\mathbf{v}-⨍_Y \mathbf{v} = -\nabla\xi. \tag{2.13}$$

Similarly, for the upper bound, we assume that

$$\mathbf{L}_i \leq \mathbf{L}_c \qquad \forall\, i=0,1,\cdots,N, \tag{2.14}$$

and hence the inequality (2.10) holds with "$\geq$" replaced by "$\leq$". Then the inner minimum of the r.h.s. of (2.2) can be bounded from above as

$$\begin{aligned}\min_{\nabla\cdot\mathbf{v}=f}\int_Y\cdots \quad &\leq \quad \min_{-\Delta\xi=f}\int_Y \Big\{(-\nabla\nabla\xi+\mathbf{F})\cdot[\mathbf{L}(\mathbf{x},\mathcal{O})-\mathbf{L}_c](-\nabla\nabla\xi+\mathbf{F}) \\ &\qquad\qquad +(-\nabla\nabla\xi+\mathbf{F})\cdot\mathbf{L}_c(-\nabla\nabla\xi+\mathbf{F})\Big\} \\ &\leq \sum_{i=0}^N\int_{\Omega_i}\frac{1}{\triangle c_i}(f+\mathrm{Tr}\mathbf{F})^2 + k_c\int_Y f^2 + \mathbf{F}\cdot\mathbf{L}_c\mathbf{F}. \end{aligned} \tag{2.15}$$

Plugging the r.h.s. of (2.9) or (2.15) into (2.2) and solving the outer minimization problem in (2.2), we find that for any $\mathbf{F}\in\mathbb{R}^{n\times n}$,

$$\min_{f\in L^2_{per}(Y)}\Big\{\sum_{i=0}^N\int_{\Omega_i}\frac{1}{\triangle c_i}(f+\mathrm{Tr}\mathbf{F})^2 + k_c\int_Y f^2\Big\} = (\mathrm{Tr}\mathbf{F})^2/\triangle c_*, \tag{2.16}$$

and the minimizer is unique and given by

$$f(\mathbf{x}) = \mathrm{Tr}(\mathbf{F})\frac{\triangle c_i-\triangle c_*}{\triangle c_*(1+k_c\triangle c_i)} \qquad \text{if } \mathbf{x}\in\Omega_i,\ i=0,1,\cdots,N. \tag{2.17}$$

Noticing the conditions (2.8) and (2.14) and the directions of the inequalities in (2.9) and (2.15), by (2.2) we obtain that for any $\mathbf{F}\in\mathbb{R}^{n\times n}$,

$$\begin{cases}\mathbf{F}\cdot\mathbf{L}^e(\mathcal{O})\mathbf{F} \geq \mathbf{F}\cdot\mathbf{L}_c\mathbf{F}+(\mathrm{Tr}\mathbf{F})^2/\triangle c_* & \text{if } \mathbf{L}(\mathbf{x},\mathcal{O})\geq\mathbf{L}_c, \\ \mathbf{F}\cdot\mathbf{L}^e(\mathcal{O})\mathbf{F} \leq \mathbf{F}\cdot\mathbf{L}_c\mathbf{F}+(\mathrm{Tr}\mathbf{F})^2/\triangle c_* & \text{if } \mathbf{L}(\mathbf{x},\mathcal{O})\leq\mathbf{L}_c.\end{cases} \tag{2.18}$$

We remark that the above bounds are equivalent to the HS bounds in their classic form [15, 16] and can be obtained by using the Hashin-Shtrikman's variational principle [29, 28], see details in the Appendix. The bounds (2.18) are microstructure-independent in the sense that the number $\triangle c_*$ depends only on the materials properties $\mathbf{L}_i$ and the volume fractions θ_i of the microstructure $\mathcal{O}$. Further, we notice that the well-orderedness conditions, i.e., (2.8) and (2.14), are weaker than the well-orderedness conditions $\mathbf{L}(\mathbf{x},\mathcal{O}) \geq$ (or $\leq$) $\mathbf{L}_0$ in the usual derivation of the HS bounds [28]. In the setting of elasticity, the well-orderedness of bulk modulus is not required by the conditions (2.8) or (2.14) and our bounds (2.18) recover the Walpole's bounds on bulk modulus [39].

Subsequently, by the (lower or upper) HS bound (2.18) is attainable for $\mathbf{F} \in \mathbb{R}^{n\times n}$ we mean one of the inequality of (2.18) holds as an equality for some microstructure $\mathcal{O}$. Since only one of the conditions $\mathbf{L}(\mathbf{x},\mathcal{O}) \geq \mathbf{L}_c$ and $\mathbf{L}(\mathbf{x},\mathcal{O}) \leq \mathbf{L}_c$ can be satisfied after $\mathbf{L}_c$ being specified, it will be clear from the context which inequality in (2.18) is under consideration.

We now study the attainment conditions for the microstructure $\mathcal{O}$ such that the HS bounds (2.18) hold as equalities. We first consider the lower HS bound, i.e., the case $\mathbf{L}(\mathbf{x},\mathcal{O}) \geq \mathbf{L}_c$, and assume that for some $\mathbf{F} \in \mathbb{R}^{n\times n}_{sym}$ the first inequality in (2.18) holds as an equality. Let $\mathbf{u}$ be the corresponding solution to (2.3) with zero average on Y. Tracking back our argument, we find that the first inequality in (2.18) holds as an equality implies that ($\mathcal{C}1$) the inequality (2.10) holds as an equality, ($\mathcal{C}2$) the inequality (2.11) holds as an equality, and ($\mathcal{C}3$) $f = \nabla\cdot\mathbf{u}$ is exactly the minimizer given by (2.17). By (2.10) and (2.12), ($\mathcal{C}1$) implies

$$(\mathbf{L}_i - \mathbf{L}_c)(\nabla\mathbf{u} + \mathbf{F}) = \frac{\mathrm{Tr}(\nabla\mathbf{u} + \mathbf{F})}{\triangle c_i}\mathbf{I} \qquad \text{on } \Omega_i,\ i = 0, \cdots, N. \tag{2.19}$$

By (2.11) and (2.13), ($\mathcal{C}2$) implies

$$\mathbf{u} = -\nabla\xi, \tag{2.20}$$

for some scalar function ξ and, finally, by (2.17) ($\mathcal{C}3$) implies

$$\nabla\cdot\mathbf{u} = \mathrm{Tr}(\mathbf{F})\frac{\triangle c_i - \triangle c_*}{\triangle c_*(1 + k_c\triangle c_i)} \qquad \text{if } \mathbf{x}\in\Omega_i,\ i = 0, 1, \cdots, N. \tag{2.21}$$

Conversely, if (2.19), (2.20) and (2.21) are true, we can easily check that the first inequality in (2.18) indeed holds as an equality. Lumping (2.19), (2.20), (2.21) together, by (2.5) we write them as the following overdetermined problem

$$\begin{cases} \Delta\xi = \sum_{i=0}^N p_i\chi_{\Omega_i} & \text{on } Y, \\ \nabla\nabla\xi = \mathbf{Q}_i & \text{on } \Omega_i,\ i = 1, \cdots, N, \\ \text{periodic boundary conditions} & \text{on } \partial Y, \end{cases} \tag{2.22}$$

where the symmetric matrices $\mathbf{Q}_i$ ($i = 1, \cdots, N$) are given by

$$\mathbf{Q}_i = \mathbf{F} - \mathrm{Tr}(\mathbf{F})\frac{(1 + k_c\triangle c_*)}{\triangle c_*(1 + k_c\triangle c_i)}(\mathbf{L}_i - \mathbf{L}_c)^{-1}\mathbf{I}, \tag{2.23}$$

and constants p_i ($i = 0, \cdots, N$), satisfying $\theta_0 p_0 + \sum_{i=1}^N \theta_i p_i = 0$, are given by

$$p_i = \mathrm{Tr}(\mathbf{Q}_i) = \frac{\mathrm{Tr}(\mathbf{F})(\triangle c_* - \triangle c_i)}{\triangle c_*(1 + k_c\triangle c_i)} \quad (i = 0, 1, \cdots, N). \tag{2.24}$$

In particular, the second of (2.22) and (2.23) follow from (2.19), (2.20), and that $\mathbf{L}_i - \mathbf{L}_c$ is invertible on $\mathbb{R}^{n\times n}_{sym}$ for $i = 1, \cdots, N$.

