



NEW METHOD TO GENERATE THREE-POINT BOUNDS ON EFFECTIVE PROPERTIES OF COMPOSITES: APPLICATION TO VISCOELASTICITY

L. V. GIBIANSKY and S. TORQUATO*

Princeton Materials Institute and Department of Civil Engineering and Operations Research,
Princeton University, Princeton, NJ 08540, U.S.A.

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ABSTRACT

The general goal of this paper is to develop a new approach for bounding the effective moduli (e.g. elastic moduli, conductivity, or thermal expansion coefficients) of composite materials. Specifically, this paper aims to: (1) formulate a procedure that combines the translation method and the Beran procedure into one powerful method that has the advantages of both approaches; and (2) apply the new method to study effective properties of viscoelastic composites. The new method enables one to get the most restrictive three-point geometrical-parameter bounds. In particular, we obtain new three-point bounds on the complex bulk modulus of an isotropic viscoelastic composite. The new bounds are given by the outermost of several circular arcs in the complex bulk modulus plane. They take into account three-point statistical information and thus are much more restrictive than previously known two-point bounds. © 1998 Elsevier Science Ltd. All rights reserved

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1. INTRODUCTION

Composite materials abound in nature and in man-made situations. It is known that effective properties of composites strongly depend on the microstructure. Even in the rare situations when the microstructure of a composite is completely known, it is computationally intensive to find the composite properties exactly. Generally, rigorous geometry-independent bounds on the composite moduli are of great value since they provide a benchmark for testing experimental results and approximation theories, and may provide an indicator of whether the effective response of a given composite is extreme (optimal) in the sense of being close to the bounds. Moreover, optimal bounds are important in the context of structural optimization since the microstructures that achieve the bounds are often the best candidates for use in the design of a structure.

Bounds on the effective properties of composites have attracted much attention in the literature [see the reviews of Christensen (1979), Willis (1981), Hashin (1983), Lurie and Cherkhaev (1986b), and Torquato (1991), to mention a few]. There exist

* Author to whom correspondence should be addressed. E-mail: torquato@matter.princeton.edu

several approaches that allow one to obtain rigorous bounds. Most of them are based on variational principles that define the effective properties. Each approach has its own advantages and disadvantages.

For example, the translation method originated by Lurie and Cherkaev (1984, 1986a,b), Murat and Tartar (1985), and Tartar (1985) has proved to be the most powerful tool to obtain tight geometrically-independent bounds on the composite moduli. The method has been shown to be especially successful in obtaining cross-property bounds even if the governing equations are uncoupled [see Cherkaev and Gibiansky (1992), and Gibiansky and Torquato (1993, 1995a, 1996)]. A new boost to the utility of the translation method was given by the discovery of Cherkaev and Gibiansky (1992) and Milton (1991b) that the special fractional-linear Y -transformation of the effective properties tensors allows one to simplify drastically all of the algebraic calculations involved in the derivation of the bounds. This transformation was also successfully used by Gibiansky and Milton (1993a,b) and Milton and Berryman (1997) in the derivation of the bounds on the effective complex moduli of viscoelastic composites by using the Hashin–Shtrikman (1963) method. A variational definition of the Y -tensor given by Milton (1991b) explains why this transformation is so important for the theory of composites. However, in the present form, a defect of the translation method is a lack of a rigorous means of taking into account more subtle microstructural information, such as three-point geometrical parameters.

A distinguished feature of the perturbation-solution method originated by Beran (1965) is its ability to incorporate, into the rigorous bounds, three-point correlation function information in the form of the three-point geometrical parameters. As was shown by Beran (1965), Beran and Molyneux (1966), McCoy (1970), Silnutzer (1972), Milton (1981a,b,c, 1982), such information allows one to improve upon the Hashin–Shtrikman conductivity and elastic moduli bounds significantly. However, in its present form, this method has a disadvantage in treating more complex problems such as cross-property bounds or viscoelasticity.

We examine these two procedures in order to use the Y -transformation to simplify the derivation of the Beran bounds. We then combine both approaches to create a new method that has advantages of both of the aforementioned “parent” methods. Specifically, we incorporate three-point statistics into the bounding procedure, as in the Beran method. The procedure results in several free parameters that allow us to “tune” the method to get the best possible bounds, as in the translation method. Our method is based on the variational principles describing the Y -tensor rather than the effective properties tensor itself. This simplifies the calculations and allows one to solve more challenging problems.

We apply our method to obtain complex bulk modulus bounds involving three-point geometrical parameters. Harmonic oscillations in viscoelastic media can be described in the quasistatic limit by the elasticity equations but with the complex fields and complex phase moduli. Complex bulk moduli bounds of this paper naturally follow a series of recent articles which developed the new approach to the complex viscoelastic moduli bounds. Specifically, Gibiansky and Milton (1993a,b) and Milton and Berryman (1997) found complex bulk and shear moduli bounds for a planar and three-dimensional composite with *fixed* phase volume fractions by using the translation and Hashin–Shtrikman method. Gibiansky and Lakes (1993, 1997) sug-

gested a simple method that uses the fixed-volume-fraction bounds to obtain bounds on the effective complex moduli of composites with *arbitrary* phase volume fractions, i.e. containing less information about the microstructure. In contrast, we use the new method to incorporate in the bounds three-point statistical information about the microstructure, in addition to the phase volume fractions. The new complex moduli bounds depend on three-point ζ -parameters and are much more restrictive than known two-point bounds.

Although our procedure will be applied to the viscoelasticity problem, the method is not restricted to any particular problem. It can be applied to generate three-point complex conductivity bounds, pure elastic bulk and shear moduli bounds, cross-property conductivity-elastic moduli bounds, etc. For example, for the conductivity problem, it improves upon the Beran (1965) bounds and reproduces the best available Milton (1981b) three-point bounds. For the complex conductivity problem, our method reproduces the best available Milton (1981a,b,c) bounds on the complex conductivity of isotropic composite in two dimensions and improves upon those bounds in three dimensions.

The new method further develops an idea that was used recently by Gibiansky and Torquato (1995b) who reviewed and improved known three-point bounds on the effective elastic moduli of planar two-phase composites. In our paper (1995b), we presented the geometrical-parameter bounds in a simple form by using the Y -transformation, and then used the translation method inequality and the Silnutzer (1972) three-point bounds to obtain the best available three-point bounds on the bulk and shear moduli of a planar composite. A similar idea was implemented by Helsing (1993) who used translation in the Hashin–Shtrikman procedure to obtain the bounds on the effective conductivity of a polycrystal.

