PERGAMON

International Journal of Solids and Structures 38 (2001) 7945–7965

INTERNATIONAL JOURNAL OF SOLIDS and STRUCTURES

www.elsevier.com/locate/ijsolstr

On optimal zeroth-order bounds with application to Hashin–Shtrikman bounds and anisotropy parameters

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Received 29 June 1998

Abstract

A new result presented in this paper is the evaluation of the Hashin–Shtrikman bounds for composites composed of arbitrarily anisotropic constituents. To date, evaluation of the Hashin–Shtrikman bounds are limited to composites with isotropic constituents or to polycrystalline composites with specific crystal symmetries. The generality of the exact result presented herein is achieved through a reinterpretation of Kröner's (J. Mech. Phys. Solids 25 (1977) 137) recursive relations for nth-order bounds and the optimal zeroth-order ($n = 0$) bound. The definitions of optimal zeroth-order bounds are extended to all even-ordered tensors and procedures are presented to evaluate these bounds for all second- and fourth-order tensors. While optimal zeroth-order bounds are not new, the ability to calculate them for fourth-order tensors of arbitrary symmetry is new. Utilizing the zeroth-order bounds, material anisotropy parameters are defined which quantify the extent of anisotropy for even-ordered tensors.

Keywords: Zeroth-order bounds; Hashin–Shtrikman bounds; Material anisotropy parameters

1. Introduction

Bounds are of interest in both experimental and theoretical work as a tool for validating or invalidating data and theory. In the field of effective properties of composite materials two orders of bounds are widely utilized: the first-order bounds, which are more commonly referred to as the Voigt/Reuss bounds (Voigt, 1928; Reuss, 1929; Hill, 1952; Paul, 1960; Milton and Kohn, 1988), and second-order bounds, which are more commonly known as Hashin–Shtrikman bounds (Hashin and Shtrikman, 1962a,b,c, 1963; Milton and Kohn, 1988; Kröner, 1977). To date, explicit expressions in the literature for the Hashin–Shtrikman

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PII: S0020-7683(00)00393-0

bounds are limited to composites consisting of an arbitrary number of *isotropic* constituents (Hashin and Shtrikman, 1963). For polycrystals with specific crystal symmetry (Watt and Peselnick, 1980) have prescribed procedures for evaluating the Hashin–Shtrikman bounds on the effective elastic properties. For more general material symmetry the procedures presented in this paper are required to evaluate the Hashin–Shtrikman bounds.

This capability is achieved through a reinterpretation of Kröner's (1977) nth-order bounds. Kröner presents recursive expressions for the evaluation of nth-order bounds on the effective elastic moduli for macroscopically homogeneous and isotropic composites in terms of the bounds of order $n-2$ for $n \geqslant 2$. A bound of nth order assumes homogeneity and isotropy of the correlation functions describing the spatial distribution of the constituents up to and including order n. Thus, every time Kröner's recursive relation is utilized to yield nth-order bounds from the $n-2$ order bounds there is the implied assumption of homogeneity and isotropy of the n and $n-1$ correlation functions. To evaluate these nth-order bounds, two "seeds" are required to initiate the recursive relations. These seeds are the zeroth- and first-order bounds. The odd-order bounds can be calculated from the first-order ($n = 1$) Voigt/Reuss bounds. The calculation of the even-order bounds (which include the Hashin–Shtrikman $n = 2$ bounds) require the zeroth-order ($n = 0$) bounds. In this paper we specifically address what are zeroth-order bounds and how to calculate them. While zeroth-order bounds are not new, the ability to evaluate them for fourth-order tensors of arbitrary symmetry is new.

Zeroth-order bounds are not novel. Optimal zeroth-order bounds have been previously defined and evaluated for a stiffness tensor with cubic material symmetry (Kröner, 1977). For the cubic (and thus isotropic) material symmetry group, the calculation of the optimal zeroth-order bounds is trivial. However, for more general classes of material symmetry it has been unknown how to evaluate these optimal zeroth-order bounds for fourth-order tensors.

Procedures are presented herein for calculating zeroth-order bounds for second- and fourth-order tensors. The ability to calculate these bounds for individual constituents of arbitrary anisotropy is novel and is significant for at least the following three reasons. First, zeroth-order bounds are rigorous bounds. Second, they permit calculation of all higher-order even-ordered bounds, including the Hashin–Shtrikman bounds, (see Section 6) for an arbitrary composite so long as it is macroscopically homogeneous and macroscopically isotropic. That is the constituents of the composite can be of *arbitrary* anisotropy. Third, they provide a means by which we quantify the extent of a material's anisotropy (see Section 8).

This paper is outlined as follows. In Section 2 the necessary mathematical notation is presented. The notation is fairly general to accommodate the definition of optimal zeroth-order bounds for all even-ordered tensors given in Section 3. Also presented in Section 3 are five theorems which are proven in Appendix A. The purpose for this formal presentation is to simplify the derivation of results in the remainder of the paper. Section 3 concludes with Kröner's definition of an optimal zeroth-order bound – which is only applicable to fourth-order tensors with no less than cubic symmetry. Sections 4 and 5 present derivations for the evaluation of the optimal zeroth-order bounds for second- and fourth-order tensors, respectively. These optimal zeroth-order bounds serve as the even "seed" for the recursive relations presented in Section 6 for the nth-order bounds. The odd "seed" is the Voigt/Reuss bound. In particular, Kröner's nth-order bounds are reinterpreted and presented in terms of Wu's tensor. These results are presented for both second- and fourth-order tensors. In Section 7 an example is presented which evaluates the bounds for $n = 0, 1, 2$ and ∞ for a graphite uranium dioxide composite using the methodology presented in Sections 5 and 6. Section 8 utilizes the optimal zeroth-order bounds to define anisotropy parameters to quantify the extent of anisotropy of a tensor. There is a single anisotropy parameter for second-order tensors and, in general, three anisotropy parameters for a fourth-order tensor. However, since our particular application to fourth-order tensors is the elastic modulus, the number of non-trivial anisotropy parameters is two – the minor symmetries of the elastic stiffness result in the third anisotropy parameter being zero implying perfect isotropy. Summary remarks are made in Section 9.

2. Preliminaries

Let $\mathscr{T}^m$ denote the set of order m tensors. Let $\boldsymbol{U} \in \mathscr{T}^n$ and $\boldsymbol{V} \in \mathscr{T}^m$ where $m, n \geqslant 1$ are not, in general, equal. We now define the following contraction operations:

$$\boldsymbol{U}\,\boldsymbol{V} = U_{i_1 \ldots i_{n-1} k}\, V_{k j_2 \ldots j_m}\, \boldsymbol{e}_{i_1} \otimes \cdots \otimes \boldsymbol{e}_{i_{n-1}} \otimes \boldsymbol{e}_{j_2} \otimes \cdots \otimes \boldsymbol{e}_{j_m}, \tag{1}$$

$$\boldsymbol{U} : \boldsymbol{V} = U_{i_1 \ldots i_{n-2} k_1 k_2}\, V_{k_1 k_2 j_3 \ldots j_m}\, \boldsymbol{e}_{i_1} \otimes \cdots \otimes \boldsymbol{e}_{i_{i-2}} \otimes \boldsymbol{e}_{j_3} \otimes \cdots \otimes \boldsymbol{e}_{j_m}, \tag{2}$$

$$\boldsymbol{U} \cdot \boldsymbol{V} = \begin{cases} U_{k_1 \ldots k_n}\, V_{k_1 \ldots k_n j_{n+1} \ldots j_m}\, \boldsymbol{e}_{j_{n+1}} \otimes \cdots \otimes \boldsymbol{e}_{j_m} & \text{if } n < m, \\ U_{k_1 \ldots k_n}\, V_{k_1 \ldots k_n} & \text{if } n = m, \\ U_{i_1 \ldots i_{n-m} k_1 \ldots k_m}\, V_{k_1 \ldots k_m}\, \boldsymbol{e}_{i_1} \otimes \cdots \otimes \boldsymbol{e}_{i_{n-m}} & \text{if } n > m, \end{cases} \tag{3}$$

The transpose of a tensor $\boldsymbol{T} \in \mathscr{T}^{2n}$ is a tensor $\boldsymbol{T}^T \in \mathscr{T}^{2n}$ defined by

$$\boldsymbol{w} \cdot \boldsymbol{T} \cdot \boldsymbol{v} = \boldsymbol{v} \cdot \boldsymbol{T}^T \cdot \boldsymbol{w} \quad \forall\, \boldsymbol{v}, \boldsymbol{w} \in \mathscr{T}^n \tag{4}$$

or, in component form, $T^T_{i_1 \ldots i_n j_1 \ldots j_n} = T_{j_1 \ldots j_n i_1 \ldots i_n}$. The symmetric and skew-symmetric operators are defined as $\boldsymbol{T}^S := \frac{1}{2}(\boldsymbol{T} + \boldsymbol{T}^T)$ and $\boldsymbol{T}^A := \frac{1}{2}(\boldsymbol{T} - \boldsymbol{T}^T)$, respectively. Let $\mathscr{T}^{2n}_{\mathrm{S}}$ and $\mathscr{T}^{2n}_{\mathrm{A}}$ denote the sets of symmetric and skew-symmetric order $2n$ tensors, respectively.

For $\boldsymbol{T} \in \mathscr{T}^{4n}_{\mathrm{S}}$ (note that $\boldsymbol{T}^T = \boldsymbol{T}$) with components $T_{i_1 \ldots i_{4n}}$ with respect to the basis $\{\boldsymbol{e}_i\}$ we define the following operators:

$$\boldsymbol{T}^s := \tfrac{1}{2}(T_{i_1 \ldots i_n i_{n+1} \ldots i_{2n} i_{2n+1} \ldots \ldots i_{4n}} + T_{i_{n+1} \ldots i_{2n} i_1 \ldots i_n i_{2n+1} \ldots \ldots i_{4n}})\, \boldsymbol{e}_{i_1} \otimes \cdots \otimes \boldsymbol{e}_{i_{4n}}, \tag{5}$$

$$\boldsymbol{T}^a := \tfrac{1}{2}(T_{i_1 \ldots i_n i_{n+1} \ldots i_{2n} i_{2n+1} \ldots \ldots i_{4n}} - T_{i_{n+1} \ldots i_{2n} i_1 \ldots i_n i_{2n+1} \ldots \ldots i_{4n}})\, \boldsymbol{e}_{i_1} \otimes \cdots \otimes \boldsymbol{e}_{i_{4n}}, \tag{6}$$

which yield the symmetric and skew-symmetric parts of $\boldsymbol{T}$ with respect to the first $2n$ indices. Since $\boldsymbol{T}^T = \boldsymbol{T}$, these operators also yield the symmetric and skew-symmetric parts of $\boldsymbol{T}$ with respect to the last $2n$ indices.

The trace operator tr applied to $\boldsymbol{T} \in \mathscr{T}^{2n}$ with components $T_{i_1 \ldots i_{2n}}$ with respect to a basis $\{\boldsymbol{e}_i\}$ is defined as $\mathrm{tr}\,\boldsymbol{T} = T_{i_1 \ldots i_n i_1 \ldots i_n}$ which is an invariant. When applied to a second-order tensor $\boldsymbol{u} \in \mathscr{T}^2$ and a fourth-order tensor $\boldsymbol{U} \in \mathscr{T}^4$, the trace operator yields the standard results: $\mathrm{tr}\,\boldsymbol{u} = u_{ii}$ and $\mathrm{tr}\,\boldsymbol{U} = U_{ijij}$.