Similar calculations prevail for the attainment of the upper HS bound, i.e., the case $\mathbf{L}(\mathbf{x}, \mathcal{O}) \le \mathbf{L}_c$, which will not be repeated here.

The overdetermined problem (2.22) places strong restrictions on the microstructure $\mathcal{O} = (\Omega_1, \cdots, \Omega_N)$. For its analogy with an ellipsoid and its extremal properties as presented above, we call the collection of domains $(\Omega_1, \cdots, \Omega_N)$ a **periodic E-inclusions** [23]. The important parameters describing the properties of a periodic E-inclusion are the symmetric matrices $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_N)$ and the volume fractions $\Theta = (\theta_1, \cdots, \theta_N)$; they are related with the materials properties and applied average field by (2.23) for an optimal composite attaining the HS bound (2.18).

We summarize below.

Theorem 2.1. *Consider a periodic $(N+1)$-phase composite* (2.1). *Let $\mathbf{L}_c$ be given by* (2.6), *$\triangle c_*$ be given by* (2.7), *and assume $(\mathbf{L}_0, \cdots, \mathbf{L}_N)$ satisfy* (2.5).

(i) (**HS bound**). *The effective tensor of the composite, given by* (2.2), *satisfies the HS bounds* (2.18).

(ii) (**Attainment condition**). *For some $\mathbf{F} \in \mathbb{R}^{n\times n}_{sym}$, the HS bound* (2.18) *is attained by a periodic microstructure $\mathcal{O}$ if, and only if the microstructure $\mathcal{O}$ is the corresponding periodic E-inclusion, i.e., the overdetermined problem* (2.22) *admits a solution $\xi \in W^{2,2}_{per}(Y)$ for $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_N)$ given by* (2.23).

From the above theorem, in particular, the attainment condition, we see that the attainability of the HS bound for an average applied field $\mathbf{F} \in \mathbb{R}^{n\times n}_{sym}$ is equivalent to the existence of the corresponding periodic E-inclusion. In the next section we will study conditions on symmetric matrices $\mathbb{K}$ and volume fractions Θ such that the corresponding periodic E-inclusion exists or does not exists. This is a more generic problem than the attainability of HS bounds since it is independent of materials properties. After obtaining conditions for the existence or non-existence of periodic E-inclusions, by (2.23) we can translate these conditions to conditions on the materials properties $(\mathbf{L}_0, \cdots, \mathbf{L}_N)$, volume fractions Θ and average applied field $\mathbf{F}$ such that the HS bounds (2.18) are attainable or unattainable.

Moreover, we notice that optimal microstructures, e.g., confocal ellipsoids and multi-rank laminations, attain the HS bounds for many composites of different materials. From the viewpoint of equations (2.22) and (2.23), this corresponds to equation (2.23) has many different sets of solutions of $(\mathbf{L}_c, \mathbf{L}_0 \cdots, \mathbf{L}_N)$ and $\mathbf{F}$ for given symmetric matrices $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_N)$ and volume fractions $\Theta = (\theta_1, \cdots, \theta_N)$. Therefore, it is useful to relate the attainability of the HS bounds of composites of one set of materials to the attainability of the HS bounds of composites of a different set of materials. From part (ii) of Theorem 2.1, we have

Corollary 2.2. *Let $(\mathbf{L}_c, \mathbf{L}_0, \cdots, \mathbf{L}_N)$ and $(\mathbf{L}'_c, \mathbf{L}'_0, \cdots, \mathbf{L}'_N)$ be two sets of tensors satisfying* (2.5) *and* (2.6), *and assume that $\mathbf{F}, \mathbf{F}' \in \mathbb{R}^{n\times n}_{sym}$ satisfy*

$$\begin{aligned} &\mathbf{F} - \mathrm{Tr}(\mathbf{F})\frac{(1 + k_c\triangle c_*)}{\triangle c_*(1 + k_c\triangle c_i)}(\mathbf{L}_i - \mathbf{L}_c)^{-1}\mathbf{I} \\ &\quad = \mathbf{F}' - \mathrm{Tr}(\mathbf{F}')\frac{(1 + k'_c\triangle c'_*)}{\triangle c'_*(1 + k'_c\triangle c'_i)}(\mathbf{L}'_i - \mathbf{L}'_c)^{-1}\mathbf{I} \qquad \forall\, i = 1, \cdots, N, \end{aligned} \tag{2.25}$$

where $k'_c, \triangle c'_0, \cdots, \triangle c'_N, \triangle c'_*$ *as in* (2.7) *with* $\mathbf{L}_i$ *replaced by* $\mathbf{L}'_i$ *for all* $i = c, 0, \cdots, N$. *Then the periodic composite* (2.1) *of materials* $(\mathbf{L}_0, \cdots, \mathbf{L}_N)$ *attains the HS bound* (2.18) *for* $\mathbf{F}$ *if, and only if the periodic composite* (2.1) *of materials* $(\mathbf{L}'_0, \cdots, \mathbf{L}'_N)$ *attains the HS bound* (2.18) *for* $\mathbf{F}'$.

3. Optimal microstructures: E-inclusions

The existence of periodic E-inclusions is addressed in a separate publication [23] for a variety of symmetric matrices $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_N)$ and volume fractions $\Theta = (\theta_1, \cdots, \theta_N)$. Below we find sufficient conditions on $\mathbb{K}$ and Θ such that a corresponding periodic E-inclusion can be found or does not exist. This problem is closely related with the problems studied in Müller & Šverák [30]. For the restrictions and constructions of periodic E-inclusions, it will be convenient to restate the concept of periodic E-inclusions in terms of gradient Young measures.

Definition 3.1. *Let* $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_N) \subset \mathbb{R}^{n\times n}_{sym}$, $\Theta = (\theta_1, \cdots, \theta_N) \in (0,1)^N$, $\theta_0 = 1 - \sum_{i=1}^N \theta_i \in (0,1)$, *and* $p_0 \in \mathbb{R}$ *be such that* $\theta_0 p_0 + \sum_{i=1}^N \theta_i \mathrm{Tr}(\mathbf{Q}_i) = 0$. *Corresponding to* $\mathbb{K}$ *and* Θ, *a* **sequential E-inclusion** *is a homogeneous gradient Young measure* ν *that is generated by a sequence in* $W^{1,p}(Y)$ *for any* $1 \le p < \infty$, *has zero center of mass, and satisfies*

$$\nu = \sum_{i=1}^N \theta_i \delta_{\mathbf{Q}_i} + \theta_0 \mu \ \text{ with } \ \mathrm{supp}\,\mu \subset \{X \in \mathbb{R}^{n\times n}_{sym} : \mathrm{Tr}(X) = p_0\}, \tag{3.1}$$

where $\delta_{\mathbf{Q}_i}$ *is the Dirac mass at* $\mathbf{Q}_i$ *and* μ *is a probability measure.*

To see the motivation behind the above definition, we assume there exists a periodic E-inclusion such that (2.22) admits a solution $\xi \in W^{2,2}_{per}(Y)$ for symmetric matrices $\mathbb{K}$ and volume fractions Θ and let $\mathbf{v}^{(k)}(\mathbf{x})$ be $\nabla\xi(k\mathbf{x})/k$ restricted to Y. Then the gradient sequence $\nabla\mathbf{v}^{(k)}$ generates the corresponding sequential E-inclusion, where the Dirac masses at $\mathbf{Q}_i$ arise from Ω_i for $i = 1, \cdots, N$ and the requirement on $\mathrm{supp}\,\mu$ arises from Ω_0, see (2.22). The converse is also true in the following sense: if there exists a sequential E-inclusion, there exists a sequence of microstructures $\mathcal{O}^{(k)} = (\Omega_0^{(k)}, \cdots, \Omega_N^{(k)})$ and a sequence $\xi^{(k)} \in W^{2,p}_{per}(Y)$ for any $1 \le p < \infty$ such that for any continuous $f : \mathbb{R}^{n\times n} \to \mathbb{R}$ with compact support,

$$\lim_{k\to+\infty} \int_Y f(\nabla\nabla\xi^{(k)} - \sum_{i=1}^N \mathbf{Q}_i \chi_i^{(k)} + (p_0 - \Delta\xi^{(k)})\mathbf{I}\chi_0^{(k)}) = f(0), \tag{3.2}$$

where $\chi_i^{(k)}$ is the characteristic function of $\Omega_i^{(k)}$. Note that equation (3.2) implies $\mathbf{L}^e(\mathcal{O}^{(k)})$ attains the HS bounds (2.18) as $k \to +\infty$ if $(\mathbf{L}_c, \mathbf{L}_0, \cdots, \mathbf{L}_N)$ are as in Theorem 2.1 and $\mathbf{F}$, $\mathbb{K}$, Θ satisfy (2.23). Thus, the attainment condition in Theorem 2.1 may be stated as: *For some* $\mathbf{F} \in \mathbb{R}^{n\times n}_{sym}$, *the HS bound* (2.18) *is attainable if, and only if there exists a sequential E-inclusion with symmetric matrices* $\mathbb{K}$ *and volume fractions* Θ *given by* (2.23).