Milton (1991b) and Gibiansky and Torquato (1995b) have noticed that the formulas for the geometrical-parameters bounds can be greatly simplified by using a special fraction-linear Y -transformation [Milton (1991b), Cherkaev and Gibiansky (1992)]. Moreover, Gibiansky and Torquato (1995b) also noticed that another similar transformation [that was called Y_ζ transformation in the cited paper but is called the Z -transformation in this paper] allows one to simplify further the geometrical-parameter bounds. We study the roots of this simplicity and obtain geometrical-parameters complex bulk modulus bounds in the form of the Z -transformed moduli. Similar successive Y -transformations were considered for the two-dimensional conductivity problem by Milton (1987) and by Clark and Milton (1995).

The structure of the paper is the following. In Section 2 we first review the translation method and the perturbation solution procedure (that we call Beran's method), and describe the properties of the so-called Y -transformation. Then we combine these techniques into the new method having the advantages of both the translation and Beran methods. Section 3 is devoted to the viscoelasticity problem: we state the problem and use the new method to get three-point bounds on the effective complex bulk modulus of a two-phase viscoelastic composite. In Section 4 we give an explicit prescription on how to construct complex bulk modulus bounds and discuss results for three-point complex bulk modulus bounds. The reader who is mainly interested in applications need only read Sections 2.1 and 3.1 for the precise statement of the problems and Section 4.3 where the results are formulated.

2. THREE-POINT BOUNDS: NEW METHOD

We incorporate and integrate the essential ingredients of the translation and Beran methods in order to formulate a new method that possesses advantages of both approaches.

2.1. Variational principle and the tensor of effective properties

Consider an intensity vector \mathbf{e} , a flux vector \mathbf{j} and let them be related by the linear constitutive relation

$$\mathbf{j} = \mathbf{D} : \mathbf{e}, \quad (2.1)$$

where \mathbf{D} is a positive-definite, symmetric property tensor and the symbol $:$ denotes an appropriate contraction-type operation. The intensity and the flux vectors are not arbitrary; they possess some differential properties. For example, in the elasticity problem, $\mathbf{e} = [\nabla \mathbf{u} + (\nabla \mathbf{u})^T]/2$ is the second-order strain tensor, \mathbf{j} is the divergence-free ($\nabla \cdot \mathbf{j} = 0$) second-order stress tensor, \mathbf{D} is the fourth-order stiffness tensor, and $:$ is a contraction with respect to two indices.

We consider a two-phase statistically homogeneous composite. The property tensor \mathbf{D} at the point \mathbf{x} can be expressed in terms of the characteristic functions $\chi_i(\mathbf{x})$ of phases $i = 1, 2$ according to

$$\mathbf{D}(\mathbf{x}) = \chi_1(\mathbf{x})\mathbf{D}_1 + \chi_2(\mathbf{x})\mathbf{D}_2, \quad (2.2)$$

where

$$\chi_i(\mathbf{x}) = \begin{cases} 1, & \text{if } \mathbf{x} \in \text{phase } i, \\ 0, & \text{otherwise,} \end{cases} \quad (2.3)$$

and \mathbf{D}_1 and \mathbf{D}_2 are the properties of phase 1 and phase 2, respectively. By performing the volume averaging of (2.3), one finds

$$\langle \chi_i(\mathbf{x}) \rangle = f_i, \quad i = 1, 2, \quad (2.4)$$

where angular brackets denote volume averaging and f_i is the volume fraction of phase i .

One can define the tensor of effective properties \mathbf{D}_* via the variational principle

$$\mathbf{e}_0 : \mathbf{D}_* : \mathbf{e}_0 = \min_{\substack{\mathbf{e}(\mathbf{x}) \\ \langle \mathbf{e}(\mathbf{x}) \rangle = \mathbf{e}_0 \\ \mathbf{e}(\mathbf{x}) \in \mathcal{E}_e}} \langle \mathbf{e}(\mathbf{x}) : \mathbf{D}(\mathbf{x}) : \mathbf{e}(\mathbf{x}) \rangle \quad (2.5)$$

where the minimum operation is taken over the admissible \mathbf{e} -fields with given average value \mathbf{e}_0 . The set \mathcal{E}_e is the set of the fields possessing the required differential properties. For example, in the elastic problem $\mathbf{e}(\mathbf{x}) \in \mathcal{E}_e$ means that $\mathbf{e}(\mathbf{x}) = [\nabla \mathbf{u}(\mathbf{x}) + (\nabla \mathbf{u}(\mathbf{x}))^T]/2$ for some displacement $\mathbf{u}(\mathbf{x})$.

Alternatively, the effective properties can be defined via the conjugate variational principle, namely,

$$\mathbf{j}_0 : \mathbf{F}_* : \mathbf{j}_0 = \min_{\substack{\langle \mathbf{j}(\mathbf{x}) \rangle = \mathbf{j}_0 \\ \mathbf{j}(\mathbf{x}) \in \mathcal{E}_j}} \langle \mathbf{j}(\mathbf{x}) : \mathbf{F}(\mathbf{x}) : \mathbf{j}(\mathbf{x}) \rangle, \quad (2.6)$$

where

$$\mathbf{F}(\mathbf{x}) = \mathbf{D}^{-1}(\mathbf{x}) = \chi_1(\mathbf{x})\mathbf{F}_1 + \chi_2(\mathbf{x})\mathbf{F}_2, \quad \mathbf{F}_i = \mathbf{D}_i^{-1}, \quad i = 1, 2, \quad (2.7)$$

and the minimum is taken over the admissible \mathbf{j} -fields with given average value \mathbf{j}_0 . For example, admissible flux fields in the case of elasticity are the divergence-free symmetric stress tensor fields. These two definitions are equivalent, i.e. $\mathbf{F}_* = \mathbf{D}^{-1}_*$. The effective tensor \mathbf{D}_* does not depend on the load (i.e. does not depend on the average fields \mathbf{e}_0 or \mathbf{j}_0), but depends on the phase properties, volume fractions, and microstructure of the composite.