For $\boldsymbol{t} \in \mathscr{T}^2$ we define the spherical and deviatoric operators, respectively, as $\boldsymbol{t}^{sph} := \frac{1}{3}(\mathrm{tr}\,\boldsymbol{t})\,\boldsymbol{i}$ and $\boldsymbol{t}^{dev} := \boldsymbol{t} - \boldsymbol{t}^{sph}$ where $\boldsymbol{i}$ is the second-order identity tensor.

We now define the operation $\boldsymbol{\Pi}_q[\boldsymbol{T}]$ which yields a new tensor as a result of transforming the body on which the tensor $\boldsymbol{T}$ is defined according to the tensor $\boldsymbol{q} \in \mathscr{O}$ where $\mathscr{O}$ is the set of second-order orthogonal tensors. For $\boldsymbol{T} \in \mathscr{T}^n$, with components $T_{i_1 i_2 \ldots i_n}$ with respect to the basis $\{\boldsymbol{e}_i\}$, the operation $\boldsymbol{\Pi}_q[\boldsymbol{T}]$ yields

$$\boldsymbol{\Pi}_q[\boldsymbol{T}] := \Pi_{q^{\mathrm{T}}}[T_{i_1 i_2 \ldots i_n}]\, \boldsymbol{e}_{i_1} \otimes \boldsymbol{e}_{i_2} \otimes \cdots \otimes \boldsymbol{e}_{i_n}, \tag{7}$$

where

$$\Pi_q[T_{i_1 i_2 \ldots i_n}] := q_{i_1 j_1}\, q_{i_2 j_2} \ldots q_{i_n j_n}\, T_{j_1 j_2 \ldots j_n}. \tag{8}$$

The volume average of a tensor $\boldsymbol{T}(\boldsymbol{x}) \in \mathscr{T}^n$ will be denoted by $\langle \boldsymbol{T} \rangle$.

For fourth-order identity tensors we have the following definitions:

$$\boldsymbol{I} := \delta_{ik}\, \delta_{jl}\, \boldsymbol{e}_i \otimes \boldsymbol{e}_j \otimes \boldsymbol{e}_k \otimes \boldsymbol{e}_l, \tag{9}$$

$$\boldsymbol{I}^{\mathrm{s}} := \boldsymbol{I}^s = \tfrac{1}{2}(\delta_{ik}\, \delta_{jl} + \delta_{il}\, \delta_{jk})\, \boldsymbol{e}_i \otimes \boldsymbol{e}_j \otimes \boldsymbol{e}_k \otimes \boldsymbol{e}_l, \tag{10}$$

$$\boldsymbol{I}^{\mathrm{a}} := \boldsymbol{I}^a = \tfrac{1}{2}(\delta_{ik}\, \delta_{jl} - \delta_{il}\, \delta_{jk})\, \boldsymbol{e}_i \otimes \boldsymbol{e}_j \otimes \boldsymbol{e}_k \otimes \boldsymbol{e}_l, \tag{11}$$

$$\boldsymbol{I}^{\text{sph}} := \tfrac{1}{3}\delta_{ij}\,\delta_{kl}\,\boldsymbol{e}_i \otimes \boldsymbol{e}_j \otimes \boldsymbol{e}_k \otimes \boldsymbol{e}_l, \tag{12}$$

$$\boldsymbol{I}^{\text{dev}} := (\delta_{ik}\,\delta_{jl} - \tfrac{1}{3}\delta_{ij}\,\delta_{kl})\,\boldsymbol{e}_i \otimes \boldsymbol{e}_j \otimes \boldsymbol{e}_k \otimes \boldsymbol{e}_l, \tag{13}$$

$$\boldsymbol{I}^{\text{dev,s}} := \boldsymbol{I}^{\text{dev}} : \boldsymbol{I}^{\text{s}} = \boldsymbol{I}^{\text{s}} : \boldsymbol{I}^{\text{dev}} = \boldsymbol{I}^{\text{s}} - \boldsymbol{I}^{\text{sph}}, \tag{14}$$

where δ_{ij} is the Kronecker delta.

The nth-order bounds on $\boldsymbol{T}$ are expressed as

$$\boldsymbol{T}^{n-} \leqslant \boldsymbol{T} \leqslant \boldsymbol{T}^{n+}, \tag{15}$$

where, for example, $\boldsymbol{T} \leqslant \boldsymbol{T}^{+}$ implies that

$$\boldsymbol{x} \cdot (\boldsymbol{T} - \boldsymbol{T}^{+}) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{n}. \tag{16}$$

In the remaining sections bounds on effective second- and fourth-order properties of a composite are presented. In this regard we make use of the following notation. Let $\boldsymbol{k}^{*} \in \mathscr{T}_{k}$ where $\mathscr{T}_{k}$ is the set of positive definite, symmetric second-order tensors and $\boldsymbol{C}^{*} \in \mathscr{T}_{\text{C}}$ where $\mathscr{T}_{\text{C}}$ is the set of positive definite, fourth-order tensors with both major and minor symmetries, denote effective properties to be bounded on the domain $\mathscr{R} \subseteq \Re^{3}$. The constitutive relation $\boldsymbol{k}(\boldsymbol{C})$ on $\mathscr{R}$ is assumed to be piecewise homogeneous thus permitting a partition of $\mathscr{R}$ denoted by $\{\mathscr{R}_r\}$ s.t. $\boldsymbol{k}^{r} := \boldsymbol{k}|\mathscr{R}_r$ ($\boldsymbol{C}^{r} := \boldsymbol{C}|\mathscr{R}_r$) is homogeneous. The ratio of the volume of $\mathscr{R}_r$ to the total volume of $\mathscr{R}$ is given by the rth phase volume fraction c_r.

3. Optimal zeroth-order bounds

In this section we define a zeroth-order bound to a tensor $\tilde{\boldsymbol{T}} \in \mathscr{T}^{2n}$ which is based on the relation (16). Since $\boldsymbol{x} \cdot \tilde{\boldsymbol{T}} \cdot \boldsymbol{x} \equiv \boldsymbol{x} \cdot (\tilde{\boldsymbol{T}}^{\text{S}}) \cdot \boldsymbol{x}$ it follows that a zeroth-order bound to a tensor $\tilde{\boldsymbol{T}}^{\text{S}}$ is a zeroth-order bound to the tensor $\tilde{\boldsymbol{T}}$. As a result – without any loss in generality – we shall only consider the zeroth-order bounds of a tensor $\boldsymbol{T} \in \mathscr{T}^{2n}_{\text{S}}$, where $\mathscr{T}^{2n}_{\text{S}}$ is the set of all symmetric (in the sense $\boldsymbol{T}^{T} = \boldsymbol{T}$) tensors.

A tensor $\boldsymbol{A} \in \mathscr{T}^{2n}_{\text{S}}$ is a zeroth-order *upper* bound to $\boldsymbol{T} \in \mathscr{T}^{2n}_{\text{S}}$ if

$$\boldsymbol{x} \cdot (\boldsymbol{\Pi}_q[\boldsymbol{T}] - \boldsymbol{A}) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{n} \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^{+} \tag{17}$$

or, equivalently, if

$$\boldsymbol{x} \cdot (\boldsymbol{T} - \boldsymbol{\Pi}_q[\boldsymbol{A}]) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{n} \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^{+}, \tag{18}$$

where $\mathscr{O}^{+}$ is the set of proper orthogonal, second-order tensors.

Let $\mathscr{A}^{+}$ denote the set of all zeroth-order upper bounds to $\boldsymbol{T} \in \mathscr{T}^{2n}_{\text{S}}$. The optimal zeroth-order upper bound, denoted by $\boldsymbol{T}^{0+}$, to a tensor $\boldsymbol{T} \in \mathscr{T}^{2n}_{\text{S}}$ is the element of $\mathscr{A}^{+}$ such that

$$\boldsymbol{x} \cdot (\boldsymbol{T}^{0+} - \boldsymbol{A}) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{n} \text{ and } \forall \boldsymbol{A} \in \mathscr{A}^{+}. \tag{19}$$

Since all proper orthogonal transformations of $\boldsymbol{A}$ are also upper bounds (see Eq. (18)), we can rewrite Eq. (19) as

$$\boldsymbol{x} \cdot (\boldsymbol{T}^{0+} - \boldsymbol{\Pi}_q[\boldsymbol{A}]) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{n},\ \forall \boldsymbol{A} \in \mathscr{A}^{+} \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^{+} \tag{20}$$

or, equivalently, as

$$\boldsymbol{x} \cdot (\boldsymbol{\Pi}_q[\boldsymbol{T}^{0+}] - \boldsymbol{A}) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{n},\ \forall \boldsymbol{A} \in \mathscr{A}^{+} \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^{+}. \tag{21}$$

In a similar manner the lower bound may be defined by replacing $\leqslant$ with $\geqslant$ and $\mathscr{A}^{+}$ with $\mathscr{A}^{-}$ in Eqs. (17)–(21), where $\mathscr{A}^{-}$ is the set of all zeroth-order lower bounds to $\boldsymbol{T}$.

In order to simplify the presentation of Sections 4 and 5 we state here the following six theorems whose proofs are provided in Appendix A. The first theorem states that the optimal zeroth-order bounds for a tensor $\boldsymbol{T} \in \mathscr{T}_{\mathrm{S}}^{2n}$ are isotropic. The special case of a fourth-order tensor $\boldsymbol{T} \in \mathscr{T}_{\mathrm{C}}$ has been previously proven (Kröner, 1977, Section 4).

Theorem 1. *The optimal zeroth-order bounds* $\boldsymbol{T}^{0+}$, $\boldsymbol{T}^{0-} \in \mathscr{T}_{S}^{2n}$, *to a tensor* $\boldsymbol{T} \in \mathscr{T}_{S}^{2n}$, *are isotropic.*

The second and third theorems state properties of the maximum and minimum eigenvalues of the differences $\boldsymbol{T} - \boldsymbol{T}^{0+}$ and $\boldsymbol{T} - \boldsymbol{T}^{0-}$, respectively.

Theorem 2. *For* $\boldsymbol{T}$, $\boldsymbol{T}^{0+} \in \mathscr{T}_{S}^{2n}$, $\boldsymbol{T}^{0+}$ *being the optimal zeroth-order upper bound to* $\boldsymbol{T}$, *the largest eigenvalue of the difference* $\boldsymbol{T} - \boldsymbol{T}^{0+}$ *is zero.*

Theorem 3. *For* $\boldsymbol{T}$, $\boldsymbol{T}^{0-} \in \mathscr{T}_{S}^{2n}$, $\boldsymbol{T}^{0+}$ *being the optimal zeroth-order lower bound to* $\boldsymbol{T}$, *the smallest eigenvalue of the difference* $\boldsymbol{T} - \boldsymbol{T}^{0-}$ *is zero.*

The remaining three theorems are elementary but they are presented in this formal manner to ease the presentation in the following sections. The fourth and fifth theorems are beneficial for the evaluation of optimal zeroth-order bounds. They decompose the equation $\boldsymbol{x} \cdot \boldsymbol{A} \cdot \boldsymbol{x} \leqslant (\geqslant) 0$ into a set of uncoupled equations.

Theorem 4. *For* $\boldsymbol{A} \in \mathscr{T}_{\mathrm{S}}^{4n}$, *the expression* $\boldsymbol{x} \cdot \boldsymbol{A} \cdot \boldsymbol{x} \leqslant (\geqslant) 0$, $\forall \boldsymbol{x} \in \mathscr{T}^{2n}$ *holds iff* $\boldsymbol{x} \cdot \boldsymbol{A}^{s} \cdot \boldsymbol{x} \leqslant (\geqslant) 0$, $\forall \boldsymbol{x} \in \mathscr{T}_{S}^{2n}$ *and* $\boldsymbol{x} \cdot \boldsymbol{A}^{a} \cdot \boldsymbol{x} \leqslant (\geqslant) 0$, $\forall \boldsymbol{x} \in \mathscr{T}_{A}^{2n}$.