(*a*) *Restrictions on periodic E-inclusions*

There are non-obvious restrictions on matrices $\mathbb{K}$ and volume fractions Θ such that the overdetermined problem (2.22) admits a solution. Liu, James & Leo [23] have shown that $\mathbb{K}$ and Θ necessarily satisfy

$$\sum_{i=1}^{N} \theta_i \mathrm{Tr}(\mathbf{Q}_i)\mathbf{Q}_i + \frac{1}{\theta_0}\sum_{j=1}^{N} \theta_j \mathrm{Tr}(\mathbf{Q}_j)\sum_{i=1}^{N} \theta_i \mathbf{Q}_i - \sum_{i=1}^{N} \theta_i \mathbf{Q}_i^2$$
$$= \theta_0 \fint_{\Omega_0} (\nabla\nabla\xi)^2 \geq \frac{1}{\theta_0}[\sum_{i=1}^{N} \theta_i \mathbf{Q}_i]^2, \tag{3.3}$$

which follows from (2.22), the divergence theorem $\int_Y \Delta\xi\nabla\nabla\xi = \int_Y (\nabla\nabla\xi)^2$, and the Jensen's inequality.

From the basic relation between gradient Young measures and quasiconvex functions [20, 21], we have the following necessary and sufficient condition for the probability measure ν in (3.1) to be a sequential E-inclusion: for any quasiconvex functions $\psi : \mathbb{R}^{n\times n} \to \mathbb{R}$ satisfying $|\psi(X)| \leq C(|X|^p + 1)$ for some $C > 0$ and some $1 \leq p < \infty$,

$$\int_{\mathbb{R}^{n\times n}} \psi(X)\, d\nu(X) = \sum_{i=1}^{N} \theta_i \psi(\mathbf{Q}_i) + \theta_0 \int_{\mathbb{R}^{n\times n}} \psi(X)\, d\mu(X) \geq \psi(0). \tag{3.4}$$

Applying (3.4) to $\mathbb{R}^{n\times n}_{sym} \ni \psi(X) = \mathbf{m}\cdot(\mathrm{Tr}(X)X - X^2)\mathbf{m}$ for any $\mathbf{m} \in \mathbb{R}^n$ (ψ is in fact a null Lagrangian in this second gradient context), we obtain (3.3), see details in [23]. Unfortunately, few explicit quasiconvex functions are known to yield useful restrictions on $\mathbb{K}$ and Θ.

However, if the periodic E-inclusion $(\Omega_1, \cdots, \Omega_N)$ corresponding to $\mathbb{K}$ and Θ is a priori assumed to be Lipschitz and so the solution to the overdetermined problem (2.22) belongs to $W^{2,\infty}_{per}(Y)$ [19], we can derive useful restrictions on $\mathbb{K}$ and Θ by the maximum principle. To see this, for any unit vector $\mathbf{m} \in \mathbb{R}^n$ let $v_{\mathbf{m}} = \mathbf{m}\cdot(\nabla\nabla\xi)^2\mathbf{m}$. Since $\xi \in W^{2,\infty}_{per}(Y)$, $v_{\mathbf{m}} \in C^\infty(\Omega_0)\cap C(\bar{\Omega}_0)$. By the first equality of (2.22), we have

$$\nabla\nabla\xi(\mathbf{x}+) = \nabla\nabla\xi(\mathbf{x}-) + (p_0 - p_i)\mathbf{n}\otimes\mathbf{n} = \mathbf{Q}_i + (p_0 - p_i)\mathbf{n}\otimes\mathbf{n}$$

for any $\mathbf{x} \in \partial\Omega_i \cap \partial\Omega_0$, where $\mathbf{x}+$ ($\mathbf{x}-$) denotes the boundary value approached from inside (outside) Ω_0, and $\mathbf{n}$ is the unit normal on $\partial\Omega_i$. Further, we find

$$\begin{cases} \Delta v_{\mathbf{m}} \geq 0 & \text{on } \Omega_0, \\ v_{\mathbf{m}} = \mathbf{m}\cdot\Lambda_i(\mathbf{n})\mathbf{m} & \text{on } \partial\Omega_0\cap\partial\Omega_i, i = 1, \cdots, N, \end{cases} \tag{3.5}$$

where $\Lambda_i(\mathbf{n}) = [\mathbf{Q}_i + (p_0 - p_i)\mathbf{n}\otimes\mathbf{n}]^2$. By the maximum principle we conclude that

$$|\nabla\nabla\xi|^2 \leq \max\{\mathrm{Tr}(\Lambda_i(\mathbf{n})) : |\mathbf{n}| = 1, i = 1, \cdots N\} \qquad \text{on } \Omega_0, \tag{3.6}$$

and that

$$\text{l.h.s. of (3.3)} \leq \theta_0\lambda_M\mathbf{I}, \tag{3.7}$$

where

$$\lambda_M := \max\{\mathbf{m}\cdot\Lambda_i(\mathbf{n})\mathbf{m} :\ i = 1, \cdots, N,\ |\mathbf{m}| = |\mathbf{n}| = 1\}.$$

In particular, if $\mathbf{Q}_i = \mathbf{I}p_i/n$, taking the trace of (3.3), by (3.6) we arrive at

$$\sum_{i=1}^{N} \theta_i p_i^2 \le \theta_0 p_{i^*}^2 + 2p_{i^*} \sum_{j=1}^{N} \theta_j p_j, \tag{3.8}$$

where i^* is the integer such that the r.h.s. of (3.8) is maximized among all $i \in \{1, \cdots, N\}$.

(b) Constructions of sequential E-inclusions

Using a convexity property of gradient Young measures we can conveniently construct complicated sequential E-inclusions from simple periodic E-inclusions. For brevity, we refer to a gradient Young measure generated by a bounded sequence in $W^{1,p}$ as a $W^{1,p}$ gradient Young measure. Recall the following two theorems:

Theorem 3.1. (Theorem 3.1, *Kinderlehrer & Pedregal [20]) Let ν_1 and ν_2 be two homogeneous $W^{1,\infty}$ gradient Young measures with zero center of mass. Then for each $\lambda \in (0,1)$, the measure $(1-\lambda)\nu_1 + \lambda\nu_2$ is also a homogeneous $W^{1,\infty}$ gradient Young measure with zero center of mass.*

Theorem 3.2. (Theorem 3, *Liu, James & Leo [23]) Let $\mathbf{Q} \in \mathbb{R}^{n\times n}_{sym}$ be either negative semi-definite or positive semi-definite. Then for each $\theta \in (0,1)$, there exists a $W^{1,\infty}$ sequential E-inclusion of form*

$$\nu = \theta\delta_{\mathbf{Q}} + (1-\theta)\mu. \tag{3.9}$$

From Theorem 3.2 and Theorem 3.1, we have

Theorem 3.3. *Let $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_N)$ be either all negative semi-definite or all positive semi-definite and $\Theta = (\theta_1, \cdots, \theta_N)$ satisfy*

$$\theta_1, \cdots, \theta_N \in (0,1), \qquad \theta_0 = 1 - \sum_{i=1}^{N} \theta_i \in (0,1). \tag{3.10}$$

Then there exists a $W^{1,\infty}$ sequential E-inclusion of form

$$\nu = \sum_{i=1}^{N} \theta_i \delta_{\mathbf{Q}_i} + \theta_0\mu. \tag{3.11}$$

Proof. We prove the theorem by induction. If $N = 1$, the theorem holds by Theorem 3.2. Assume the theorem holds for $1 \le N \le k$, below we show the theorem holds for $N = k+1$.