The variational principles (2.5) and (2.6) can be used to obtain bounds on the effective tensors \mathbf{D}_* and \mathbf{F}_* . For example, energy of any trial field $\hat{\mathbf{e}}(\mathbf{x}) \in \mathcal{E}_e$, $\langle \hat{\mathbf{e}}(\mathbf{x}) \rangle = \mathbf{e}_0$ gives the upper bound on the effective energy via (2.5), thus leading to the bounds on the effective tensor \mathbf{D}_* . In the following sections we review two methods to obtain such bounds.

2.2. Translation method

In this section we give a brief review of the translation method. Recent advances of the method have been discussed in detail in papers by Milton and Kohn (1988), Milton (1990, 1991a,b), Cherkaev and Gibiansky (1992, 1993), and Gibiansky and Torquato (1995a,b, 1996). Therefore, we only outline the main results that are used in our new procedure.

Let the constant matrices \mathbf{T}_e and \mathbf{T}_j be associated with the quasiconvex quadratic forms of the vectors $\mathbf{e}(\mathbf{x})$ and $\mathbf{j}(\mathbf{x})$, respectively, i.e.

$$\langle \mathbf{e}(\mathbf{x}) \rangle : \mathbf{T}_e : \langle \mathbf{e}(\mathbf{x}) \rangle \leq \langle \mathbf{e}(\mathbf{x}) : \mathbf{T}_e : \mathbf{e}(\mathbf{x}) \rangle, \quad \forall \mathbf{e}(\mathbf{x}) \in \mathcal{E}_e, \quad (2.8)$$

$$\langle \mathbf{j}(\mathbf{x}) \rangle : \mathbf{T}_j : \langle \mathbf{j}(\mathbf{x}) \rangle \leq \langle \mathbf{j}(\mathbf{x}) : \mathbf{T}_j : \mathbf{j}(\mathbf{x}) \rangle, \quad \forall \mathbf{j}(\mathbf{x}) \in \mathcal{E}_j, \quad (2.9)$$

for any vector fields $\mathbf{e}(\mathbf{x}) \in \mathcal{E}_e$ and $\mathbf{j}(\mathbf{x}) \in \mathcal{E}_j$ with appropriate differential properties. The reader is referred, e.g. to Ball *et al.* (1981), and Dacorogna (1982) for a proper definition and properties of the quasiconvex functions.

Consider two different two-phase composites having identical microstructures. One of them contains phases with properties $\mathbf{F}_1, \mathbf{F}_2$ and possesses the effective tensor \mathbf{F}_* . The other one contains the same phases "translated" by the matrix \mathbf{T}_j , i.e. with properties $\hat{\mathbf{F}}_1 = \mathbf{F}_1 - \mathbf{T}_j$, $\hat{\mathbf{F}}_2 = \mathbf{F}_2 - \mathbf{T}_j$, and possesses the effective tensor $\hat{\mathbf{F}}_*$. We assume that both phases of the translated composite are positive semi-definite, i.e.

$$\hat{\mathbf{F}}_1 = \mathbf{F}_1 - \mathbf{T}_j \geq 0, \quad \hat{\mathbf{F}}_2 = \mathbf{F}_2 - \mathbf{T}_j \geq 0. \quad (2.10)$$

Then one can prove that

$$\mathbf{F}_* - \mathbf{T}_j \geq \hat{\mathbf{F}}_*, \quad (2.11)$$

or, equivalently

$$(\mathbf{F}_* - \mathbf{T}_j)^{-1} \leq \hat{\mathbf{F}}_*^{-1}. \quad (2.12)$$

Milton (1990) was the first to present the translation method in such a form and gave to the method its present name. In what follows we will show how to obtain a lower bound on the effective tensor, \mathbf{F}_* by using the inequality (2.12).

Remark: Similarly, one can consider a translated composite with the phases $\hat{\mathbf{D}}_1 = \mathbf{D}_1 - \mathbf{T}_e \geq 0$, $\hat{\mathbf{D}}_2 = \mathbf{D}_2 - \mathbf{T}_e \geq 0$, and the effective tensor $\hat{\mathbf{D}}_*$ and show that $\mathbf{D}_* - \mathbf{T}_e \geq \hat{\mathbf{D}}_*$. By using this inequality, one can obtain a lower bound on the effective tensor \mathbf{D}_* (i.e. an upper bound on the inverse tensor $\mathbf{F}_* = \mathbf{D}_*^{-1}$).

Consider the “translated” composite with the phase properties $\hat{\mathbf{D}}_1 = (\mathbf{F}_1 - \mathbf{T}_j)^{-1}$, and $\hat{\mathbf{D}}_2 = (\mathbf{F}_2 - \mathbf{T}_j)^{-1}$ and the effective tensor $\hat{\mathbf{D}}_* = \hat{\mathbf{F}}_*^{-1}$. As follows from the variational principle (2.5),

$$\mathbf{e}_0 : \hat{\mathbf{D}}_* : \mathbf{e}_0 \leq \langle \hat{\mathbf{e}}(\mathbf{x}) : \hat{\mathbf{D}}(\mathbf{x}) : \hat{\mathbf{e}}(\mathbf{x}) \rangle, \quad (2.13)$$

where $\hat{\mathbf{e}}(\mathbf{x}) \in \mathcal{E}_e$ is any appropriate trial field with given average value $\langle \hat{\mathbf{e}}(\mathbf{x}) \rangle = \mathbf{e}_0$. Expressing the local properties of the translated composite via the local properties of the original composite, and by using the inequality (2.12) we arrive at the bound

$$\mathbf{e}_0 : (\mathbf{F}_* - \mathbf{T}_j)^{-1} : \mathbf{e}_0 \leq \langle \hat{\mathbf{e}}(\mathbf{x}) : (\mathbf{F}(\mathbf{x}) - \mathbf{T}_j)^{-1} : \hat{\mathbf{e}}(\mathbf{x}) \rangle. \quad (2.14)$$

One can choose $\hat{\mathbf{e}}(\mathbf{x})$ to be a constant field $\hat{\mathbf{e}}(\mathbf{x}) = \mathbf{e}_0$ thus leading to the bound

$$\mathbf{e}_0 : (\mathbf{F}_* - \mathbf{T}_j)^{-1} : \mathbf{e}_0 \leq \mathbf{e}_0 : \langle (\mathbf{F}(\mathbf{x}) - \mathbf{T}_j)^{-1} \rangle : \mathbf{e}_0, \quad (2.15)$$

or equivalently,

$$(\mathbf{F}_* - \mathbf{T}_j)^{-1} \leq \langle (\mathbf{F}(\mathbf{x}) - \mathbf{T}_j)^{-1} \rangle. \quad (2.16)$$