Theorem 5. *For* $\boldsymbol{A} \in \mathscr{T}_{S}^{4}$ *with a spherical eigentensor, the expression* $\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} \leqslant (\geqslant) 0$, $\forall \boldsymbol{x} \in \mathscr{T}^{2}$, *holds iff* $\boldsymbol{x}^{sph} : \boldsymbol{A} : \boldsymbol{x}^{sph} \leqslant (\geqslant) 0$, $\forall \boldsymbol{x} \in \mathscr{T}^{2}$, *and* $\boldsymbol{x}^{dev} : \boldsymbol{A} : \boldsymbol{x}^{dev} \leqslant (\geqslant) 0$, $\forall \boldsymbol{x} \in \mathscr{T}^{2}$.

The final theorem relates the traces of two $2n$-order tensors given that the difference of the two tensors is, say, negative semi-definite.

Theorem 6. *For* $\boldsymbol{A}, \boldsymbol{B} \in \mathscr{T}^{2n}$, $\boldsymbol{x} \cdot (\boldsymbol{A} - \boldsymbol{B}) \cdot \boldsymbol{x} \leqslant 0$, $\forall \boldsymbol{x} \in \mathscr{T}^{n}$ *implies that* $\operatorname{tr} \boldsymbol{A} \leqslant \operatorname{tr} \boldsymbol{B}$.

In the remainder of this section Knöner's definition of an optimal zeroth-order bound is presented. As will be seen, it is a complete definition only for fourth-order tensors with a spherical eigentensor. The definitions given above are valid for all even ordered tensors regardless of material symmetry.

3.1. Kröner's definition

Let $\boldsymbol{T} \in \mathscr{T}_{\mathrm{C}}$ where $\mathscr{T}_{\mathrm{C}}$ is the set of positive definite, fourth-order tensors with the symmetries $T_{jikl} = T_{ijkl} = T_{klij} = T_{lkij}$. Kröner (1977, Section 4) defines the optimal zeroth-order upper bound – characterized by the two components T_{1111}^{0+} and T_{2323}^{0+} – to the tensor $\boldsymbol{T}$ by T_{1111}^{0+} and T_{2323}^{0+} being the maximum values attained by T_{1111} and T_{2323} as $\boldsymbol{T}$ undergoes an arbitrary coordinate transformation. In other words, letting $T_{ijkl}^{q} = \Pi_{q}[T_{ijkl}]$ for $\boldsymbol{q} \in \mathscr{O}^{+}$ the optimal upper bound, as defined by Kröner, is given by

$$T_{1111}^{0+} = \max_{q \in \mathscr{O}^{+}} T_{1111}^{q}, \qquad T_{2323}^{0+} = \max_{q \in \mathscr{O}^{+}} T_{2323}^{q}. \tag{22}$$

Certainly, it must be true that

$$T_{1111}^{0+} \geqslant \max_{q \in \mathscr{O}^{+}} T_{1111}^{q}, \qquad T_{2323}^{0+} \geqslant \max_{q \in \mathscr{O}^{+}} T_{2323}^{q}, \tag{23}$$

however, it is not obvious that Eq. (22) is sufficient to ensure that $\boldsymbol{T}^{0+}$ is an upper bound, i.e., that Eq. (18) holds with $\boldsymbol{A} = \boldsymbol{T}^{0+}$. Kröner does not provide a proof of sufficiency.

It can be shown that if $\boldsymbol{T}$ possesses a spherical eigentensor then Eq. (22) is sufficient for determining the optimal upper bound. Recall that if $\boldsymbol{T}$ has cubic or isotropic symmetry then $\boldsymbol{T}$ has a spherical eigentensor. For more general classes of symmetry, however, the sufficiency of Eq. (22) is not known.

4. Calculation of second-order optimal bounds

Let $\boldsymbol{t} \in \mathscr{T}^2_{\mathrm{S}}$ be a tensor for which optimal zeroth-order bounds are to be calculated. Let $(\lambda_i, \boldsymbol{v}_i)$, $i \in \{1,2,3\}$, denote the three eigenvalue–eigentensor pairs of the tensor $\boldsymbol{t}$ and define $\lambda_{\max} := \max_i \lambda_i$ and $\lambda_{\min} := \min_i \lambda_i$. From Theorem 1 the upper and lower optimal bounds of $\boldsymbol{t}$, $\boldsymbol{t}^{0+}$ and $\boldsymbol{t}^{0-}$, are isotropic. Thus, the optimal bounds may be represented in the form $\boldsymbol{t}^{0+} = t^{0+}\boldsymbol{i}$ and $\boldsymbol{t}^{0-} = t^{0-}\boldsymbol{i}$ where t^{0+}, $t^{0-} \in \Re$.

Consider the eigenanalysis of $\boldsymbol{t} - \boldsymbol{t}^{0+}$:

$$[\boldsymbol{t} - (\bar{\lambda}_i + t^{0+})\boldsymbol{i}]\bar{\boldsymbol{v}}_i = \boldsymbol{0}, \tag{24}$$

where $(\bar{\lambda}_i, \bar{\boldsymbol{v}}_i)$, $i \in \{1,2,3\}$ are the three eigenvalue–eigentensor pairs of the difference $\boldsymbol{t} - \boldsymbol{t}^{0+}$. It is easily shown that $(\bar{\lambda}_i + t^{0+}, \bar{\boldsymbol{v}}_i) = (\lambda_i, \boldsymbol{v}_i)$, thus, $\bar{\lambda} = \lambda - t^{0+}$. Taking the max of both sides gives $\bar{\lambda}_{\max} = \lambda_{\max} - t^{0+}$. From Theorem 2 the largest eigenvalue of $\boldsymbol{t} - \boldsymbol{t}^{0+}$ is zero; that is, $\bar{\lambda}_{\max} = 0$. Thus,

$$t^{0+} = \lambda_{\max} := \{\text{largest eigenvalue of } \boldsymbol{t}\}. \tag{25}$$

Similarly, using Theorem 3, it can be deduced that

$$t^{0-} = \lambda_{\min} := \{\text{smallest eigenvalue of } \boldsymbol{t}\}. \tag{26}$$

Since optimal zeroth-order bounds are isotropic (Theorem 1), the zeroth-order bounds for a composite are the two isotropic tensors which optimally bound the set of zeroth-order bounds of the constituents. By "optimally bound" it is meant that there do not exist other isotropic tensors which provide a better, or "tighter", zeroth-order bound. Therefore, the upper and lower zeroth-order bounds for a composite system are given by $\boldsymbol{k}^{0+} = k^{0+}\boldsymbol{i}$ and $\boldsymbol{k}^{0-} = k^{0-}\boldsymbol{i}$ where

$$k^{0+} := \max_r \lambda^r_{\max} \qquad k^{0-} := \min_r \lambda^r_{\min} \tag{27}$$

and $\lambda^r_{\max}$ and $\lambda^r_{\min}$ denote the maximum and minimum eigenvalues of $\boldsymbol{k}^r$, respectively.

5. Calculation of fourth-order optimal bounds

Let $\boldsymbol{T} \in \mathscr{T}^4_{\mathrm{S}}$ be a tensor for which optimal zeroth-order bounds are to be calculated. From Theorem 1 the upper and lower optimal bounds of $\boldsymbol{T}$: $\boldsymbol{T}^{0+}$ and $\boldsymbol{T}^{0-}$, are isotropic and may, thus, take the form

$$\boldsymbol{T}^{0+} = 3\kappa^{0+}\boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0+}\boldsymbol{I}^{\mathrm{dev,s}} + 2\eta^{0+}\boldsymbol{I}^{\mathrm{a}}, \tag{28}$$

$$\boldsymbol{T}^{0-} = 3\kappa^{0-}\boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0-}\boldsymbol{I}^{\mathrm{dev,s}} + 2\eta^{0-}\boldsymbol{I}^{\mathrm{a}}, \tag{29}$$

where $\kappa^{0+}, \mu^{0+}, \eta^{0+}, \kappa^{0-}, \mu^{0-}, \eta^{0-} \in \Re$.

We provide proofs for the evaluation of the upper bound exclusively. The evaluation of the lower bound proceeds in a similar manner and will not be presented below. However, final results for $\boldsymbol{T}^{0-}$ will be presented.

Restricting attention to fourth-order tensors when applying Theorem 4 to Eq. (18) with $\boldsymbol{A} = \boldsymbol{T}^{0+}$, and noting that $\boldsymbol{T}^{0+}$ is isotropic, yields the uncoupled conditions

$$\boldsymbol{x} : [\boldsymbol{T}^s - (\boldsymbol{T}^{0+})^s] : \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2_{\mathrm{S}}, \tag{30}$$

$$\boldsymbol{x} : [\boldsymbol{T}^a - (\boldsymbol{T}^{0+})^a] : \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2_{\mathrm{A}}, \tag{31}$$

where $(\boldsymbol{T}^{0+})^s = 3\kappa^{0+}\boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0+}\boldsymbol{I}^{\mathrm{dev,s}}$ and $(\boldsymbol{T}^{0+})^a = 2\eta^{0+}\boldsymbol{I}^{\mathrm{a}}$. Theorem 2 applies to the expression within brackets for both Eqs. (30) and (31).

5.1. Calculation of η^{0+}

We proceed to evaluate $(\boldsymbol{T}^{0+})^a$. Let $(\lambda_i, \boldsymbol{v}_i)$, $i \in \{1, 2, 3\}$, denote the three eigenvalue–eigentensor pairs of the tensor of $\boldsymbol{T}^a$, where $\lambda_i \in \Re$ and $\boldsymbol{v}_i \in \mathscr{T}^2_{\mathrm{A}}$. Define $\lambda_{\max} := \max_i \lambda_i$ and $\lambda_{\min} := \min_i \lambda_i$. We now consider the eigenanalysis of $\boldsymbol{T}^a - (\boldsymbol{T}^{0+})^a$ which takes the form

$$[\boldsymbol{T}^a - (\bar{\lambda}_i + 2\eta^{0+})\boldsymbol{I}^{\mathrm{a}}] : \bar{\boldsymbol{v}} = \boldsymbol{0}. \tag{32}$$

The solution of Eq. (32) is $(\bar{\lambda}_i + 2\eta^{0+}, \bar{\boldsymbol{v}}_i) = (\lambda_i, \boldsymbol{v}_i)$. Thus, $\bar{\lambda}_i = \lambda_i - 2\eta^{0+}$. Taking the max of both sides gives $\bar{\lambda}_{\max} = \lambda_{\max} - 2\eta^{0+}$. From Theorem 2 the largest eigenvalue of $\boldsymbol{T}^a - (\boldsymbol{T}^{0+})^a$ is zero. Thus, $\bar{\lambda}_{\max} = 0$ which leads to

$$2\eta^{0+} = \lambda_{\max} := \{\text{largest eigenvalue of } \boldsymbol{T}^a\}. \tag{33}$$

Proceeding in a similar manner for the optimal lower bound yields

$$2\eta^{0-} = \lambda_{\min} := \{\text{smallest eigenvalue of } \boldsymbol{T}^a\}. \tag{34}$$

If $\boldsymbol{T}$ has both minor diagonal symmetries, i.e., $\boldsymbol{T}^a = \boldsymbol{0}$, then $\eta^{0+} = \eta^{0-} = 0$.