Let $\Theta = (\theta_1, \cdots, \theta_{k+1})$ satisfy (3.10) for $N = k+1$. By multiplying the generating sequence $\mathbf{v}^{(k)}$ by any constant $a \in \mathbb{R}$, we see that there exists a $W^{1,\infty}$ sequential E-inclusions with $a\mathbb{K}$ and Θ if there exists a $W^{1,\infty}$ sequential E-inclusions with $\mathbb{K} = (\mathbf{Q}_1, \cdots, \mathbf{Q}_{k+1})$ and Θ. Therefore, without loss of generality we assume that $\mathbb{K}$ are all negative semi-definite. Let $p_0 \in \mathbb{R}$ be such that $\theta_0 p_0 + \sum_{i=1}^{k+1} \theta_i \mathrm{Tr}(\mathbf{Q}_i) = 0$ and $\alpha \in (0,1)$ be such that $\alpha\mathrm{Tr}(\mathbf{Q}_{k+1}) + (1-\alpha)p_0 = 0$. If $p_0 = 0$, the theorem is trivial since $\mathrm{Tr}(\mathbf{Q}_i) \le 0$ for all $i = 1, \cdots, k+1$. If $\mathrm{Tr}(\mathbf{Q}_i) = 0$ for some i, the theorem follows from the inductive assumption, Theorem 3.1, and the Direct mass supported at zero matrix is a gradient Young measure. Subsequently we assume $p_0 > 0$ and $\mathrm{Tr}(\mathbf{Q}_i) < 0$ for all $i = 1, \cdots, k+1$.

Direct calculations verify that

$$\alpha = \frac{p_0}{p_0 - \mathrm{Tr}(\mathbf{Q}_{k+1})} > \frac{p_0 - p_0 \sum_{i=1}^k \theta_i + \sum_{i=1}^k \theta_i \mathrm{Tr}(\mathbf{Q}_i)}{p_0 - \mathrm{Tr}(\mathbf{Q}_{k+1})} = \theta_{k+1}, \tag{3.12}$$

and that $\theta_{k+1}\mathrm{Tr}(\mathbf{Q}_{k+1}) + \theta_0 p_0 > 0$,

$$\frac{\theta_{k+1}}{\alpha}(1-\alpha) = \frac{-\mathrm{Tr}(\mathbf{Q}_{k+1})\theta_{k+1}}{p_0} < \theta_0. \tag{3.13}$$

Define λ and θ'_i $(i = 1, \cdots, k)$ by

$$\lambda = \frac{\theta_{k+1}}{\alpha}, \quad (1-\lambda)\theta'_0 + \lambda(1-\alpha) = \theta_0 \quad \text{and} \quad (1-\lambda)\theta'_i = \theta_i \ . \tag{3.14}$$

From (3.12) and (3.13), we see that $\lambda \in (0,1)$ and $\theta'_0, \cdots, \theta'_k > 0$. In particular, $\theta'_0 > 0$ follows from (3.13) and $(1-\lambda)\theta'_0 = \theta_0 - \lambda(1-\alpha) = \theta_0 - \frac{\theta_{k+1}}{\alpha}(1-\alpha)$. Further,

$$\sum_{i=0}^k \theta'_i = \frac{1}{1-\lambda}\sum_{i=0}^k \theta_i - \frac{\lambda(1-\alpha)}{1-\lambda} = \frac{-\lambda + \sum_{i=0}^{k+1}\theta_i}{1-\lambda} = 1.$$

Thus, $(\theta'_1, \cdots, \theta'_k)$ satisfy (3.10) for $N = k$. By the inductive assumption, for $N = k$ we have the existence of a $W^{1,\infty}$ sequential E-inclusion

$$\nu_1 = \sum_{i=1}^k \theta'_i \delta_{\mathbf{Q}_i} + \theta'_0 \mu_1, \tag{3.15}$$

where μ_1 is a probability measure with

$$\mathrm{supp}\, \mu_1 \subset \{X \in \mathbb{R}^{n\times n}_{sym} : \ \sum_{i=1}^k \theta'_i \mathrm{Tr}(\mathbf{Q}_i) + \theta'_0 \mathrm{Tr}(X) = 0\}.$$

By Theorem 3.2, we also have the existence of a $W^{1,\infty}$ sequential E-inclusion

$$\nu_2 = \alpha\delta_{\mathbf{Q}_{k+1}} + (1-\alpha)\mu_2, \tag{3.16}$$

where μ_2 is a probability measure with $\mathrm{supp}\, \mu_2 \subset \{X \in \mathbb{R}^{n\times n}_{sym} : \ \mathrm{Tr}(X) = p_0\}$.

Let p'_0 be such that $\sum_{i=1}^k \theta'_i \mathrm{Tr}(\mathbf{Q}_i) + \theta'_0 p'_0 = 0$. From (3.14), we have $\sum_{i=1}^k \frac{\theta_i \mathrm{Tr}(\mathbf{Q}_i)}{1-\lambda} + \frac{p'_0}{1-\lambda}[\theta_0 - \lambda(1-\alpha)] = 0$, which, by (3.13) and the definition of p_0, implies

$$\begin{aligned} 0 &= [\theta_0 + \frac{\theta_{k+1}\mathrm{Tr}(\mathbf{Q}_{k+1})}{p_0}]p'_0 - \theta_{k+1}\mathrm{Tr}(\mathbf{Q}_{k+1}) - p_0\theta_0 \\ &= \frac{1}{p_0}[\theta_0 p_0 + \theta_{k+1}\mathrm{Tr}(\mathbf{Q}_{k+1})](p'_0 - p_0). \end{aligned} \tag{3.17}$$

If $\theta_0 p_0 + \theta_{k+1}\mathrm{Tr}(\mathbf{Q}_{k+1}) = 0$, by (3.13) and (3.14) we have $\theta'_0 = 0$. Thus, $p_0 = p'_0$ and we define

$$\nu := \lambda\nu_2 + (1-\lambda)\nu_1 = \sum_{i=1}^{k+1} \theta_i \delta_{\mathbf{Q}_i} + \theta_0 \mu, \tag{3.18}$$

where the equality follows from (3.14), (3.15) and (3.16), $\mu = \frac{\lambda(1-\alpha)}{\theta_0}\mu_2 + \frac{(1-\lambda)\theta'_0}{\theta_0}\mu_1$ is a probability measure with supp $\mu \subset \{X \in \mathbb{R}^{n\times n}_{sym} :\ \mathrm{Tr}(X) = p_0\}$. From Theorem 3.1 and Definition 3.1, we see that ν defined by (3.18) is a $W^{1,\infty}$ sequential E-inclusion corresponding to $\mathbb{K}$ and Θ. The proof of the theorem is completed. □

4. Applications

The practical problem we attempt to solve is to characterize the effective tensors that one can obtain by mixing multiple (≥ 3) materials with given volume fractions, i.e., the G_Θ-closure problem. We do not yet have a complete answer to this problem. The progress lies in a series of sufficient conditions such that the HS bounds (2.18) are attainable or unattainable. These results follow from Theorem 2.1, the restrictions and existence of periodic E-inclusions. We address composites of conductive materials and elastic materials.