For a two-phase medium this reduces to

$$(\mathbf{F}_* - \mathbf{T}_j)^{-1} \leq f_1 (\mathbf{F}_1 - \mathbf{T}_j)^{-1} + f_2 (\mathbf{F}_2 - \mathbf{T}_j)^{-1}. \quad (2.17)$$

Relation (2.17) is the main inequality of the translation method. To get the strongest bound one can optimize this inequality over the set of the translation matrices \mathbf{T}_j which correspond to the quasiconvex quadratic forms [i.e. those which satisfy the inequality (2.9)]. Note that the variational principles (2.5) and (2.6) are only valid for the materials with nonnegative tensor of properties. In the considered case it leads to the restrictions (2.10) on the admissible translation matrix \mathbf{T}_j . These restrictions are very important for the method.

Our new procedure diverges from the described translation method at the level of the choice of the trial field. Instead of using the constant trial field in (2.14), we will use the first-order perturbation solution as a trial field. Such a field takes into account additional information about the microstructure of the composite, resulting in sharper bounds.

2.3. Beran approach

Here we outline the aforementioned standard Beran approach to finding trial fields in the variational principles (2.5) and (2.6) that lead to tight bounds on the effective

moduli. Following Milton (1982) we use Fourier series representations which greatly simplify the derivation.

Without loss of generality we assume that the composite is a periodic material. Then the characteristic function $\chi_1(\mathbf{x})$, the local properties tensors $\mathbf{D}(\mathbf{x})$, $\mathbf{F}(\mathbf{x})$, and the fields $\mathbf{j}(\mathbf{x})$ and $\mathbf{e}(\mathbf{x})$ can be represented as Fourier series:

$$\chi_1(\mathbf{x}) = f_1 + \sum_{\mathbf{k} \neq 0} \omega(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}, \quad i^2 = -1, \quad (2.18)$$

$$\mathbf{D}(\mathbf{x}) = \langle \mathbf{D} \rangle + (\mathbf{D}_1 - \mathbf{D}_2) \sum_{\mathbf{k} \neq 0} \omega(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}, \quad (2.19)$$

$$\mathbf{F}(\mathbf{x}) = \langle \mathbf{F} \rangle + (\mathbf{F}_1 - \mathbf{F}_2) \sum_{\mathbf{k} \neq 0} \omega(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}, \quad (2.20)$$

$$\mathbf{j}(\mathbf{x}) = \mathbf{j}_0 + \sum_{\mathbf{k} \neq 0} \mathbf{J}(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}, \quad \mathbf{j}_0 = \langle \mathbf{j} \rangle, \quad (2.21)$$

$$\mathbf{e}(\mathbf{x}) = \mathbf{e}_0 + \sum_{\mathbf{k} \neq 0} \mathbf{E}(\mathbf{k}) e^{i\mathbf{k} \cdot \mathbf{x}}, \quad \mathbf{e}_0 = \langle \mathbf{e} \rangle, \quad (2.22)$$

where \mathbf{k} is the Fourier wave-vector, and $\omega(\mathbf{k})$, $\mathbf{J}(\mathbf{k})$ and $\mathbf{E}(\mathbf{k})$ are the Fourier coefficients of the functions $\chi(\mathbf{x})$, $\mathbf{j}(\mathbf{x})$ and $\mathbf{e}(\mathbf{x})$, respectively. The local fields $\mathbf{j}(\mathbf{x})$ and $\mathbf{e}(\mathbf{x})$ are connected via the constitutive equation (2.1), and the effective property tensor $\mathbf{D}_* = \mathbf{F}^{-1} \mathbf{F}_*$ is the proportionality coefficient between the average fields \mathbf{e}_0 and \mathbf{j}_0 , i.e.

$$\mathbf{j}_0 = \mathbf{D}_* : \mathbf{e}_0. \quad (2.23)$$

The Fourier coefficients $\mathbf{J}(\mathbf{k})$ and $\mathbf{E}(\mathbf{k})$ of the fields $\mathbf{j}(\mathbf{x})$ and $\mathbf{e}(\mathbf{x})$ satisfy algebraic equalities that follow from the differential equalities in real space. For example, in the elasticity problem, the Fourier coefficients $\mathbf{E}(\mathbf{k})$ of the deformation field $\mathbf{e}(\mathbf{x}) = [\nabla \mathbf{u}(\mathbf{x}) + (\nabla \mathbf{u}(\mathbf{x}))^T]/2$ can be written as

$$\mathbf{E}(\mathbf{k}) = \frac{i}{2} (\mathbf{k} \mathbf{U}(\mathbf{k}) + \mathbf{U}(\mathbf{k}) \mathbf{k}), \quad (2.24)$$

where $\mathbf{U}(\mathbf{k})$ are the Fourier coefficients of the displacement field $\mathbf{u}(\mathbf{x})$. Similarly, the Fourier coefficients $\mathbf{J}(\mathbf{k})$ of the stress field \mathbf{j} , $\nabla \cdot \mathbf{j}(\mathbf{x}) = 0$, satisfy the relation

$$\mathbf{k} \cdot \mathbf{J}(\mathbf{k}) = 0. \quad (2.25)$$

Assuming that the dimensionless difference in the phase properties is small (of the order of $\delta \ll 1$), one can find the asymptotic expansion of the solution, i.e.

$$\begin{aligned} \mathbf{e}(\mathbf{x}) &= \mathbf{e}_0 + \delta \mathbf{e}_p(\mathbf{x}) + O(\delta^2), \\ \mathbf{j}(\mathbf{x}) &= \mathbf{j}_0 + \delta \mathbf{j}_p(\mathbf{x}) + O(\delta^2). \end{aligned} \quad (2.26)$$

For example, for an isotropic elastic composite of two isotropic phases, the Fourier coefficient $\hat{\mathbf{e}}(\mathbf{k})$ of the first-order perturbation solution for the hydrostatic average strain field $\mathbf{e}_0 = \varepsilon_0 \mathbf{I}$ is given by

$$\hat{\mathbf{e}}(\mathbf{k}) = \alpha_e \frac{\mathbf{k}\mathbf{k}}{k^2} \omega(\mathbf{k}) \quad (2.27)$$

where k is the magnitude of the vector \mathbf{k} , $k^2 = \mathbf{k} \cdot \mathbf{k}$, and α_e is some scalar constant; the exact expression for this constant is not important for our purposes here. The corresponding solution for the stress field in the case when the average stress field is hydrostatic $\tau_0 = \tau_0 \mathbf{I}$ is given by

$$\hat{\boldsymbol{\tau}}(\mathbf{k}) = \alpha_j \left(\mathbf{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right) \omega(\mathbf{k}), \quad (2.28)$$

where $\hat{\boldsymbol{\tau}}(\mathbf{k})$ is a Fourier coefficient of the stress field and α_j is a scalar constant.