5.2. Calculation of κ^{0+} and μ^{0+}

We now address the evaluation of $(\boldsymbol{T}^{0+})^s$. Let $(\lambda_i, \boldsymbol{v}_i)$, $i \in \{1, 2, \ldots, 6\}$ denote the six eigenvalue–eigentensor pairs of the tensor of $\boldsymbol{T}^s$ where $\lambda_i \in \Re$ and $\boldsymbol{v}_i \in \mathscr{T}^2_{\mathrm{S}}$. Define $\lambda_{\max} := \max_i \lambda_i$ and $\lambda_{\min} := \min_i \lambda_i$. Consider the eigenproblem

$$[\boldsymbol{T}^s - (\boldsymbol{T}^{0+})^s - \bar{\lambda}_i \boldsymbol{I}^{\mathrm{s}}] : \bar{\boldsymbol{v}}_i = \boldsymbol{0}, \tag{35}$$

whose solutions are given by the eigenvalue–eigentensor pairs $(\bar{\lambda}_i, \bar{\boldsymbol{v}}_i)$ for $i \in \{1, 2, \ldots, 6\}$.

The tensor $(\boldsymbol{T}^{0+})^{\mathrm{S}}$ is isotropic thus

$$(\boldsymbol{T}^{0+})^s \in \mathscr{A} := \{\boldsymbol{A} \mid \boldsymbol{A} = \alpha \boldsymbol{I}^{\mathrm{sph}} + \beta \boldsymbol{I}^{\mathrm{s}}, \quad \alpha, \beta \in \Re\}. \tag{36}$$

To further restrict the set $\mathscr{A}$ of possible optimal upper bounds, take $\boldsymbol{A} \in \mathscr{A}$ and consider the eigenproblem of $\boldsymbol{T}^s - \boldsymbol{A}$ which takes the form

$$[\boldsymbol{T}^s - \alpha \boldsymbol{I}^{\mathrm{sph}} - \tilde{\lambda}_i(\alpha)\boldsymbol{I}^{\mathrm{s}}] : \bar{\boldsymbol{v}}_i(\alpha) = \boldsymbol{0}, \tag{37}$$

where $\tilde{\lambda}(\alpha) := \bar{\lambda}(\alpha, \beta) + \beta$ is an eigenvalue of $\boldsymbol{T}^s - \alpha \boldsymbol{I}^{\mathrm{sph}}$ and $\bar{\lambda}(\alpha, \beta)$ is an eigenvalue of $\boldsymbol{T}^s - \boldsymbol{A}$. The beauty of Eq. (37) is that it is *not* a function of β! From Theorem 2 it is necessary that the maximum eigenvalue of $\boldsymbol{T}^s - \boldsymbol{A}$ be zero (i.e., $\bar{\lambda}_{\max}(\alpha, \beta) = 0$) in order for $\boldsymbol{A}$ to be an upper bound and more specifically a *potential* optimal upper bound. In other words, if $\bar{\lambda}_{\max}(\alpha, \beta) \neq 0$ then $\boldsymbol{A} \neq (\boldsymbol{T}^{0+})^s$.

The requirement that $\bar{\lambda}_{\max}(\alpha, \beta) = 0$ allows us to calculate β. Choose $\alpha \in \Re$. The eigenproblem (37) can then be solved for the eigenvalues $\tilde{\lambda}_i(\alpha)$ where $i \in \{1, 2, \ldots, 6\}$. Taking the max of both sides of the definition for $\tilde{\lambda}_i(\alpha)$ gives

$$\tilde{\lambda}_{\max}(\alpha) = \bar{\lambda}_{\max}(\alpha, \beta) + \beta. \tag{38}$$

After substituting $\bar{\lambda}_{\max}(\alpha, \beta) = 0$, one obtains

$$\beta = \tilde{\lambda}_{\max}(\alpha) := \{\text{largest eigenvalue of } \boldsymbol{T}^s - \alpha \boldsymbol{I}^{\text{sph}}, \quad \alpha \in \Re\}. \tag{39}$$

We now have that

$$(\boldsymbol{T}^{0+})^s \in \mathscr{A}' := \{\boldsymbol{A}' \mid \boldsymbol{A}' = \alpha \boldsymbol{I}^{\text{sph}} + \tilde{\lambda}_{\max}(\alpha)\boldsymbol{I}^s, \quad \alpha \in \Re\}. \tag{40}$$

It remains to determine which value of $\alpha \in \Re$ yields the optimal upper bound from the set $\mathscr{A}'$. From Theorem 6 it is concluded that the optimal upper bound is that element of $\mathscr{A}'$ which has the minimum trace. Here is the proof. Every $\boldsymbol{A}' \in \mathscr{A}'$ is a zeroth-order upper bound to $\boldsymbol{T}^s$ with the maximum eigenvalue of $\boldsymbol{T}^s - \boldsymbol{A}'$ equal to zero. By the definition of the optimal zeroth-order upper bound and Theorem 6, it follows that for any $\boldsymbol{A}' \in \mathscr{A}'$ for which there exists a $\boldsymbol{B}' \in \mathscr{A}'$ s.t. $\operatorname{tr} \boldsymbol{B}' < \operatorname{tr} \boldsymbol{A}'$ that $\boldsymbol{A}' \neq (\boldsymbol{T}^{0+})^s$. Since $\mathscr{A}'$ is a set parameterized by a single scalar variable – namely, α – it follows that the optimal zeroth-order upper bound is the element of $\mathscr{A}'$ with the smallest trace.

Letting

$$\tau_1(\alpha) := \operatorname{tr}(\alpha \boldsymbol{I}^{\text{sph}} + \tilde{\lambda}_{\max}(\alpha)\boldsymbol{I}^s) \tag{41}$$

$$\tau_1(\alpha) = \alpha + 6\,\tilde{\lambda}_{\max}(\alpha) \tag{42}$$

and defining $\alpha_{\min}$ to be the value of α which minimizes the function $\tau_1(\alpha)$: $\tau_1(\alpha_{\min}) = \min_{\alpha \in \Re} \tau_1(\alpha)$, the optimal zeroth-order upper bound is given by $(\boldsymbol{T}^{0+})^s = \alpha_{\min} \boldsymbol{I}^{\text{sph}} + \tilde{\lambda}_{\max}(\alpha_{\min})\boldsymbol{I}^s$ or, equivalently, by

$$2\mu^{0+} = \tilde{\lambda}_{\max}(\alpha_{\min}) := \{\text{largest eigenvalue of } \boldsymbol{T}^s - \alpha_{\min}\boldsymbol{I}^{\text{sph}}\}, \tag{43}$$

$$3\kappa^{0+} = \alpha_{\min} + \tilde{\lambda}_{\max}(\alpha_{\min}). \tag{44}$$

Proceeding in a similar manner for the optimal zeroth-order lower bound yields

$$2\mu^{0-} = \tilde{\lambda}_{\min}(\alpha_{\max}) := \{\text{smallest eigenvalue of } \boldsymbol{T}^s - \alpha_{\max}\boldsymbol{I}^{\text{sph}}\}, \tag{45}$$

$$3\kappa^{0-} = \alpha_{\max} + \tilde{\lambda}_{\min}(\alpha_{\max}), \tag{46}$$

where $\alpha_{\max}$ is the value of α which maximizes the function

$$\tau_2(\alpha) := \operatorname{tr}(\alpha \boldsymbol{I}^{\text{sph}} + \tilde{\lambda}_{\min}(\alpha)\boldsymbol{I}^s) \tag{47}$$

$$\tau_2(\alpha) = \alpha + 6\,\tilde{\lambda}_{\min}(\alpha). \tag{48}$$

That is, $\tau_2(\alpha_{\max}) = \max_{\alpha \in \Re} \tau_2(\alpha)$.

5.3. Special Case: spherical eigentensor

The evaluation of $(\boldsymbol{T}^{0+})^s$ with its two parameters is not difficult if $\boldsymbol{T}^s$ has a specific form – a form such that it has a spherical eigentensor $\lambda_{\text{sph}}\boldsymbol{i}$. Restricting attention to fourth-order tensors in applying Theorem 5 to Eq. (18) with $\boldsymbol{T} = \boldsymbol{T}^s$ and $\boldsymbol{A} = (\boldsymbol{T}^{0+})^s$, and noting that $(\boldsymbol{T}^{0+})^s$ is isotropic and thus has a spherical eigentensor, yields the uncoupled conditions

$$\boldsymbol{x}^{sph} : [\boldsymbol{T}^s - (\boldsymbol{T}^{0+})^s] : \boldsymbol{x}^{sph} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2, \tag{49}$$

$$\boldsymbol{x}^{dev} : [\boldsymbol{T}^s - (\boldsymbol{T}^{0+})^s] : \boldsymbol{x}^{dev} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2. \tag{50}$$

Theorem 2 applies to the expression within brackets of both Eqs. (49) and (50).

Since $\boldsymbol{T}^s$ has a spherical eigentensor it follows that $\bar{\boldsymbol{v}}_i = \boldsymbol{v}_i$, for $i \in \{1, 2, \ldots, 6\}$, are eigentensors of the difference $\boldsymbol{T}^s - (\boldsymbol{T}^{0+})^s$. Let λ_{sph} denote the eigenvalue of the spherical eigenmode $\boldsymbol{i}$ of $\boldsymbol{T}^s$. That is, $\exists i \in \{1, 2, \ldots, 6\}$ s.t. $\boldsymbol{v}_i \propto \boldsymbol{i}$. Let $(\hat{\lambda}_i, \hat{\boldsymbol{v}}_i)$, for $i \in \{1, 2, \ldots, 5\}$, denote the five non-spherical eigenvalue–eigentensor pairs of (λ_i, v_i). For the isotropic tensor $(\boldsymbol{T}^{0+})^s$ recall that $3\kappa^{0+}$ is the spherical eigenvalue and that $2\mu^{0+}$ is an eigenvalue of multiplicity five corresponding to the five non-spherical eigentensors.

We now consider Eq. (49) by substituting $\bar{\boldsymbol{v}}_i = \boldsymbol{i} \propto \boldsymbol{x}^{sph}$ into Eq. (35) to yield

$$(\lambda_{\mathrm{sph}} - 3\kappa^{0+} - \bar{\lambda})\boldsymbol{i} = \boldsymbol{0}. \tag{51}$$

Therefore $\bar{\lambda} = \lambda_{\mathrm{sph}} - 3\kappa^{0+}$. From Theorem 2 it is concluded that $\bar{\lambda} = 0$ and thus

$$3\kappa^{0+} = \lambda_{\mathrm{sph}} := \{\text{spherical eigenvalue of } \boldsymbol{T}^s\}. \tag{52}$$

We now consider the remaining Eq. (50) by substituting $\bar{\boldsymbol{v}}_i = \hat{\boldsymbol{v}}_i$ into Eq. (35) to yield $\bar{\lambda}_i = \hat{\lambda}_i - 2\mu^{0+}$. Taking the max of both sides gives $\bar{\lambda}_{\max} = \max_i \hat{\lambda}_i - 2\mu^{0+}$. From Theorem 2 it is concluded that $\bar{\lambda}_{\max} = 0$ and thus

$$2\mu^{0+} = \max_i \hat{\lambda}_i = \{\text{largest non-spherical eigenvalue of } \boldsymbol{T}^s\}. \tag{53}$$

Proceeding in a similar manner for the optimal zeroth-order lower bound yields

$$3\kappa^{0-} = \lambda_{\mathrm{sph}} := \{\text{spherical eigenvalue of } \boldsymbol{T}^s\} \tag{54}$$

$$= 3\kappa^{0+} \tag{55}$$

$$2\mu^{0-} = \min_i \tilde{\lambda}_i = \{\text{smallest non-spherical eigenvalue of } \boldsymbol{T}^s\}. \tag{56}$$

Isotropic or cubic symmetry of $\boldsymbol{T}^s$ is sufficient to ensure a spherical eigentensor. When $\boldsymbol{T}^s$ is isotropic, say of the form $\boldsymbol{T}^s = 3\kappa \boldsymbol{I}^{\mathrm{sph}} + 2\mu \boldsymbol{I}^{\mathrm{dev,s}}$, then $\kappa^{0+} = \kappa^{0-} = \kappa$ and $\mu^{0+} = \mu^{0-} = \mu$. When $\boldsymbol{T}^s$ has cubic symmetry then $\kappa^{0+} = \kappa^{0-} = \frac{1}{3}(T_{1111} + 2T_{1122})$, $\mu^{0+} = \max\{T_{2323}, \frac{1}{2}(T_{1111} - T_{1122})\}$ and $\mu^{0-} = \min\{T_{2323}, \frac{1}{2}(T_{1111} - T_{1122})\}$.