(*a*) *Composites of conductive materials*

Consider conductive composites of $(N+1)$-phases with conductivity tensors $0 < \mathbf{A}_0, \cdots, \mathbf{A}_N \in \mathbb{R}^{n\times n}_{sym}$ and volume fractions $\theta_0 \in (0,1)$, $\Theta = (\theta_1, \cdots, \theta_N) \in (0,1)^N$. According to (2.5), we assume

$$\mathbf{A}_0 = k_0 \mathbf{I}, \quad \mathbf{A}_N = k_N \mathbf{I}, \tag{4.1}$$

$$\mathbf{A}_0 < \mathbf{A}_1, \mathbf{A}_2, \cdots, \mathbf{A}_{N-1} < \mathbf{A}_N, \tag{4.2}$$

and denote by $\mathbf{A}^e$ the effective conductivity tensor of a composite. To use Theorem 2.1, we set $(\mathbf{L}_i)_{pjqk} = \delta_{pq}(\mathbf{A}_i)_{jk}$ for $i = 0, \cdots, N$ and choose $(\mathbf{L}_c)_{pjqk} = k_0 \delta_{pq}\delta_{jk}$ for the lower bound and $(\mathbf{L}_c)_{pjqk} = k_N \delta_{pq}\delta_{jk}$ for the upper bound. By (2.2), we verify that the effective tensor $\mathbf{L}^e$ can be written as $(\mathbf{L}^e)_{piqj} = \delta_{pq}(\mathbf{A}^e)_{ij}$. By (2.4) and (4.2), we have

$$\mathbf{A}_0 < \mathbf{H}_\Theta \leq \mathbf{A}^e \leq \mathbf{A}_\Theta < \mathbf{A}_N, \tag{4.3}$$

where $\mathbf{A}_\Theta = \sum_{i=0}^N \theta_i \mathbf{A}_i$ and $\mathbf{H}_\Theta = [\sum_{i=0}^N \theta_i \mathbf{A}_i^{-1}]^{-1}$. From (2.18) we have

$$\begin{cases} \mathrm{Tr}(\mathbf{F}^T\mathbf{F}) + (\mathrm{Tr}\mathbf{F})^2/\triangle c_*^L \leq \mathbf{A}_0^{-1/2}\mathbf{A}^e\mathbf{A}_0^{-1/2} \cdot \mathbf{F}^T\mathbf{F} & \forall \mathbf{F} \in \mathbb{R}^{n\times n}, \\ \mathrm{Tr}(\mathbf{F}^T\mathbf{F}) + (\mathrm{Tr}\mathbf{F})^2/\triangle c_*^U \geq \mathbf{A}_N^{-1/2}\mathbf{A}^e\mathbf{A}_N^{-1/2} \cdot \mathbf{F}^T\mathbf{F} & \forall \mathbf{F} \in \mathbb{R}^{n\times n}, \end{cases} \tag{4.4}$$

or equivalently by (A.1),

$$\begin{cases} \mathrm{Tr}[(\mathbf{A}_0^{-1/2}\mathbf{A}^e\mathbf{A}_0^{-1/2} - \mathbf{I})^{-1}] \leq \triangle c_*^L, \\ \mathrm{Tr}[(\mathbf{I} - \mathbf{A}_N^{-1/2}\mathbf{A}^e\mathbf{A}_N^{-1/2})^{-1}] \leq -\triangle c_*^U, \end{cases} \tag{4.5}$$

where, by (2.7),

$$\begin{cases} \triangle c_i^L = \mathrm{Tr}[(\mathbf{A}_0^{-1/2}\mathbf{A}_i\mathbf{A}_0^{-1/2} - \mathbf{I})^{-1}] > 0 & i = 1, \cdots, N, \\ \triangle c_i^U = -\mathrm{Tr}[(\mathbf{I} - \mathbf{A}_N^{-1/2}\mathbf{A}_i\mathbf{A}_N^{-1/2})^{-1}] < 0 & i = 0, \cdots, N-1, \end{cases} \tag{4.6}$$

$$\triangle c_*^L = \frac{\theta_0 + \sum_{i=1}^N \triangle c_i^L \frac{\theta_i}{1+\triangle c_i^L}}{\sum_{i=1}^N \frac{\theta_i}{1+\triangle c_i^L}} > 0, \qquad \triangle c_*^U = \frac{\theta_N + \sum_{i=0}^{N-1} \triangle c_i^U \frac{\theta_i}{1+\triangle c_i^U}}{\sum_{i=0}^{N-1} \frac{\theta_i}{1+\triangle c_i^U}} < -1. \tag{4.7}$$

We remark that the bounds (4.3), (4.5) and (4.4) are valid without assuming (4.1). We now discuss if these bounds completely describe the G_Θ-closure, i.e., the collection of effective conductivity tensors that one can obtain by mixing $(\mathbf{A}_0, \cdots, \mathbf{A}_N)$ with volume fraction $(\theta_0, \cdots, \theta_N)$. We denote by G_Θ the G_Θ-closure and G_Θ^{out} the set of symmetric matrices $\mathbf{A}^e$ that satisfy (4.4) or, equivalently, (4.3) and (4.5). The set $G_\Theta^{out} \subset \mathbb{R}^{n\times n}_{sym}$ is clearly compact and convex and contains G_Θ. Further, for some $\mathbf{A}^e \in G_\Theta^{out}$, if both inequalities in (4.5) are strict and $\mathbf{A}^e < \mathbf{A}_\Theta$, then $\mathbf{A}^e > \mathbf{H}_\Theta$. Thus, the HS bounds describe a generic boundary point of G_Θ^{out} in the sense that

$$\partial G_\Theta^{out} = \mathcal{S}^L \cup \mathcal{S}^U \cup \{\mathbf{A}^e \in G_\Theta^{out} : \ \mathrm{rank}(\mathbf{A}_\Theta - \mathbf{A}^e) < n\}, \tag{4.8}$$

where

$$\begin{aligned} \mathcal{S}^L &= \{\mathbf{A}^e \in G_\Theta^{out} : \ \mathrm{Tr}[(\mathbf{A}_0^{-1/2}\mathbf{A}^e\mathbf{A}_0^{-1/2} - \mathbf{I})^{-1}] = \triangle c_*^L\}, \\ \mathcal{S}^U &= \{\mathbf{A}^e \in G_\Theta^{out} : \ \mathrm{Tr}[(\mathbf{A}_N^{-1/2}\mathbf{A}^e\mathbf{A}_N^{-1/2} - \mathbf{I})^{-1}] = \triangle c_*^U\}, \end{aligned} \tag{4.9}$$

are two hypersurfaces in $\mathbb{R}^{n\times n}_{sym}$ defined by the HS bounds (4.5). As demonstrated by the following theorem, the attainability of the HS bounds, i.e., $\mathcal{S}^L \cup \mathcal{S}^U$, plays an important role in estimating how well G_Θ^{out} approximates G_Θ.

Theorem 4.1. *Consider conductive composites of* $(N+1)$*-phases with conductivity tensors* $\mathbf{A}_0 < \mathbf{A}_1, \cdots, \mathbf{A}_{N-1} < \mathbf{A}_N$. *If* $(\mathcal{S}^L \cup \mathcal{S}^U) \subset G_\Theta$, *then*

$$G_\Theta(\mathbf{A}_0, \cdots, \mathbf{A}_N) = G_\Theta^{out}(\mathbf{A}_0, \cdots, \mathbf{A}_N). \tag{4.10}$$

Proof. Let $\mathbf{A}^e$ be an interior point in G_Θ^{out}, $\mathbf{A}(t) = \mathbf{A}^e + t(\mathbf{A}_\Theta - \mathbf{A}^e)$, $t_0 := \inf\{t : \mathbf{A}(t) \geq 0\}$, and $\mathcal{A} = \{\mathbf{A}(t) : t_0 < t < 1\}$. By (4.8) we have $\mathrm{rank}(\mathbf{A}_\Theta - \mathbf{A}^e) = n$. We verify that neither of the endpoints of $\mathcal{A}$ is contained in G_Θ^{out} since $\mathbf{A}(t_0) - \mathbf{H}_\Theta$ is not positive semi-definite and $\mathrm{Tr}[(\mathbf{I} - \mathbf{A}_N^{-1/2}\mathbf{A}(1)\mathbf{A}_N^{-1/2})^{-1}] > -\triangle c_*^U$. Therefore, $\mathcal{A} \cap \partial G_\Theta^{out}$ contains at least two distinct points $\mathbf{A}(t_1)$ and $\mathbf{A}(t_2)$ that satisfy $\mathbf{A}(t_1) < \mathbf{A}(0) < \mathbf{A}(t_2)$. Further, $\mathrm{rank}(\mathbf{A}_\Theta - \mathbf{A}(t)) = \mathrm{rank}((1-t)(\mathbf{A}_\Theta - \mathbf{A}^e)) = n$ for any $t_0 < t < 1$. By (4.8), we see both $\mathbf{A}(t_1)$ and $\mathbf{A}(t_2)$ are contained in $\mathcal{S}^L \cup \mathcal{S}^U$. Since $\mathcal{S}^L \cup \mathcal{S}^U \subset G_\Theta$ and the G-closure of $\mathbf{A}(t_1)$ and $\mathbf{A}(t_2)$ is closed and convex (see [12]), we infer $\mathbf{A}^e = \mathbf{A}(0) \in G_\Theta$. □

Grabovsky [12] has shown that all HS bounds are attainable and hence $G_\Theta^{out} = G_\Theta$ for two-phase well-ordered conductive composites. In general, not all HS bounds are attainable for multiphase composites. Below we give sufficient conditions for the HS bounds become attainable or unattainable.