To obtain three-point bounds, one can use trial fields of the form

$$\hat{\mathbf{e}}(\mathbf{x}) = \mathbf{e}_0 + \alpha_e \mathbf{e}_p(\mathbf{x}), \quad (2.29)$$

or

$$\hat{\mathbf{j}}(\mathbf{x}) = \mathbf{j}_0 + \alpha_j \mathbf{j}_p(\mathbf{x}), \quad (2.30)$$

in the variational principles (2.5) and (2.6). Here $\mathbf{e}_p(\mathbf{x})$ and $\mathbf{j}_p(\mathbf{x})$ are the first-order fluctuation parts of the fields $\mathbf{e}(\mathbf{x})$ and $\mathbf{j}(\mathbf{x})$. By using the Fourier representations (2.18)–(2.22) of the fields and the local properties, one can calculate the average energy of such trial fields (and thus bound the effective properties) in terms of integrals over the periodic cell that depend on the characteristic functions. For the bulk modulus bounds, the integrals are expressed in terms of the phase volume fraction $f_1 = 1 - f_2$ and the three-point geometrical parameter $\zeta_1 = 1 - \zeta_2$. Finally, one optimizes the resulting bounds with respect to the constants α_e or α_j to obtain tight three-point bounds on the effective properties. This procedure is described in a variety of papers, including Beran (1965), Beran and Molyneux (1966), McCoy (1970), Silnutzer (1972), Torquato (1980), Milton (1981a,b, 1982). Three-point bounds on the effective moduli of multiphase composites were obtained by Phan Thien and Milton (1982, 1983).

As we see, Beran-type bounds lack free parameters that are present in the translation method in the form of the translation matrix \mathbf{T} , but have the advantage that they use more precise trial fields. In the following sections, we will show how to integrate these methods into one procedure that will have advantages of both approaches. Before doing so, we will first introduce a special fractional linear transformation that allows us to simplify the form of the bounds.

2.4. *Y-transformation*

2.4.1. Definition and properties. The fractional linear *Y*-transformation [Cherkaev and Gibiansky (1992), Milton (1991b)] of the effective property tensor is given by

$$\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) = -f_2 \mathbf{F}_1 - f_1 \mathbf{F}_2 + f_1 f_2 (\mathbf{F}_1 - \mathbf{F}_2) : (f_1 \mathbf{F}_1 + f_2 \mathbf{F}_2 - \mathbf{F}_*)^{-1} : (\mathbf{F}_1 - \mathbf{F}_2). \quad (2.31)$$

Notice that this transformation enables one to express the inequality (2.17) in the more compact form

$$\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) + \mathbf{T}_j \geq 0. \quad (2.32)$$

The \mathbf{Y} -transformation possesses the following remarkable properties:

$$\mathbf{Y}(\mathbf{F}_*^{-1}, \mathbf{F}_1^{-1}, \mathbf{F}_2^{-1}) = \mathbf{Y}^{-1}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2), \quad (2.33)$$

$$\mathbf{Y}(\mathbf{F}_* - \mathbf{T}, \mathbf{F}_1 - \mathbf{T}, \mathbf{F}_2 - \mathbf{T}) = \mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) + \mathbf{T}, \quad (2.34)$$

$$\mathbf{Y}(\mathbf{F}_1, \mathbf{F}_1, \mathbf{F}_2) = -\mathbf{F}_1, \quad \mathbf{Y}(\mathbf{F}_2, \mathbf{F}_1, \mathbf{F}_2) = -\mathbf{F}_2. \quad (2.35)$$

These properties can be proved by a direct substitution of the expression (2.31) for the function $\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2)$ into the formulas (2.33)–(2.35). Note also that the function $\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2)$ is a monotonic function of its first argument, i.e.

$$\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) - \mathbf{Y}(\hat{\mathbf{F}}_*, \mathbf{F}_1, \mathbf{F}_2) \geq 0 \quad \text{if and only if } \mathbf{F}_* - \hat{\mathbf{F}}_* \geq 0. \quad (2.36)$$

Thus, the translation inequality (2.12) can be written as

$$\mathbf{Y}((\mathbf{F}_* - \mathbf{T}_j)^{-1}, (\mathbf{F}_1 - \mathbf{T}_j)^{-1}, (\mathbf{F}_2 - \mathbf{T}_j)^{-1}) \leq \mathbf{Y}((\hat{\mathbf{F}}_*^{-1}, (\mathbf{F}_1 - \mathbf{T}_j)^{-1}, (\mathbf{F}_2 - \mathbf{T}_j)^{-1}), \quad (2.37)$$

or in the equivalent form

$$[\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) + \mathbf{T}_j]^{-1} \leq \mathbf{Y}((\hat{\mathbf{F}}_*^{-1}, (\mathbf{F}_1 - \mathbf{T}_j)^{-1}, (\mathbf{F}_2 - \mathbf{T}_j)^{-1}) \quad (2.38)$$

that we will use in the following sections.