5.4. Composite system

Again, since optimal zeroth-order bounds are isotropic the zeroth-order bounds for a composite are the two isotropic tensors which optimally bound the set of zeroth-order bounds of the constituents. Note that for two isotropic fourth-order tensors: $\boldsymbol{A} = 3\kappa_A \boldsymbol{I}^{\mathrm{sph}} + 2\mu_A \boldsymbol{I}^{\mathrm{dev,s}}$ and $\boldsymbol{B} = 3\kappa_B \boldsymbol{I}^{\mathrm{sph}} + 2\mu_B \boldsymbol{I}^{\mathrm{dev,s}}$, that $\boldsymbol{A} \leqslant \boldsymbol{B}$ iff (i) $\kappa_A \leqslant \kappa_B$ and (ii) $\mu_A \leqslant \mu_B$. Thus, the upper and lower zeroth-order bounds for a composite system are given by

$$\boldsymbol{C}^{0+} = 3\kappa^{0+} \boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0+} \boldsymbol{I}^{\mathrm{dev,s}}, \tag{57}$$

$$\boldsymbol{C}^{0-} = 3\kappa^{0-} \boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0-} \boldsymbol{I}^{\mathrm{dev,s}}, \tag{58}$$

where

$$\kappa^{0+} := \max_r \kappa_r^{0+} \qquad \mu^{0+} := \max_r \mu_r^{0+}, \tag{59}$$

$$\kappa^{0-} := \min_r \kappa_r^{0-} \qquad \mu^{0-} := \min_r \mu_r^{0-} \tag{60}$$

and κ_r^{0+} and κ_r^{0-} (μ_r^{0+} and μ_r^{0-}) are the upper and lower zeroth-order bounds, respectively, of the bulk (shear) modulus for arbitrarily anisotropic $\boldsymbol{C}^r$. Note that $\boldsymbol{C}^{0+}$ and $\boldsymbol{C}^{0-}$ are *not* necessarily elements of the set of constituent zeroth-order bounds $\{(\boldsymbol{C}^r)^{0+}, (\boldsymbol{C}^r)^{0-}\}$.

6. *n*th order bounds

Kröner (1977) presents expressions for the nth-order bounds in terms of the bounds of order $n-2$. In obtaining bounds of order n from the bounds of order $n-2$ it is assumed that the correlation functions of order n and $n-1$ are homogeneous and isotropic. The bounds of order $n-2$ have already assumed homogeneity and isotropy of the correlation functions up to and including order $n-2$. Kröner's expressions are manipulated algebraically to yield tensorial expressions similar to the $\boldsymbol{T}$ (i.e., Wu's tensor) and $\boldsymbol{W}$ tensors (Benveniste, 1986) where the shape of the inclusions happen to be spherical. These results are presented below.

The evaluation of the following expressions are easily performed after mapping the tensorial expressions to equivalent matrical expressions (Nadeau and Ferrari, 1998). These mappings, however, are not trivial.

6.1. Second-order tensors

The nth-order bounds ($n \geqslant 2$) are given recursively by

$$\boldsymbol{k}^{n+} = \langle \boldsymbol{k}\boldsymbol{t}_{n-2}\rangle \langle \boldsymbol{t}_{n-2}\rangle^{-1}, \tag{61}$$

$$\boldsymbol{k}^{n-} = \left[\langle \boldsymbol{\rho}\boldsymbol{w}_{n-2}\rangle \langle \boldsymbol{w}_{n-2}\rangle^{-1}\right]^{-1}, \tag{62}$$

where

$$\boldsymbol{t}_{n-2} := \left[\boldsymbol{i} + \boldsymbol{p}_{n-2}(\boldsymbol{k} - \boldsymbol{k}^{(n-2)+})\right]^{-1}, \tag{63}$$

$$\boldsymbol{w}_{n-2} := \left[\boldsymbol{i} + \boldsymbol{q}_{n-2}(\boldsymbol{\rho} - \boldsymbol{\rho}^{(n-2)+})\right]^{-1}, \tag{64}$$

$$\boldsymbol{p}_{n-2} := \boldsymbol{e}(\boldsymbol{k}^{(n-2)+})^{-1} = \boldsymbol{e}\boldsymbol{\rho}^{(n-2)-}, \tag{65}$$

$$\boldsymbol{q}_{n-2} := (\boldsymbol{\rho}^{(n-2)+})^{-1}(\boldsymbol{i} - \boldsymbol{e}) = \boldsymbol{k}^{(n-2)-}(\boldsymbol{i} - \boldsymbol{e}), \tag{66}$$

and where $\boldsymbol{e} := (1/3)\boldsymbol{i}$. Note that $(1/3)\boldsymbol{i}$ also happens to be the second-order Eshelby tensor (Hatta and Taya, 1985) for a *spherical* inclusion. The tensors $\boldsymbol{k}^{0+}$ and $\boldsymbol{k}^{0-}$ are the zeroth-order upper and lower bounds on $\boldsymbol{k}^{*}$ for the composite (see Section 4). Note that $\boldsymbol{k}$ and $\boldsymbol{\rho}$ (and, thus, $\boldsymbol{t}_{(n-2)}$ and $\boldsymbol{w}_{(n-2)}$) are the only spatially variable quantities in Eqs. (61)–(66).

6.2. Fourth-order tensors

The nth-order bounds ($n \geqslant 2$) are given by

$$\boldsymbol{C}^{n+} = \langle \boldsymbol{C} : \boldsymbol{T}_{n-2}\rangle : \langle \boldsymbol{T}_{n-2}\rangle^{-1}, \tag{67}$$

$$\boldsymbol{C}^{n-} = \left[\langle \boldsymbol{S} : \boldsymbol{W}_{n-2}\rangle : \langle \boldsymbol{W}_{n-2}\rangle^{-1}\right]^{-1}, \tag{68}$$

where

$$\boldsymbol{T}_{n-2} := \left[\boldsymbol{I}^{\mathrm{s}} + \boldsymbol{P}_{n-2} : (\boldsymbol{C} - \boldsymbol{C}^{(n-2)+})\right]^{-1}, \tag{69}$$

$$\boldsymbol{W}_{n-2} := \left[\boldsymbol{I}^{\mathrm{s}} + \boldsymbol{Q}_{n-2} : (\boldsymbol{S} - \boldsymbol{S}^{(n-2)+})\right]^{-1}, \tag{70}$$

$$\boldsymbol{P}_{n-2} := \boldsymbol{E}_{(n-2)+} : (\boldsymbol{C}^{(n-2)+})^{-1} = \boldsymbol{E}_{(n-2)+} : \boldsymbol{S}^{(n-2)-}, \tag{71}$$

$$\boldsymbol{Q}_{n-2} := (\boldsymbol{S}^{(n-2)+})^{-1} : (\boldsymbol{I}^{s} - \boldsymbol{E}_{(n-2)-}) = \boldsymbol{C}^{(n-2)-} : (\boldsymbol{I}^{s} - \boldsymbol{E}_{(n-2)-}), \tag{72}$$

and where $\boldsymbol{C}^{(n-2)+}$ and $\boldsymbol{C}^{(n-2)-}$ are the upper and lower bounds of order $(n-2)$ on $\boldsymbol{C}^{*}$ for the composite and $\boldsymbol{E}_{(n-2)+}$ and $\boldsymbol{E}_{(n-2)-}$ are equivalent to the fourth-order Eshelby tensors (Eshelby, 1957; Mura, 1987) for a spherical inclusion in an isotropic matrix of constitution $\boldsymbol{C}^{(n-2)+}$ and $\boldsymbol{C}^{(n-2)-}$, respectively.

6.3. Hashin–Shtrikman bounds

Bounds of second-order for *macroscopically homogeneous and isotropic* effective properties are more commonly known as Hashin–Shtrikman bounds and they were arrived at by variational or minimum energy methods. These bounds are explicitly a function of the constitutive relations and volume fractions of the individual phases but they are *also* implicitly a function of the microstructure. Second-order bounds assume homogeneity and isotropy of the correlation functions up to and including order $n = 2$. If *only* constitutive relations and phase volume fractions are known then the appropriate bounds are of order $n = 1$. Despite statements to the contrary in the literature second-order bounds rely on additional microstructural information, namely, the homogeneity and isotropy of the correlation function of order $n = 2$.

6.3.1. Second-order tensors

The Hashin–Shtrikman bounds (Hashin and Shtrikman, 1962b; Milton and Kohn, 1988), given by Eqs. (61)–(66) when $n = 2$, are valid, and capable of evaluation, for *arbitrary* material constitution and number of constituents – so long as the effective properties are macroscopically homogeneous and isotropic. This is in distinction to expressions in the literature for Hashin–Shtrikman bounds which are limited to *isotropic* constituents for multi-constituent composites (Hashin and Shtrikman, 1962b).

It is of interest to note that under certain circumstances equivalence may be found between the Hatta–Taya effective property predictions (Hatta and Taya, 1985; Nadeau and Ferrari, 1995) and the Hashin–Shtrikman bounds. When a bi-constituent composite in the form of an isotropic matrix reinforced with spherical fibers which have a uniform ODF and the matrix material constitution, in addition to being isotropic, is an optimal zeroth-order bound of the composite then the prediction of Hatta–Taya corresponds to one of the two Hashin–Shtrikman bounds. In particular, if $\boldsymbol{k}^{m} = \boldsymbol{k}^{0+}$ then the Hatta–Taya approximation is equivalent to the Hashin–Shtrikman upper bound and if $\boldsymbol{k}^{m} = \boldsymbol{k}^{0-}$ then the Hatta–Taya approximation is equivalent to the Hashin–Shtrikman lower bound. This correspondence has been previously observed (Benveniste, 1986) for multi-phase composites with *isotropic* constituents.

6.3.2. Fourth-order tensors

The Hashin–Shtrikman bounds (Hashin and Shtrikman, 1962a,c, 1963), given by Eqs. (67)–(72) when $n = 2$, are valid, and capable of evaluation, for *arbitrary* material constitution and number of constituents – so long as the effective properties are macroscopically homogeneous and isotropic. This is in distinction to expressions in the literature which are limited to *isotropic* constituents for multi-constituent composites (Hashin and Shtrikman, 1963) and to cubic (Hashin and Shtrikman, 1962c) and hexagonal, trigonal and tetragonal (Watt and Peselnick, 1980) single crystal symmetries for polycrystals.