Corollary 4.2. *Let*

$$\begin{cases}\mathbf{Q}_i^L = (\mathbf{A}_0^{-1/2}\mathbf{A}^e\mathbf{A}_0^{-1/2} - \mathbf{I})^{-1} - \frac{1+\triangle c_*^L}{1+\triangle c_i^L}(\mathbf{A}_0^{-1/2}\mathbf{A}_i\mathbf{A}_0^{-1/2} - \mathbf{I})^{-1} & \text{if } \mathbf{A}^e \in \mathcal{S}^L,\\ \mathbf{Q}_i^U = -(\mathbf{I} - \mathbf{A}_N^{-1/2}\mathbf{A}^e\mathbf{A}_N^{-1/2})^{-1} + \frac{1+\triangle c_*^U}{1+\triangle c_i^U}(\mathbf{I} - \mathbf{A}_N^{-1/2}\mathbf{A}_i\mathbf{A}_N^{-1/2})^{-1} & \text{if } \mathbf{A}^e \in \mathcal{S}^U,\end{cases}$$

$\mathbb{K}^L = (\mathbf{Q}_1^L, \cdots, \mathbf{Q}_N^L)$ *and* $\mathbb{K}^U = (\mathbf{Q}_0^U, \cdots, \mathbf{Q}_{N-1}^U)$. *(i) If* $\mathbb{K}^L$ *(*$\mathbb{K}^U$*) are all positive semi-definite or all negative semi-definite, then the effective tensor* $\mathbf{A}^e \in \mathcal{S}^L$ $(\mathcal{S}^U)$ *is attainable; (ii) if* $\mathbb{K}^L$ *(*$\mathbb{K}^U$*) and* Θ *violates* (3.3) *or* (3.7) *or* (3.8)*, then the effective tensor* $\mathbf{A}^e \in \mathcal{S}^L$ $(\mathcal{S}^U)$ *is unattainable.*

Proof. By Theorem A.1, equation (A.2) and Theorem 2.1, equation (2.23), we see that $\mathbf{A}^e \in \mathcal{S}^L$ ($\mathbf{A}^e \in \mathcal{S}^U$) is attainable if and only if there exists a sequential E-inclusion corresponding to $\mathbb{K}^L$ ($\mathbb{K}^U$) and Θ. Part (i) of the Corollary follows from Theorem 3.3 and part (ii) follows from the restrictions on periodic E-inclusions, i.e., (3.3), (3.7) and (3.8). □

Below we specialize the above results to isotropic composites of $(N+1)$-isotropic phases of $0 < k_0 < k_1 < \cdots < k_{N-1} < k_N$. Denote by k^e the effective conductivity of the composite. Then the HS bounds (4.4) can be written as

$$k_0 + nk_0/\triangle c_*^L =: k^L \le k^e \le k^U := k_N + nk_N/\triangle c_*^U, \tag{4.11}$$

where

$$\begin{cases}\triangle c_*^L = \frac{\theta_0 + \sum_{i=1}^N \theta_i \triangle c_i^L/(1+\triangle c_i^L)}{\sum_{i=1}^N \theta_i/(1+\triangle c_i^L)},\\ \triangle c_*^U = \frac{\theta_N + \sum_{i=0}^{N-1} \theta_i \triangle c_i^U/(1+\triangle c_i^U)}{\sum_{i=0}^{N-1} \theta_i/(1+\triangle c_i^U)},\end{cases} \qquad \begin{cases}\triangle c_i^L = \frac{nk_0}{k_i - k_0},\\ \triangle c_i^U = \frac{nk_N}{k_i - k_N}.\end{cases} \tag{4.12}$$

Further, by Corollary 4.2, the lower (upper) HS bound in (4.11) is attainable if there exists a periodic E-inclusion corresponding to $\mathbb{K}^L = (\frac{p_1^L}{n}\mathbf{I}, \cdots, \frac{p_N^L}{n}\mathbf{I})$ ($\mathbb{K}^U = (\frac{p_0^U}{n}\mathbf{I}, \cdots, \frac{p_{N-1}^U}{n}\mathbf{I})$) and volume fractions Θ, where

$$\begin{cases}p_i^L = \mathrm{Tr}(\mathbf{Q}_i^L) = \frac{\triangle c_*^L - \triangle c_i^L}{(1+\triangle c_i^L)} & i = 1, \cdots, N,\\ p_i^U = \mathrm{Tr}(\mathbf{Q}_i^U) = \frac{\triangle c_*^U - \triangle c_i^U}{(1+\triangle c_i^U)} & i = 0, \cdots, N-1.\end{cases} \tag{4.13}$$

By part (i) of Corollary 4.2, we conclude that *the lower HS bound* $k^L \le k^e$ *is attainable if* $k^L \le k_1$ *and that the upper bound* $k^e \le k^U$ *is attainable if* $k^U \ge k_{N-1}$. We remark that these attainability results concerning isotropic composites of isotropic materials were first shown by Milton [26].

We now discuss the implication of (3.8). By (4.13) and (4.12), direct calculations reveal that

$$\frac{p_j^L}{p_i^L} - 1 = \frac{\rho_{ji}}{\theta_0 - \sum_{k=1}^N \theta_k \rho_{ki}}, \qquad \rho_{ji} = \frac{\triangle c_i^L - \triangle c_j^L}{(1+\triangle c_j^L)} = \frac{nk_0(k_j - k_i)}{(k_j + (n-1)k_0)(k_i - k_0)}.$$

Note that ρ_{ji} does not depend on volume fractions. By (3.8) we conclude that if

$$\sum_{k=1}^N \theta_k \rho_{ki^*}^2 > (\theta_0 - \sum_{k=1}^N \theta_k \rho_{ki^*})^2, \tag{4.14}$$

then there exists no Lipschitz periodic E-inclusions, and hence the lower HS bound k^L is unattainable (by Lipschitz microstructures). We remark that, specialized to two dimensions and three-phase composites, the above conditions on unattainable HS bounds have been shown in [1, 7]. Similar results hold for the upper bound, which we will not repeat here.

(*b*) *Composites of elastic materials*

We now consider elastic composites of $(N+1)$ phases with elasticity tensors given by $\mathbf{L}_0, \cdots, \mathbf{L}_N$ and volume fractions $\theta_0, \cdots, \theta_N \in (0,1)$. Let μ_0 (μ_N) be the greatest (least) number such that for all $0 \neq \mathbf{F} \in \mathbb{R}^{n\times n}_{sym}$ with $\mathrm{Tr}(\mathbf{F}) = 0$,

$$2\mu_0|\mathbf{F}|^2 \leq \min_{i\in\{0,\cdots,N\}} \mathbf{F}\cdot\mathbf{L}_i\mathbf{F} \qquad (2\mu_N|\mathbf{F}|^2 \geq \max_{i\in\{0,\cdots,N\}} \mathbf{F}\cdot\mathbf{L}_i\mathbf{F}), \tag{4.15}$$

and κ_c^L (κ_c^U) be the least (greatest) number in $\{\mathbf{I}\cdot\mathbf{L}_i\mathbf{I}/n^2 : \ i = 0,\cdots,N\}$, $(\mathbf{L}_c^L)_{piqj} = \mu_0(\delta_{ij}\delta_{pq} + \delta_{pj}\delta_{iq}) + (\kappa_c^L - 2\mu_0/n)\delta_{ip}\delta_{jq}$, and $(\mathbf{L}_c^U)_{piqj} = \mu_N(\delta_{ij}\delta_{pq} + \delta_{pj}\delta_{iq}) + (\kappa_c^U - 2\mu_N/n)\delta_{ip}\delta_{jq}$. Choosing $\mathbf{L}_c^L$ as the comparison tensor for the lower bound and $\mathbf{L}_c^U$ as the comparison tensor for the upper bound, we write the HS bounds (2.18) as for any $\mathbf{F}\in\mathbb{R}^{n\times n}$,