As was found by Milton (1991b), the \mathbf{Y} -tensor has a very natural variational interpretation. He suggested to decompose any field $\mathbf{e}(\mathbf{x})$ as a sum

$$\mathbf{e}(\mathbf{x}) = \mathbf{e}_0 + \mathbf{p}_e^*(\mathbf{x}) + \mathbf{P}_e(\mathbf{x}). \quad (2.39)$$

Here $\mathbf{e}_0 = \langle \mathbf{e}(\mathbf{x}) \rangle$ is the constant part of the field $\mathbf{e}(\mathbf{x})$. The field $\mathbf{p}_e^*(\mathbf{x})$ has a zero-average value, $\langle \mathbf{p}_e^*(\mathbf{x}) \rangle = 0$, and is constant in each of the phases, i.e.

$$\mathbf{p}_e^*(\mathbf{x}) = \begin{cases} \frac{1}{f_1} \langle \chi_1(\mathbf{x})(\mathbf{e}(\mathbf{x}) - \mathbf{e}_0) \rangle, & \text{if } \mathbf{x} \in \text{phase 1;} \\ \frac{1}{f_2} \langle \chi_2(\mathbf{x})(\mathbf{e}(\mathbf{x}) - \mathbf{e}_0) \rangle, & \text{if } \mathbf{x} \in \text{phase 2.} \end{cases} \quad (2.40)$$

Finally, $\mathbf{P}_e(\mathbf{x})$ is the remaining part of the $\mathbf{e}(\mathbf{x})$ with zero-average value and zero-averages in each phase, i.e.

$$\langle \chi_1(\mathbf{x}) \mathbf{P}_e(\mathbf{x}) \rangle = 0, \quad \langle \chi_2(\mathbf{x}) \mathbf{P}_e(\mathbf{x}) \rangle = 0. \quad (2.41)$$

The field $\mathbf{p}_e^*(\mathbf{x})$ can be written in the form

$$\mathbf{p}_e^*(\mathbf{x}) = (f_2 \chi_1(\mathbf{x}) - f_1 \chi_2(\mathbf{x})) \mathbf{v}_e = (\chi_1(\mathbf{x}) - f_1) \mathbf{v}_e, \quad (2.42)$$

where \mathbf{v}_e is a constant tensor. The constant tensor $\mathbf{Y}_D = \mathbf{Y}(\mathbf{D}_*, \mathbf{D}_1, \mathbf{D}_2)$ has a variational interpretation in terms of the components of the field \mathbf{e} . The \mathbf{Y}_D -tensor acts locally, is independent of \mathbf{x} and is defined via the relations

$$\langle \mathbf{p}_e^*(\mathbf{x}) : \mathbf{Y}_D : \mathbf{p}_e^*(\mathbf{x}) \rangle = \min_{\substack{\mathbf{P}_e(\mathbf{x}) \\ \langle \chi_e(\mathbf{x}) \mathbf{P}_e(\mathbf{x}) \rangle = 0 \\ \mathbf{p}_e^*(\mathbf{x}) + \mathbf{P}_e(\mathbf{x}) \in \mathcal{E}_e}} \langle \mathbf{P}_e(\mathbf{x}) : \mathbf{D}(\mathbf{x}) : \mathbf{P}_e(\mathbf{x}) \rangle. \quad (2.43)$$

Similarly, one can obtain the conjugate variational principle which defines the tensor $\mathbf{Y}_F = \mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) = \mathbf{Y}_D^{-1}$, i.e.

$$\langle \mathbf{p}_j^*(\mathbf{x}) : \mathbf{Y}_F : \mathbf{p}_j^*(\mathbf{x}) \rangle = \min_{\substack{\mathbf{P}_j(\mathbf{x}) \\ \langle \chi_j(\mathbf{x}) \mathbf{P}_j(\mathbf{x}) \rangle = 0 \\ \mathbf{p}_j^*(\mathbf{x}) + \mathbf{P}_j(\mathbf{x}) \in \mathcal{E}_j}} \langle \mathbf{P}_j(\mathbf{x}) : \mathbf{F}(\mathbf{x}) : \mathbf{P}_j(\mathbf{x}) \rangle, \quad (2.44)$$

where $\mathbf{p}_j^*(\mathbf{x})$ and $\mathbf{P}_j(\mathbf{x})$ are defined similar to $\mathbf{p}_e^*(\mathbf{x})$ and $\mathbf{P}_e(\mathbf{x})$ but for the $\mathbf{j}(\mathbf{x})$ -field, i.e.

$$\mathbf{j}(\mathbf{x}) = \mathbf{j}_0 + \mathbf{p}_j^*(\mathbf{x}) + \mathbf{P}_j(\mathbf{x}). \quad (2.45)$$

The variational definitions of the \mathbf{Y} -tensors allow one to obtain bounds on effective properties of a composite. For example, the inequality (2.32) can be proved directly by using the variational principle (2.44) [see Milton (1991b)].

2.4.2. Three-point bounds in terms of \mathbf{Y} -transformations. As was discussed and explored by Gibiansky and Torquato (1995b), the three-point bounds have a simpler form in terms of the \mathbf{Y} -transformations (2.31) of the effective moduli. In this section we will discuss the basis of this simplicity and obtain bounds directly in terms of the functions \mathbf{Y}_D or \mathbf{Y}_F .

Given the Fourier coefficients $\mathbf{E}(\mathbf{k})$ of the first-order perturbation solution $\hat{\mathbf{e}}(\mathbf{x})$, one can decompose it into three parts similar to (2.39). The first one is the constant average field \mathbf{e}_0 . The second part is the field $\mathbf{p}_e^*(\mathbf{x})$ which is constant in the phases and has average value equal to zero. This means that the Fourier coefficients $\hat{\mathbf{p}}_e^*(\mathbf{k})$ of the field $\mathbf{p}_e^*(\mathbf{x})$ are proportional to the Fourier coefficients $\omega(\mathbf{k})$ of the characteristic function $\chi_1(\mathbf{x})$ and can be written as

$$\hat{\mathbf{p}}_e^*(\mathbf{k}) = \omega(\mathbf{k}) \mathbf{v}_e, \quad \mathbf{k} \neq 0, \quad (2.46)$$

where \mathbf{v}_e is a constant vector [cf. (2.42)]. The third contribution is the remaining part $\mathbf{P}_e(\mathbf{x})$ of the perturbation solution. The Fourier coefficients $\hat{\mathbf{P}}_e(\mathbf{k})$ of this field are given by the difference

$$\hat{\mathbf{P}}_e(\mathbf{k}) = \mathbf{E}(\mathbf{k}) - \hat{\mathbf{p}}_e^*(\mathbf{k}), \quad \mathbf{k} \neq 0. \quad (2.47)$$

Substituting these fields into the variational principle (2.43), one can get the bound on the \mathbf{Y}_D tensor in the form

$$\langle \mathbf{p}_e^*(\mathbf{x}) : \mathbf{Y}_D : \mathbf{p}_e^*(\mathbf{x}) \rangle \leq \langle \mathbf{P}_e(\mathbf{x}) : \mathbf{D}(\mathbf{x}) : \mathbf{P}_e(\mathbf{x}) \rangle \quad (2.48)$$

Evaluation of the left- and right-hand sides of the inequality (2.48) is a matter of straightforward calculation. An arbitrary constant multiplying the fluctuating part of the trial field in the original formulation of the Beran method, cancels out in such a derivation. This allows us to avoid the optimization over this constant. In Section 5 we will show how to apply this procedure to the viscoelasticity problem.