It is of interest to note that under certain circumstances equivalence may be found between the Mori–Tanaka effective property predictions (Mori and Tanaka, 1973; Benveniste, 1987) and the Hashin–Shtrikman bounds. When a bi-constituent composite in the form of an isotropic matrix reinforced with anisotropic, spherical fibers which have a uniform ODF, and the matrix material constitution, in addition to being isotropic, is an optimal zeroth-order bound of the composite, then the Mori–Tanaka prediction corresponds to one of the two Hashin–Shtrikman bounds. In particular, if $\boldsymbol{C}^{m} = \boldsymbol{C}^{0+}$ then the

Mori–Tanaka approximation is equivalent to the Hashin–Shtrikman upper bound and if $\boldsymbol{C}^{\mathrm{m}} = \boldsymbol{C}^{0-}$ then the Mori–Tanaka approximation is equivalent to the Hashin–Shtrikman lower bound. This correspondence has been previously observed for two-phase (Weng, 1984) and multi-phase (Norris, 1989) composites with isotropic constituents and for the general case of multi-constituent composites with anisotropic constituents (Weng, 1990).

7. Bounds on a graphite–uranium dioxide composite

In this section, utilizing the developments presented above, bounds corresponding to $n = 0,\ 1,\ 2$ and ∞ are presented for the elastic bulk κ and shear μ moduli for a macroscopically isotropic graphite–uranium dioxide (C–UO_2) composite. Of particular interest in this example is the calculation of the Hashin–Shtrikman bounds for the composite because each of the individual constituents are anisotropic. The Hashin–Shtrikman bound calculation (as well as higher order bound calculations) is made possible due to the previous developments in Sections 5 and 6. This composite is used in nuclear reactor fuel rods and it is assumed that all crystals are randomly oriented.

Single crystal graphite has hexagonal symmetry and its five independent elastic moduli (Kelly, 1981, Table 3.2, p. 74) are presented in Table 1. Single crystal uranium dioxide has cubic symmetry and its three independent elastic moduli (Simmons and Wang, 1971, code = 12076) are presented in Table 2. Bounds on the bulk κ and shear μ moduli of polycrystalline graphite and uranium dioxide are tabulated in Table 3. This table was compiled by first evaluating the optimal zeroth-order bounds utilizing Section 5, then using the results of Section 6 to compute the bounds corresponding to $n = 2, 4, 6, \ldots, \infty$. Second, the first-order Voigt/Reuss bounds were evaluated and then the relations of Section 6 could be used again to compute the bounds corresponding to $n = 3, 5, 7, \ldots, \infty$.

The bounds for a macroscopically isotropic graphite–uranium dioxide composite are now presented. Plots of the results are in the form of modulus versus volume fraction α of uranium dioxide. The bounds on the bulk modulus are presented in Fig. 1 while the bounds on the shear modulus are presented in Fig. 2.

Table 1
Single crystal elastic moduli for graphite

Component	Modulus (GPa)
C_{1111}	1060
C_{1122}	180
C_{1133}	15
C_{3333}	36.5
C_{2323}	4.5

Table 2
Single crystal elastic moduli for uranium dioxide (UO_2)

Component	Modulus (GPa)
C_{1111}	396.0
C_{1122}	121.0
C_{1212}	64.1

Table 3
Bounds on the bulk κ and shear μ moduli of graphite and uranium dioxide

	Graphite (GPa)	Uranium dioxide (GPa)
κ^{0+}	512.59	212.67
κ^{1+}	286.28	212.67
κ^{2+}	204.17	212.67
$\vdots$		
κ^{∞}	89.04	212.67
$\vdots$		
κ^{2-}	42.63	212.67
κ^{1-}	35.76	212.67
κ^{0-}	30.25	212.67
μ^{0+}	440.00	137.50
μ^{1+}	219.57	93.46
μ^{2+}	149.27	88.28
$\vdots$		
μ^{∞}	54.09	87.19
$\vdots$		
μ^{2-}	16.34	86.44
μ^{1-}	10.26	81.50
μ^{0-}	4.50	64.10

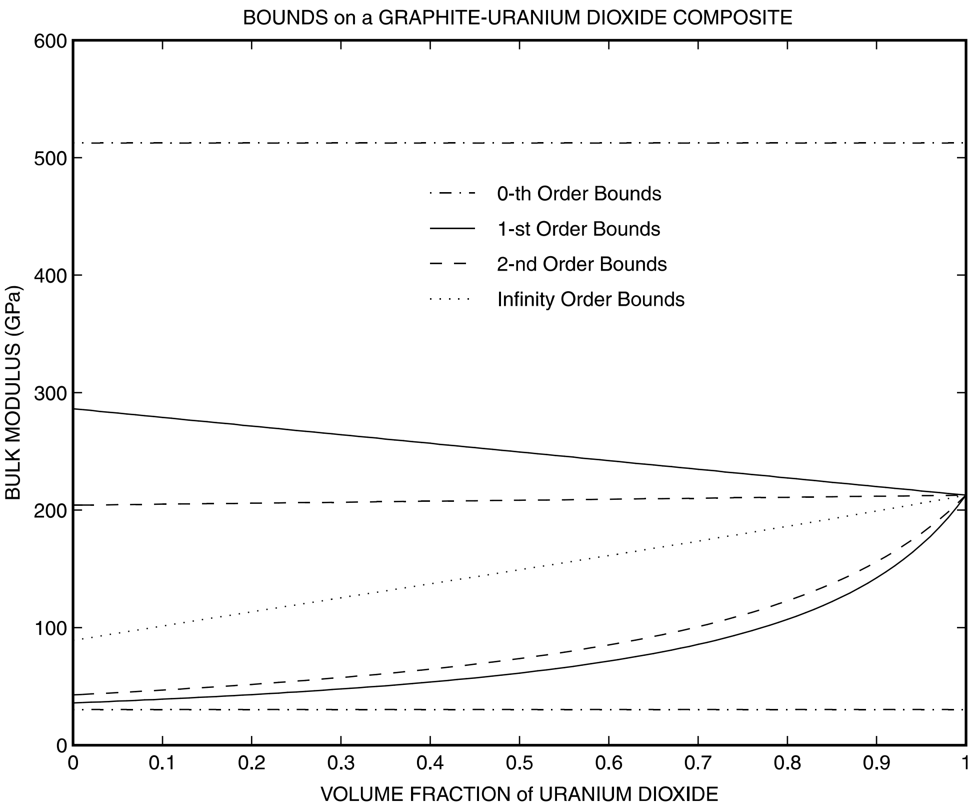

Fig. 1. Graphite–uranium dioxide composite: bulk modulus bounds.

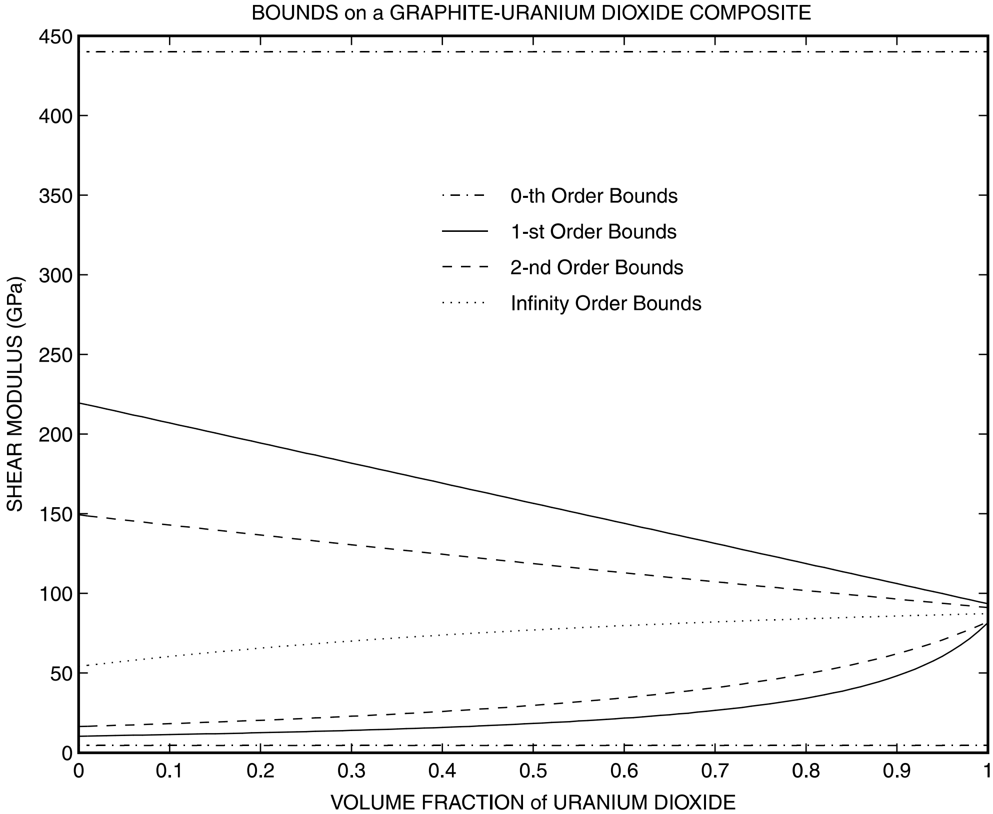

Fig. 2. Graphite–uranium dioxide composite: shear modulus bounds.

These results were achieved by first determining the optimal zeroth-order and first-order bounds for each constituent. These results can be found in Table 3. Once the bounds ($n = 0, 1$) are known for each constituent it is possible to evaluate the zeroth- and first-order bounds of the composite. The zeroth-order bounds of the composite are evaluated using Eqs. (59) and (60). In the case of the first-order composite bounds it is necessary to know the volume fraction of the uranium dioxide. Once these "seeds" – which are the zeroth- and first-order bounds of the composite – are determined, the results of Section 6 are employed to evaluate all of the higher order bounds ($n \geqslant 2$). In Figs. 1 and 2 the bounds corresponding to $n = 0$, 1, 2 and ∞ are presented. The upper and lower infinity-order bounds, which are equivalent to the self-consistent approximation (Kröner, 1977), are coincident, thus, for example, we may define $\kappa^{\infty} := \kappa^{\infty+} = \kappa^{\infty-}$.

Of most practical importance in this example is the evaluation of the Hashin–Shtrikman bounds for a composite with anisotropic constituents. Of less practical importance is the evaluation of the zeroth-order bounds, but as has been previously stated, evaluation of the zeroth-order bounds permits easy evaluation of higher even-order bounds such as the Hashin–Shtrikman bounds.

In performing the tensorial operations necessary to evaluate the zeroth-order bounds, and in implementing the recursive relations of Section 6, it is tremendously helpful to employ the tensor-to-matrix mappings of Nadeau and Ferrari (1998). Through the use of these mappings it is possible to transform the tensorial operations to equivalent standard matrix operations. The calculations for this example were accomplished using the matrix manipulation package, MATLAB.

8. Material anisotropy parameters

An application of zeroth-order bounds is the characterization, or quantification, of material property anisotropy. In this section we present parameters which characterize the anisotropy of generally anisotropic second- and fourth-order properties. This section concludes with two examples: calculation of the anisotropy parameters for (i) the nearly isotropic cubic crystal of tungsten (W) and (ii) one of the most anisotropic materials known – graphite.

8.1. Second-order tensors

A second-order tensor $\boldsymbol{t} \in \mathscr{T}_S^2$ defines a surface: $\boldsymbol{x} \cdot \boldsymbol{t}\boldsymbol{x} = 1$ where $\boldsymbol{x} \in \mathscr{T}^1$. For example, if $\boldsymbol{t}$ is positive definite, then $\boldsymbol{x} \cdot \boldsymbol{t}\boldsymbol{x} = 1$ defines an ellipsoid. With respect to a particular basis the tensor $\boldsymbol{t}$ has components:

$$[k_{ij}] = \begin{bmatrix} \lambda_1 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{bmatrix}, \tag{73}$$

where λ_1, λ_2 and λ_3 are the eigenvalues of $\boldsymbol{t}$. The semi-major axes of the ellipsoidal surface are $\sqrt{1/\lambda_1}$, $\sqrt{1/\lambda_2}$ and $\sqrt{1/\lambda_3}$.