$$\mathbf{F}\cdot\mathbf{L}_c^L\mathbf{F} + (\mathrm{Tr}\mathbf{F})^2/\triangle c_*^L \leq \mathbf{F}\cdot\mathbf{L}^e\mathbf{F} \leq \mathbf{F}\cdot\mathbf{L}_c^U\mathbf{F} + (\mathrm{Tr}\mathbf{F})^2/\triangle c_*^U, \tag{4.16}$$

where, by (2.7),

$$\begin{cases}\triangle c_i^L = \mathbf{I}\cdot(\mathbf{L}_i - \mathbf{L}_c^L)^{-1}\mathbf{I}, \\ \triangle c_i^U = \mathbf{I}\cdot(\mathbf{L}_i - \mathbf{L}_c^U)^{-1}\mathbf{I},\end{cases} \qquad \begin{cases}\triangle c_*^L = \frac{\sum_{i=0}^N \theta_i \triangle c_i^L/(1/k_c^L + \triangle c_i^L)}{\sum_{i=0}^N \theta_i/(1/k_c^L+\triangle c_i^L)}, \\ \triangle c_*^U = \frac{\sum_{i=0}^N \theta_i \triangle c_i^U/(1/k_c^U + \triangle c_i^U)}{\sum_{i=0}^N \theta_i/(1/k_c^U+\triangle c_i^U)},\end{cases} \tag{4.17}$$

$k_c^L = \kappa_c^L + 2\mu_0(1-1/n)$ and $k_c^U = \kappa_c^U + 2\mu_N(1-1/n)$. It is worthwhile noticing that the lower (upper) bound in (4.16) is valid for general anisotropic elasticity tensors $\mathbf{L}_0,\cdots,\mathbf{L}_N$. Below we assume that $\mathbf{L}_0$ and $\mathbf{L}_N$ are isotropic with shear modulus μ_0 and μ_N and equation (2.5) is satisfied.

Unlike conductivity problems we cannot determine the effective tensor $\mathbf{L}^e$ by (A.2). Thus, it is more difficult to show the attainability of a given effective elasticity tensor. Nevertheless, we can discuss the attainability of a particular component of the effective elasticity tensor, e.g., the bulk modulus. By Theorem 2.1 and Theorem 3.3, the lower bound in (4.16) is attainable for some $\mathbf{F}\in\mathbb{R}^{n\times n}_{sym}$ if

$$\mathbf{Q}_i^L = \mathbf{F} - \mathrm{Tr}(\mathbf{F})\frac{(1+k_c^L\triangle c_*^L)}{\triangle c_*^L(1+k_c^L\triangle c_i^L)}(\mathbf{L}_i - \mathbf{L}_c^L)^{-1}\mathbf{I} \ \ (i=1,\cdots,N) \tag{4.18}$$

are all negative semi-definite or all positive semi-definite, whereas the upper bound in (4.16) is attainable for some $\mathbf{F}\in\mathbb{R}^{n\times n}_{sym}$ if

$$\mathbf{Q}_i^U = \mathbf{F} - \mathrm{Tr}(\mathbf{F})\frac{(1+k_c^U\triangle c_*^U)}{\triangle c_*^U(1+k_c^U\triangle c_i^U)}(\mathbf{L}_i - \mathbf{L}_c^U)^{-1}\mathbf{I} \ \ (i=0,\cdots,N-1) \tag{4.19}$$

are all negative semi-definite or all positive semi-definite.

We now focus on the bulk modulus. Let $\kappa^e = \mathbf{I} \cdot \mathbf{L}^e \mathbf{I}/n^2$ be the effective bulk modulus of the composite. Choosing $\mathbf{F} = \mathbf{I}$ in (4.16) we obtain

$$\kappa_c^L + 1/\triangle c_*^L \le \kappa^e \le \kappa_c^U + 1/\triangle c_*^U, \tag{4.20}$$

which coincides with the Walpole's bounds [39] for bulk modulus. If we assume that $\mathbf{L}_0, \cdots, \mathbf{L}_N$ are all isotropic tensors, then the symmetric matrices in (4.18) and (4.19) can be written as

$$\mathbf{Q}_i^L = \frac{\mathrm{Tr}(\mathbf{Q}_i^L)}{n}\mathbf{I} = \frac{(\triangle c_*^L - \triangle c_i^L)}{\triangle c_*^L(1 + k_c^L \triangle c_i^L)}\mathbf{I}, \qquad \mathbf{Q}_i^U = \frac{\mathrm{Tr}(\mathbf{Q}_i^U)}{n}\mathbf{I} = \frac{(\triangle c_*^U - \triangle c_i^U)}{\triangle c_*^U(1 + k_c^U \triangle c_i^U)}\mathbf{I},$$

and hence $\mathbb{K}^L = (\mathbf{Q}_1^L, \cdots, \mathbf{Q}_N^L)$ ($\mathbb{K}^U = (\mathbf{Q}_0^L, \cdots, \mathbf{Q}_{N-1}^L)$) are negative semi-definite or positive semi-definite is equivalent to

$$\triangle c_*^L \ge \max_{i \in \{1, \cdots, N\}} \triangle c_i^L \quad \text{or} \quad \triangle c_*^L \le \min_{i \in \{1, \cdots, N\}} \triangle c_i^L \tag{4.21}$$

$$\big(\triangle c_*^U \le \min_{i \in \{0, \cdots, N-1\}} \triangle c_i^U \quad \text{or} \quad \triangle c_*^U \ge \max_{i \in \{0, \cdots, N-1\}} \triangle c_i^U\big). \tag{4.22}$$

Therefore, we conclude that the lower bound in (4.20) is attainable if (4.21) is satisfied while the upper bound in (4.20) is attainable if (4.22) is satisfied. These attainability results have been obtained by Milton [26]. Sufficient conditions for unattainable HS bounds follow from similar discussions as for conductive composites, which we will not repeat here.

5. Summary and discussion

We have derived a necessary and sufficient condition for the HS bounds to be attainable. This condition yields a simple characterization of the optimal gradient fields and motivates us to introduce the concept of (sequential) E-inclusions. A special quasiconvex function and the maximum principle are used to restrict sequential E-inclusions, while a convexity property of gradient Young measures is used to show the existence of a class of sequential E-inclusions. From these results, we find sufficient conditions on the attainable and unattainable HS bounds for composites of any finite number of conductive materials or elastic materials in any dimensions.

We have restricted ourselves to periodic composites for the ease of the definition of the effective tensors (cf. (2.2)) and the formal proofs. Since any effective tensors can be approximated arbitrarily well by those of periodic microstructures, the results in this paper shall remain valid without assuming periodicity.

Since the G-closure of two well-ordered conductive materials can be realized by multi-rank laminations [24, 36, 12], Theorem 2.1 suggests that sequential E-inclusions in Theorem 3.2 can all be realized by multi-rank laminations. Further, it is sufficient to consider simple laminations to prove Theorem 3.1, see [20]. From these two facts we may infer that sequential E-inclusions in Theorem 3.3, and therefore all attainable HS bounds in Section 4, can be realized by multi-rank laminations. A formal proof of this statement is not pursued here.

To establish the existence of sequential E-inclusions without assuming positive or negative semi-definite symmetric matrices $\mathbb{K}$ (cf., Theorem 3.3), we have to resort to the conventional way of constructions. We are aware of three types of constructions that can give rise to sequential E-inclusions not covered by Theorem 3.3:

1. In the case of two dimensions ($n = 2$) and three phases ($N = 2$), the Sigmund and coworker's constructions [32, 11] in effect asserts the existence of sequential E-inclusions corresponding to $\mathbb{K} = (p_1\mathbf{I}/2, p_2\mathbf{I}/2)$ and $\Theta = (\theta_1, \theta_2)$ if (3.8) is satisfied, i.e.,

$$\theta_1(\frac{p_1}{p_2} - 1)^2 \leq 1 \quad \text{or} \quad \theta_2(\frac{p_2}{p_1} - 1)^2 \leq 1.$$

   We remark that the result of [2, 7] implies the above condition is also necessary for the existence of sequential E-inclusions with $\mathbb{K}$ being isotropic matrices in two dimensions.