Remark : Note that the trial field $\hat{\mathbf{e}}(\mathbf{x})$ may depend on more than one constant, e.g. for the shear modulus bounds. In this case only one of these constants cancels out in our derivation. We will still need to optimize the bound over the remaining constants.

2.5. New method

In the preceding sections we collected all of the ingredients that are necessary to formulate our new bounding procedure. In this section we give a step-by-step prescription on how to obtain the new bounds.

In order to find the upper bound on the effective properties tensor \mathbf{D}_* (or, equivalently, the lower bound on the effective properties tensor \mathbf{F}_*) we will

- study the differential properties of the fields $\mathbf{j}(\mathbf{x})$ and find the appropriate translation matrices \mathbf{T}_j that correspond to the quasiconvex quadratic forms of the fields $\mathbf{j}(\mathbf{x})$;
- consider the “translated” composite with the phase properties $\hat{\mathbf{F}}_1 = \mathbf{F}_1 - \mathbf{T}_j$ and $\hat{\mathbf{F}}_2 = \mathbf{F}_2 - \mathbf{T}_j$ and composite properties $\hat{\mathbf{F}}_*$, or equivalently, $\hat{\mathbf{D}}_1 = (\mathbf{F}_1 - \mathbf{T}_j)^{-1}$, $\hat{\mathbf{D}}_2 = (\mathbf{F}_2 - \mathbf{T}_j)^{-1}$, and $\hat{\mathbf{D}}_* = \hat{\mathbf{F}}_*^{-1}$;
- use the translation inequality (2.12) in the form (2.38) and the variational principle (2.43) for the translated composite in the form

$$\langle \mathbf{p}_e^*(\mathbf{x}) : \mathbf{Y}(\hat{\mathbf{F}}_*^{-1}, (\mathbf{F}_1 - \mathbf{T}_j)^{-1}, (\mathbf{F}_2 - \mathbf{T}_j)^{-1}) : \mathbf{p}_e^*(\mathbf{x}) \rangle \leq \langle \mathbf{P}_e(\mathbf{x}) : (\mathbf{F}(\mathbf{x}) - \mathbf{T}_j)^{-1} : \mathbf{P}_e(\mathbf{x}) \rangle \quad (2.49)$$

to get the inequality

$$\langle \mathbf{p}_e^*(\mathbf{x}) [\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) + \mathbf{T}_j]^{-1} : \mathbf{p}_e^*(\mathbf{x}) \rangle \leq \langle \mathbf{P}_e(\mathbf{x}) : (\mathbf{F}(\mathbf{x}) - \mathbf{T}_j)^{-1} : \mathbf{P}_e(\mathbf{x}) \rangle, \quad (2.50)$$

where $\mathbf{p}_e^*(\mathbf{x})$ and $\mathbf{P}_e(\mathbf{x})$ are defined by (2.39) for any trial field $\hat{\mathbf{e}}(\mathbf{x})$.

- find the Fourier representation of the first-order perturbation solution $\hat{\mathbf{e}}(\mathbf{x})$ and decompose it into the parts $\hat{\mathbf{p}}_e^*(\mathbf{k})$ and $\hat{\mathbf{P}}_e(\mathbf{k})$;
- evaluate the left- and right-hand sides of the inequality (2.50) with the trial fields $\hat{\mathbf{p}}_e^*(\mathbf{k})$ and $\hat{\mathbf{P}}_e(\mathbf{k})$ in terms of the geometrical parameters $\zeta_1 = 1 - \zeta_2$.

As a result of the described procedure, we will arrive at the required lower bound on the effective properties tensor \mathbf{F}_* .

In order to get the other bound, one needs to repeat the above outlined procedure for the dual problem, i.e. study the differential properties of the $\mathbf{e}(\mathbf{x})$ field, find the translation matrices \mathbf{T}_e that correspond to the quasiconvex quadratic forms of the $\mathbf{e}(\mathbf{x})$ -fields, translate the tensors \mathbf{D}_i by the translation \mathbf{T}_e , invert the translated phase properties and use variational principle (2.44) instead of (2.43).

In the next section we apply the suggested method to the important problem of finding geometrical-parameters bounds on the effective complex bulk modulus of a two-phase viscoelastic composite.

Remarks :

- (i) In general, one should consider the problem with “translated” phases in order to find the perturbation solution. However, in the viscoelasticity problem that we will

study in the next section, one can use the trial fields found for the pure elastic composite without any translation.

(ii) Independently, Milton (1997) has obtained what he called the Hashin–Shtrikman variational inequality for the \mathbf{Y} -tensor which can be reduced to (2.50) by appropriate choice of the trial polarization field and the reference medium.

3. BOUNDS ON COMPLEX BULK MODULUS OF VISCOELASTIC COMPOSITE

In this section we obtain bounds on the effective complex moduli of viscoelastic composites. Although an important practical problem, progress in bounding viscoelastic moduli has been limited relative to the corresponding pure elasticity problem. The early literature on the subject is very sparse [Hashin (1965, 1970), Christensen (1969), Roscoe (1969, 1972)]. The reason for this was the lack of appropriate minimum variational principles that describe the complex elasticity problem. The situation changed when such variational principles were discovered by Cherkaev and Gibiansky (1994) [see also Milton (1990) and Fannjiang and Papanicolaou (1994)]. This enables one to apply any of the variational methods to bound effective moduli of viscoelastic composites. In particular, Gibiansky and Milton (1993a,b) have found complex moduli bounds by using the Hashin–Shtrikman and the translation methods, and Milton and Berryman (1997) obtained complex shear modulus bounds by using the Hashin–Shtrikman approach. In this section we apply our new method to obtain three-point complex bulk moduli bounds.