From the above discussion, and the results of Section 4, it follows that the optimal zeroth-order *upper* bound to $\boldsymbol{t}$ is the *smallest* sphere, centered at the origin, which can be *circumscribed* to the surface of the ellipsoid. Similarly, the optimal zeroth-order *lower* bound to $\boldsymbol{t}$ is the *largest* sphere, centered at the origin, which can be *inscribed* to the surface of the ellipsoid. In what follows we introduce a scalar parameter which quantifies the relative size of the two bounding spheres. This parameter is a measure of the anisotropy of the tensor $\boldsymbol{t}$.

Since our application is to physical material properties we now restrict attention to positive definite tensors $\boldsymbol{k}$. Let $\boldsymbol{k}^{0+} = k^{0+}\boldsymbol{i}$ and $\boldsymbol{k}^{0-} = k^{0-}\boldsymbol{i}$ denote the zeroth-order upper and lower bounds to $\boldsymbol{k}$. The anisotropy parameter for $\boldsymbol{k}$ is defined as

$$\alpha := \frac{k^{0+} - k^{0-}}{k^{0+}}. \tag{74}$$

The positive definiteness of $\boldsymbol{k}$ implies that α is bounded:

$$0 \leqslant \alpha < 1. \tag{75}$$

The tensor $\boldsymbol{k}$ is isotropic if the circumscribed and inscribed spheres have the same radius, say k, since then $k_{ij} = k\delta_{ij}$ which is isotropic. Thus, the closer α tends to zero the more isotropic the constitutive relation $\boldsymbol{k}$ while the closer α tends to one the more anisotropic.

8.2. Fourth-order tensors

In analogy with the anisotropy parameter for $\boldsymbol{t} \in \mathscr{T}_S^2$, the optimal zeroth-order bounds for a tensor $\boldsymbol{T} \in \mathscr{T}_S^4$ contain information regarding the extent of anisotropy of $\boldsymbol{T}$. That is, the relative sizes of the two bounding isotropic tensors are a measure of the anisotropy of the tensor $\boldsymbol{T}$.

Since our motivation is the quantification of anisotropy of elastic material properties we now take $\boldsymbol{T} = \boldsymbol{C} \in \mathscr{T}_C$. Let the optimal zeroth-order upper and lower bounds to $\boldsymbol{C}$, respectively take the forms

$$\boldsymbol{C}^{0+} = 3\kappa^{0+}\boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0+}\boldsymbol{I}^{\mathrm{dev,s}}, \tag{76}$$

$$\boldsymbol{C}^{0-} = 3\kappa^{0-}\boldsymbol{I}^{\mathrm{sph}} + 2\mu^{0-}\boldsymbol{I}^{\mathrm{dev,s}}. \tag{77}$$

The anisotropy of $\boldsymbol{C}$ is quantified by the following two parameters:

$$\alpha^{\kappa} := \frac{\kappa^{0+} - \kappa^{0-}}{\kappa^{0+}}, \qquad \alpha^{\mu} := \frac{\mu^{0+} - \mu^{0-}}{\mu^{0+}}. \tag{78}$$

For a general fourth-order tensor a third parameter could be defined but because the elastic modulus $\boldsymbol{C}$ is an element of $\mathscr{T}_{\mathrm{C}}$ it follows that $\boldsymbol{C}^{a} = \boldsymbol{0}$ and thus the third anisotropy parameter, which would be defined as $\alpha^{\eta} := (\eta^{0+} - \eta^{0-})/\eta^{0+}$, would always be equal to zero. The positive definiteness of $\boldsymbol{C}$ implies that both α^{κ} and α^{μ} are bounded:

$$0 \leqslant \alpha^{\kappa} < 1, \qquad 0 \leqslant \alpha^{\mu} < 1. \tag{79}$$

An anisotropy parameter tending to zero is an indication of isotropy while an anisotropy parameter tending to one is an indication of anisotropy.

An elastic tensor $\boldsymbol{C}$ is isotropic if and only if $(\alpha^{\kappa}, \alpha^{\mu})_{\mathrm{isotropic}} = (0, 0)$. A cubic material, on the other hand, is isotropic with respect to the bulk property (i.e., $\alpha^{\kappa} = 0$) but exhibits anisotropy in the shear property. In other words, for a cubic material $(\alpha^{\kappa}, \alpha^{\mu})_{\mathrm{cubic}} = (0, \alpha^{\mu})$.

8.3. Historical elastic anisotropy parameter

Characterization of the anisotropy of *cubic* materials has been noted in the literature. Depending on the particular application, characterization has been quantified by a number of parameters. Here are three such parameters:

$$\alpha_1 := \frac{2C_{2323}}{C_{1111} - C_{1122}}, \tag{80}$$

$$\alpha_2 := \frac{C_{1111} - C_{1122}}{2C_{2323}} = \frac{1}{\alpha_1}, \tag{81}$$

$$\alpha_3 := \frac{C_{1111} - C_{1122} - 2C_{2323}}{C_{1111}}. \tag{82}$$

The ratio of elastic constants on the RHS of the definition for α_1 has been referred to as Zener's anisotropy factor (Zener, 1948, p. 16). Note that $\alpha_1 \geqslant 0$, $\alpha_2 \geqslant 0$ and α_3 is unbounded. The conditions: $\alpha_1 = 1$, $\alpha_2 = 1$ and $\alpha_3 = 0$, imply isotropy. Also note that a material with $\alpha_1 = \bar{\alpha}$ is just as anisotropic (or isotropic, depending how you wish to view it) as a material with $\alpha_1 = 1/\bar{\alpha}$. Likewise, $\alpha_2 = \bar{\alpha}$ and $\alpha_2 = 1/\bar{\alpha}$ are equivalent measures of anisotropy. $\alpha_3 = \bar{\alpha}$ and $\alpha_3 = -\bar{\alpha}$ are also equivalent measures of anisotropy. In short, these quantities are not convenient parameters for comparison of cubic materials due to their non-uniqueness. The parameters introduced in this section, in addition to being applicable to arbitrary material symmetry, are unique.

8.4. Example: single crystal tungsten (W)

Single crystal tungsten (W) has cubic symmetry. As a result, tungsten's thermal conductivity is an isotropic tensor which leads to a thermal conductivity anisotropy parameter of zero: $\alpha_{\mathrm{tungsten}} = 0$.

Tungsten's three independent elastic moduli (Simmons and Wang, 1971, code = 12020) are presented in Table 4. Utilizing the results of the Section 5.3, the optimal zeroth-order bounds were calculated and they

Table 4
Single crystal elastic moduli for tungsten (W)

Component	Modulus (GPa)
C_{1111}	512.57
C_{1122}	205.82
C_{2323}	152.67

Table 5
Optimal zeroth-order bounds to the elastic properties of tungsten (W)

κ^{0+} (GPa)	κ^{0-} (GPa)	μ^{0+} (GPa)	μ^{0-} (GPa)
308.07	308.07	153.38	152.67

Table 6
Single crystal thermal conductivity moduli for graphite

Component	Modulus (W/(m K))
k_{11}	6.87
k_{33}	1840

are presented in Table 5. The anisotropy parameters for tungsten are $(\alpha^{\kappa}, \alpha^{\mu})_{\text{tungsten}} = (0.0000, 0.004629)$. Single crystal tungsten is, therefore, a nearly isotropic material in elastic behavior.

Since tungsten is a cubic material the historical anisotropy parameters of Section 8 were evaluated: $(\alpha_1)_{\text{tungsten}} = 0.9954$, $(\alpha_2)_{\text{tungsten}} = 1.0047$ and $(\alpha_3)_{\text{tungsten}} = 0.002751$.

8.5. Example: single crystal graphite

Single crystal graphite is transversely isotropic. Graphite's two independent thermal conductivity moduli (Null et al., 1973, Table 3) are presented in Table 6. The optimal zeroth-order bounds are thus $k^{0+} = 1840$ W/(m K) and $k^{0-} = 6.87$ W/(m K) which results in an anisotropy parameter of $(\alpha^{k})_{\text{graphite}} = 0.996$.

The zeroth-order bounds on the elastic moduli of graphite were calculated in Section 7 and they were presented in Table 3. The anisotropy parameters of graphite are $(\alpha^{\kappa}, \alpha^{\mu})_{\text{graphite}} = (0.941, 0.990)$.

These parameters quantify – and would appear to confirm – the belief that graphite is one of the most anisotropic materials known. Other highly anisotropic materials include, for example, GaS $((\alpha^{\kappa}, \alpha^{\mu})_{\text{GaS}} = (0.669, 0.737))$ and GaSe $((\alpha^{\kappa}, \alpha^{\mu})_{\text{GaSe}} = (0.660, 0.733))$.

9. Closure

Optimal zeroth-order bounds have been defined for all even ordered tensors. It has been proven that all optimal zeroth-order bounds are isotropic tensors. Additional properties of optimal zeroth-order bounds have also been proven. In regards to evaluation of the optimal bounds, this paper has presented a method for calculating optimal zeroth-order bounds of all second- and fourth-order tensors. As a result of this capability it is possible to evaluate the Hashin–Shtrikman bounds for all macroscopically homogeneous and isotropic composites regardless of the material symmetry of the constituents. In addition, material anisotropy parameters have been defined to quantify the extent of a material's anisotropy.

Acknowledgements

JCN was supported by the National Science Foundation (NSF) grant MSS-9215671 and MF gratefully acknowledges the support of the NSF through the National Young Investigator Award in the Mechanics and Materials program.

Appendix A. Some properties of, and pertaining to optimal bounds

Theorem 1. *The optimal zeroth-order bounds* $\boldsymbol{T}^{0+}, \boldsymbol{T}^{0-} \in \mathscr{T}_S^{2n}$, *to a tensor* $\boldsymbol{T} \in \mathscr{T}_S^{2n}$, *are isotropic.*

Proof. In regards to the upper bound, choose $\boldsymbol{A} = \boldsymbol{T}^{0+}$ in Eqs. (20) and (21) to arrive at

$$\boldsymbol{x} \cdot (\boldsymbol{T}^{0+} - \boldsymbol{\Pi}_q[\boldsymbol{T}^{0+}]) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^n \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^+ \tag{A.1}$$

and

$$\boldsymbol{x} \cdot (\boldsymbol{\Pi}_q[\boldsymbol{T}^{0+}] - \boldsymbol{T}^{0+}) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^n \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^+ \tag{A.2}$$

respectively. Multiplying Eq. (A.2) through by -1 and combining the result with Eq. (A.1) results in the conclusion that

$$\boldsymbol{x} \cdot (\boldsymbol{T}^{0+} - \boldsymbol{\Pi}_q[\boldsymbol{T}^{0+}]) \cdot \boldsymbol{x} = 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^n \text{ and } \forall \boldsymbol{q} \in \mathscr{O}^+ \tag{A.3}$$

from which it follows that

$$\boldsymbol{T}^{0+} = \boldsymbol{\Pi}_q[\boldsymbol{T}^{0+}] \quad \forall \boldsymbol{q} \in \mathscr{O}^+. \tag{A.4}$$

Since $\boldsymbol{T}^{0+} \in \mathscr{T}^{2n}$ is an even-ordered tensor it follows that Eq. (A.6) is also valid $\forall \boldsymbol{q} \in \mathscr{O}$ thus $\boldsymbol{T}^{0+}$ is isotropic. Analogous reasoning leads to the conclusion that $\boldsymbol{T}^{0-}$ is also an isotropic tensor. □

Theorem 2. *For* $\boldsymbol{T}, \boldsymbol{T}^{0+} \in \mathscr{T}_S^{2n}$, $\boldsymbol{T}^{0+}$ *being the optimal zeroth-order upper bound to* $\boldsymbol{T}$, *the largest eigenvalue of the difference* $\boldsymbol{T} - \boldsymbol{T}^{0+}$ *is zero.*

Proof. For $i \in \{1, 2, \ldots, 3^n\}$ let $(\lambda_i, \boldsymbol{v}_i)$, where $\lambda_i \in \Re$ and $\boldsymbol{v}_i \in \mathscr{T}^n$, denote the 3^n eigenvalue–eigentensor pairs of the difference $\boldsymbol{T} - \boldsymbol{T}^{0+}$. In other words,

$$\left[\boldsymbol{T} - \boldsymbol{T}^{0+} - \lambda_i \boldsymbol{I}\right] \cdot \boldsymbol{v}_i = \boldsymbol{0}. \tag{A.5}$$

Let $\lambda_{\max} := \max_i \lambda_i$. By definition of $\boldsymbol{T}^{0+}$ it is known that the difference in question is negative semi-definite. Thus all 3^n eigenvalues λ_i are less than or equal to zero; or, equivalently, $\lambda_{\max} \leqslant 0$. We are thus proving that $\lambda_{\max} = 0$.