2. In the case of two dimensions ($n = 2$) and three phases ($N = 2$), the constructions of [1] assert the existence of sequential E-inclusions not covered by Theorem 3.3. However, we do not have a simple formula on $\mathbb{K}$ and Θ associated with sequential E-inclusions that can be realized by this constructions.

3. In two and higher dimensions, $N \geq 2$, periodic E-inclusions can give rise to sequential E-inclusions that have Dirac masses supported on both negative definite matrices and positive definite matrices, see [23].

All the above constructions could be important in extending the attainable HS bounds. A systematic study is underway and will be reported in the future.

Finally, we make a few comments on possible generalizations. First of all, one notices that in Section 2 the minimization problem (2.2) may be tested with a linear combination of the second gradient of a scalar potential. Then the restriction of $m = n$ may be removed and the cross-property bounds [6, 33] may be derived. Further, the comparison material can be generalized without being restricted to the form of (2.6). We can in fact extend our argument to the comparison tensors that satisfy ($m = n$)

$$(\mathbf{L}_c)_{piqj}(\mathbf{k})_i(\mathbf{k})_j(\mathbf{k})_q = k_c|\mathbf{k}|^2(\mathbf{k})_p \qquad \forall\, \mathbf{k} \in \mathbb{R}^n \tag{5.1}$$

for some $k_c > 0$, see [23]. Further, by a linear transformation

$$\mathbf{x} \longrightarrow \mathbf{x}' = \Lambda^{-1}\mathbf{x} \qquad \text{and} \qquad \mathbf{v} \longrightarrow \mathbf{v}' = \mathbf{G}^{-1}\mathbf{v}, \tag{5.2}$$

we can extend Theorem 2.1 to comparison tensors (while other conditions on materials properties in Theorem 2.1 remain unchanged)

$$(\mathbf{L}'_c)_{piqj} = (\mathbf{G})_{rp}(\mathbf{G})_{sq}(\Lambda)_{ik}(\Lambda)_{jl}(\mathbf{L}_c)_{rksl},$$

where $\mathbf{G}$, $\Lambda \in \mathbb{R}^{n\times n}$ are invertible, $\mathbf{L}_c$ satisfies (5.1). In fact we have used the transformations (5.2) with $\Lambda = \mathbf{A}_0^{-1/2}$ (or $\Lambda = \mathbf{A}_N^{-1/2}$) and $\mathbf{G} = \mathbf{I}$ in writing the bounds as (4.5) and (4.4). The reader is invited to formulate the precise statements corresponding to Theorem 2.1 for tensors $\mathbf{L}_c$ of these forms.

Appendix: the dual HS bounds

The HS bounds can be derived from the HS variational principle [29] and usually take the following form

$$\begin{cases} \mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}\le \triangle c_* & \text{if } \mathbf{L}(\mathbf{x},\mathcal{O})\ge \mathbf{L}_c, \\ \mathbf{I}\cdot(\mathbf{L}_c-\mathbf{L}^e)^{-1}\mathbf{I}\le -\triangle c_* & \text{if } \mathbf{L}(\mathbf{x},\mathcal{O})\le \mathbf{L}_c. \end{cases} \tag{A.1}$$

The above bounds can be regarded as the dual bounds of (2.18). More precisely,

Theorem A.1. *Assume* (2.4) *and* (2.5). *Then inequalities* (2.18) *are equivalent to* (A.1). *Further, for some* $\mathbf{F}\in\mathcal{R}(\mathbf{L}^e-\mathbf{L}_c)$ *with* $\mathrm{Tr}(\mathbf{F})\neq 0$, *one of the inequalities in* (2.18) *holds as an equality if, and only if the corresponding inequality in* (A.1) *holds as an equality. In this case, we have*

$$\frac{\mathbf{F}}{\mathrm{Tr}(\mathbf{F})}=\frac{1}{\triangle c_*}(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}. \tag{A.2}$$

Proof. We note that (2.4) and (2.5) imply $\mathcal{R}(\mathbf{L}^e-\mathbf{L}_c)\supset\mathbb{R}^{n\times n}_{sym}$. Consider the case $\mathbf{L}(\mathbf{x},\mathcal{O})\ge\mathbf{L}_c$. To show (A.1) implies (2.18), by $\mathbf{L}^e\ge\mathbf{L}_c$, (1.1) and (A.1) we have

$$\sup_{\mathbf{F}\in\mathbb{R}^{n\times n}}\{2\mathbf{F}\cdot\mathbf{I}-\mathbf{F}\cdot(\mathbf{L}^e-\mathbf{L}_c)\mathbf{F}\}=\mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}\le\triangle c_*. \tag{A.3}$$

Choosing $\mathbf{F}$ with $\mathrm{Tr}(\mathbf{F})=\triangle c_*$, we see that $\mathbf{F}\cdot(\mathbf{L}^e-\mathbf{L}_c)\mathbf{F}\ge\triangle c_*=(\mathrm{Tr}\mathbf{F})^2/\triangle c_*$, which, by multiplying $\mathbf{F}$ by a such that $a\mathrm{Tr}(\mathbf{F})=\triangle c_*$, in fact holds for any $\mathbf{F}$ with $\mathrm{Tr}(\mathbf{F})\neq 0$. If $\mathrm{Tr}(\mathbf{F})=0$, the first bound in (2.18) is obvious. Further, $\mathbf{F}'=(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}$ is a maximizer of the l.h.s. of (A.3). Therefore, if the first bound in (A.1) holds as an equality, we have $\mathrm{Tr}(\mathbf{F}')=\mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}=\triangle c_*\neq 0$, and

$$\mathbf{F}'\cdot(\mathbf{L}^e-\mathbf{L}_c)\mathbf{F}'=\mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}=(\mathrm{Tr}\mathbf{F}')^2/\triangle c_*.$$

Thus, the first inequality in (2.18) holds as an equality for $a\mathbf{F}'$ with any $a\neq 0$, i.e., all $\mathbf{F}$ that satisfy (A.2).

Conversely, from the first bound in (2.18), choosing $\mathbf{F}=(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}$ we obtain the first bound in (A.1). Further, if for some $\mathbf{F}\in\mathcal{R}(\mathbf{L}^e-\mathbf{L}_c)$ with $\mathrm{Tr}(\mathbf{F})\neq 0$ the first bound in (2.18) holds as an equality, we have

$$\sup_{\mathbf{P}^0\in\mathbb{R}^{n\times n}}\{2\mathbf{P}^0\cdot\mathbf{F}-\mathbf{P}^0\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{P}^0\}=\mathbf{F}\cdot(\mathbf{L}^e-\mathbf{L}_c)\mathbf{F}=(\mathrm{Tr}\mathbf{F})^2/\triangle c_*.$$

Choosing $\mathbf{P}^0=\mathrm{Tr}(\mathbf{F})\mathbf{I}/\triangle c_*$ we have

$$\frac{2\mathrm{Tr}(\mathbf{F})^2}{\triangle c_*}-\frac{\mathrm{Tr}(\mathbf{F})^2}{\triangle c_*^2}\mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}\le(\mathrm{Tr}\mathbf{F})^2/\triangle c_*,$$

and hence $\mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}\ge\triangle c_*$, which, together with the first bound in (A.1), implies that $\mathbf{I}\cdot(\mathbf{L}^e-\mathbf{L}_c)^{-1}\mathbf{I}=\triangle c_*$, and that $\mathbf{P}^0=\mathrm{Tr}(\mathbf{F})\mathbf{I}/\triangle c_*\in\mathcal{R}(\mathbf{L}^e-\mathbf{L}_c)$ is in fact a maximizer of the l.h.s. of (A.4). On the other hand, the maximization problem in (A.4) admits the unique maximizer $(\mathbf{L}^e-\mathbf{L}_c)\mathbf{F}$ in $\mathcal{R}(\mathbf{L}^e-\mathbf{L}_c)$, which then implies (A.2). Thus, we complete the proof of Theorem A.1 for the case $\mathbf{L}(\mathcal{O},\mathbf{x})\ge\mathbf{L}_c$.

The case $\mathbf{L}_c\ge\mathbf{L}(\mathcal{O},\mathbf{x})$ can be handled similarly and will not be repeated here. □

I thank Kaushik Bhattacharya for valuable comments. I also gratefully acknowledge the financial support of the US Office of Naval Research through the MURI grant N00014-06-1-0730 and the startup funds from the University of Houston.

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