Toward a contradiction assume that $\lambda_{\max} = -\epsilon < 0$ and let

$$\boldsymbol{A} = \boldsymbol{T}^{0+} - \tfrac{1}{2}\epsilon \boldsymbol{I} \tag{A.6}$$

where $\boldsymbol{I} \in \mathscr{T}_S^{2n}$ is the identity tensor. Note that $\boldsymbol{A}$ is isotropic and thus $\boldsymbol{\Pi}_q[\boldsymbol{A}] = \boldsymbol{A} \ \forall \boldsymbol{q} \in \mathscr{O}$.

We first show that $\boldsymbol{A}$ is a zeroth-order upper bound to $\boldsymbol{T}$. We next show that $\boldsymbol{A}$ is a *better* upper bound to $\boldsymbol{T}$ than $\boldsymbol{T}^{0+}$, which is in contradiction to the definition of $\boldsymbol{T}^{0+}$, therefore $\lambda_{\max} \geqslant 0$. It being previously determined that $\lambda_{\max} \leqslant 0$ it follows then that $\lambda_{\max} = 0$.

To prove that $\boldsymbol{A}$ is a zeroth-order upper bound to $\boldsymbol{T}$ we show that all the eigenvalues of the difference, $\boldsymbol{T} - \boldsymbol{A}$, are less than or equal to zero. The eigenproblem to be considered is

$$\boldsymbol{0} = [\boldsymbol{T} - \boldsymbol{A} - \bar{\lambda}_i \boldsymbol{I}] \cdot \bar{\boldsymbol{v}}_i \tag{A.7}$$

$$\boldsymbol{0} = [\boldsymbol{T} - \boldsymbol{T}^{0+} - \left(\bar{\lambda}_i - \tfrac{1}{2}\epsilon\right) \boldsymbol{I}] \cdot \bar{\boldsymbol{v}}_i. \tag{A.8}$$

The eigenpairs are $(\bar{\lambda}_i - \frac{1}{2}\epsilon, \bar{v}_i) = (\lambda_i, \boldsymbol{v}_i)$. Thus, $\bar{\lambda}_i = \lambda_i + \frac{1}{2}\epsilon$ and, taking the max of both sides, $\bar{\lambda}_{\max} = \lambda_{\max} + \frac{1}{2}\epsilon = -\epsilon + \frac{1}{2}\epsilon = -\frac{1}{2}\epsilon < 0$. Therefore, $\boldsymbol{A}$ is a zeroth-order upper bound to $\boldsymbol{T}$.

To prove that $\boldsymbol{A}$ is a better zeroth-order upper bound to $\boldsymbol{T}$ then $\boldsymbol{T}^{0+}$ we show that $\boldsymbol{T}^{0+} - \boldsymbol{A}$ is *positive* semi-definite. Substituting Eq. (A.6) into Eq. (19) yields

$$\boldsymbol{x} \cdot (\boldsymbol{T}^{0+} - \boldsymbol{A}) \cdot \boldsymbol{x} = \tfrac{1}{2}\epsilon \boldsymbol{x} \cdot \boldsymbol{x} \geqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^n. \qquad \square \tag{A.9}$$

Theorem 3. *For $\boldsymbol{T}, \boldsymbol{T}^{0-} \in \mathscr{T}_S^{2n}$, $\boldsymbol{T}^{0-}$ being the optimal zeroth-order lower bound to $\boldsymbol{T}$, the smallest eigenvalue of the difference $\boldsymbol{T} - \boldsymbol{T}^{0-}$ is zero.*

The proof follows that of Theorem 2. $\square$

Theorem 4. *For $\boldsymbol{A} \in \mathscr{T}_S^{4n}$, the expression*

$$\boldsymbol{x} \cdot \boldsymbol{A} \cdot \boldsymbol{x} \leqslant (\geqslant) 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^{2n} \tag{A.10}$$

holds iff

$$\boldsymbol{x} \cdot \boldsymbol{A}^s \cdot \boldsymbol{x} \leqslant (\geqslant) 0 \quad \forall \boldsymbol{x} \in \mathscr{T}_S^{2n} \tag{A.11}$$

and

$$\boldsymbol{x} \cdot \boldsymbol{A}^a \cdot \boldsymbol{x} \leqslant (\geqslant) 0 \quad \forall \boldsymbol{x} \in \mathscr{T}_A^{2n}. \tag{A.12}$$

Proof. First, perform additive decompositions of $\boldsymbol{A} \in \mathscr{T}_S^{4n}$ and $\boldsymbol{x} \in \mathscr{T}^{2n}$:

$$\boldsymbol{A} = \boldsymbol{A}^s + \boldsymbol{A}^a \tag{A.13}$$

$$\boldsymbol{x} = \boldsymbol{x}^S + \boldsymbol{x}^A. \tag{A.14}$$

Substituting these decompositions into Eq. (A.10) yields

$$\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} = (\boldsymbol{x}^S + \boldsymbol{x}^A) : (\boldsymbol{A}^s + \boldsymbol{A}^a) : (\boldsymbol{x}^S + \boldsymbol{x}^A) \tag{A.15}$$

$$\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} = \boldsymbol{x}^S : \boldsymbol{A}^s : \boldsymbol{x}^S + \boldsymbol{x}^A : \boldsymbol{A}^a : \boldsymbol{x}^A. \tag{A.16}$$

Sufficiency: Obviously, if Eqs. (A.11) and (A.12) hold then from Eq. (A.16) it follows that Eq. (A.10) also holds.

Necessity: Since Eq. (A.10) is valid $\forall \boldsymbol{x} \in \mathscr{T}^{2n}$, it follows that taking $\boldsymbol{x}^A = \boldsymbol{0}$ gives Eq. (A.11) and taking $\boldsymbol{x}^S = \boldsymbol{0}$ gives Eq. (A.12). $\square$

Theorem 5. *For $\boldsymbol{A} \in \mathscr{T}_S^4$ with a spherical eigentensor, the expression*

$$\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} \leqslant (\geqslant) 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2 \tag{A.17}$$

holds iff

$$\boldsymbol{x}^{sph} : \boldsymbol{A} : \boldsymbol{x}^{sph} \leqslant (\geqslant) 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2, \tag{A.18}$$

$$\boldsymbol{x}^{dev} : \boldsymbol{A} : \boldsymbol{x}^{dev} \leqslant (\geqslant) 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^2. \tag{A.19}$$

Proof. First, perform a spherical-deviatoric decomposition of $\boldsymbol{x} \in \mathscr{T}^2$ and substitute into Eq. (A.17) to yield

$$\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} = (\boldsymbol{x}^{sph} + \boldsymbol{x}^{dev}) : \boldsymbol{A} : (\boldsymbol{x}^{sph} + \boldsymbol{x}^{dev}) \tag{A.20}$$

$$\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} = \boldsymbol{x}^{sph} : \boldsymbol{A} : \boldsymbol{x}^{sph} + \boldsymbol{x}^{sph} : \boldsymbol{A} : \boldsymbol{x}^{dev} + \boldsymbol{x}^{dev} : \boldsymbol{A} : \boldsymbol{x}^{sph} + \boldsymbol{x}^{dev} : \boldsymbol{A} : \boldsymbol{x}^{dev} \tag{A.21}$$

$$\boldsymbol{x} : \boldsymbol{A} : \boldsymbol{x} = \boldsymbol{x}^{sph} : \boldsymbol{A} : \boldsymbol{x}^{sph} + \boldsymbol{x}^{dev} : \boldsymbol{A} : \boldsymbol{x}^{dev}. \tag{A.22}$$

Eq. (A.22) follows from Eq. (A.21) because $\boldsymbol{A} : \boldsymbol{x}^{sph} = \boldsymbol{x}^{sph} : \boldsymbol{A} \propto \boldsymbol{x}^{sph}$ and $\boldsymbol{x}^{sph} : \boldsymbol{x}^{dev} = 0$.

Sufficiency: Obviously, if Eqs. (A.18) and (A.19) hold then from Eq. (A.22) it follows that Eq. (A.17) also holds.

Necessity: Since Eq. (A.17) is valid $\forall \boldsymbol{x} \in \mathscr{T}^2$, it follows that taking $\boldsymbol{x}^{dev} = \boldsymbol{0}$ gives Eq. (A.18) and taking $\boldsymbol{x}^{sph} = \boldsymbol{0}$ gives Eq. (A.19). □

Theorem 6. *For* $\boldsymbol{A}, \boldsymbol{B} \in \mathscr{T}^{2n}$

$$\boldsymbol{x} \cdot (\boldsymbol{A} - \boldsymbol{B}) \cdot \boldsymbol{x} \leqslant 0 \quad \forall \boldsymbol{x} \in \mathscr{T}^n \Rightarrow \operatorname{tr} \boldsymbol{A} \leqslant \operatorname{tr} \boldsymbol{B}. \tag{A.23}$$

Proof. Recall that the trace of a tensor $\boldsymbol{T} \in \mathscr{T}^{2n}$ is an invariant. We now prove this theorem for the case $n = 2$; the other cases follow in a similar manner. Evaluating the LHS of the implication (A.23) for the following choices of $[\boldsymbol{x}]$

$$\begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \begin{bmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \ldots, \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \tag{A.24}$$

leads, respectively, to the following component inequalities:

$$A_{1111} \leqslant B_{1111}, \qquad A_{1212} \leqslant B_{1212}, \ \ldots, \ A_{3333} \leqslant B_{3333}. \tag{A.25}$$

Summing components yields

$$A_{1111} + A_{1212} + \cdots + A_{3333} \leqslant B_{1111} + B_{1212} + \cdots + B_{3333} \tag{A.26}$$

which is equivalent to $\operatorname{tr} \boldsymbol{A} \leqslant \operatorname{tr} \boldsymbol{B}$. The proofs for $n = 1, 3, 4, 5, \ldots,$ proceed in a similar manner. □

Note: It can *not* be concluded from Theorem 6 that $A_{i_1 \ldots i_{2n}} \leqslant B_{i_1 \ldots i_{2n}}$. For example, for the case $n = 1$, *only* the following three inequalities can be deduced: $A_{11} \leqslant B_{11}$, $A_{22} \leqslant B_{22}$, and $A_{33} \leqslant B_{33}$. Nothing can be determined about the relations amongst the off-diagonal terms.

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