A more detailed review can be found in the paper by Gibiansky and Milton (1993a).

3.1. *Viscoelasticity: statement and variational principles*

We consider the steady-state harmonic oscillations in a linear viscoelastic medium. A constitutive relation for the harmonic oscillations of a viscoelastic material in the quasistatic limit is given by

$$\boldsymbol{\tau} = \mathcal{C} : \boldsymbol{\varepsilon}, \quad (3.1)$$

where $\boldsymbol{\tau} = \boldsymbol{\tau}' + i\boldsymbol{\tau}''$, $i = \sqrt{-1}$, and $\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}' + i\boldsymbol{\varepsilon}''$ are the complex stress and strain fields, and $\mathcal{C} = \mathcal{C}' + i\mathcal{C}''$ is a complex fourth-order stiffness tensor [Christensen (1971)]. Here and below a' and a'' denote the real and imaginary parts of the variable $a = a' + ia''$. The complex strain field $\boldsymbol{\varepsilon}$ is the symmetric part of the gradient of the complex displacement field \mathbf{u} , i.e.

$$\boldsymbol{\varepsilon} = \frac{1}{2}(\nabla \mathbf{u} + (\nabla \mathbf{u})^T), \quad (3.2)$$

the stress field $\boldsymbol{\tau}$ is divergence-free, i.e.

$$\nabla \cdot \boldsymbol{\tau} = 0. \quad (3.3)$$

The imaginary part of the complex stiffness tensor is nonnegative $\mathcal{C}'' \geq 0$ for media with nonnegative dissipation. We will also assume without loss of generality that $\boldsymbol{\tau}$, $\boldsymbol{\varepsilon}$, and \mathcal{C} are periodic.

The complex-valued equation (3.1) can be reformulated in terms of the real quantities as

$$\mathbf{j} = \mathbf{D} : \mathbf{e}, \quad (3.4)$$

where

$$\mathbf{j} = \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\tau}'' \end{pmatrix}, \quad \mathbf{e} = \begin{pmatrix} -\boldsymbol{\tau}' \\ \boldsymbol{\varepsilon}' \end{pmatrix}, \quad (3.5)$$

and

$$\mathbf{D} = \begin{pmatrix} (\mathcal{C}'')^{-1} & (\mathcal{C}'')^{-1} \mathcal{C}' \\ \mathcal{C}'(\mathcal{C}'')^{-1} & \mathcal{C}'' + \mathcal{C}'(\mathcal{C}'')^{-1} \mathcal{C}' \end{pmatrix}. \quad (3.6)$$

Remark: The matrix \mathbf{D} that appears in the constitutive equation (3.4) is a $d(d+1)$ -by- $d(d+1)$ matrix (where d is the spatial dimension) which can be written as a two-by-two matrix with elements that are symmetric fourth-order tensors. In the remaining part of the paper, we denote such matrices by bold capital letters, e.g. \mathbf{D} , \mathbf{T} , \mathbf{F} , etc. The fourth-order tensors will be denoted by bold capital Greek or by calligraphic letters, e.g. \mathcal{C} , \mathcal{A} , \mathcal{Y} , $\mathbf{\Lambda}$, etc, and two-by-two matrices will be denoted by capital letters, e.g. D^h , T^s , F^s , etc.

There are two complementary minimum variational principles [see Cherkaev and Gibiansky (1994), Gibiansky and Milton (1993a)] describing the viscoelasticity problem, namely,

$$\mathbf{e}_0 : \mathbf{D}_* : \mathbf{e}_0 \leq \langle \mathbf{e} : \mathbf{D}(\mathbf{x}) : \mathbf{e} \rangle, \quad \forall \mathbf{e} \in \mathcal{E}_e, \quad (3.7)$$

where \mathbf{e} is any admissible \mathbf{e} -field with given average value \mathbf{e}_0 and

$$\mathbf{j}_0 : \mathbf{F}_* : \mathbf{j}_0 \leq \langle \mathbf{j} : \mathbf{F}(\mathbf{x}) : \mathbf{j} \rangle \quad \forall \mathbf{j} \in \mathcal{E}_j, \quad (3.8)$$

where \mathbf{j} is any admissible \mathbf{j} -field with given average value \mathbf{j}_0 . Here

$$\mathbf{F} = \mathbf{D}^{-1} = \begin{pmatrix} \mathcal{C}'' + \mathcal{C}'(\mathcal{C}'')^{-1} \mathcal{C}' & -\mathcal{C}'(\mathcal{C}'')^{-1} \\ -(\mathcal{C}'')^{-1} \mathcal{C}' & (\mathcal{C}'')^{-1} \end{pmatrix}. \quad (3.9)$$

“Admissible fields” here means that the strain-type components of the \mathbf{e} - and \mathbf{j} -fields are the symmetric parts of the gradients of some displacement vectors, and the stress-type components of the \mathbf{e} and \mathbf{j} fields are symmetric and divergence-free, i.e.

$$\mathbf{e} \in \mathcal{E}_e = \left\{ \begin{pmatrix} -\boldsymbol{\tau}' \\ \boldsymbol{\varepsilon}' \end{pmatrix} \text{ such that } \begin{array}{l} \nabla \cdot \boldsymbol{\tau}' = 0 \\ \boldsymbol{\varepsilon}' = \frac{1}{2}[\nabla \mathbf{u}' + (\nabla \mathbf{u}')^T] \end{array} \right\}, \quad (3.10)$$

$$\mathbf{j} \in \mathcal{E}_j = \left\{ \begin{pmatrix} \boldsymbol{\varepsilon}'' \\ \boldsymbol{\tau}'' \end{pmatrix} \text{ such that } \begin{array}{l} \boldsymbol{\varepsilon}'' = \frac{1}{2}[\nabla \mathbf{u}'' + (\nabla \mathbf{u}'')^T] \\ \nabla \cdot \boldsymbol{\tau}'' = 0 \end{array} \right\}. \quad (3.11)$$

Remark: Note that the property matrix \mathbf{D} for the viscoelasticity problem has a very special structure, specifically, $\det \mathbf{D} = 1$, and therefore the matrix \mathbf{D} and inverse matrix $\mathbf{F} = \mathbf{D}^{-1}$ are very similar [compare (3.6) and (3.9)].

We will study an isotropic composite comprised of isotropic phases. The stiffness tensor of an isotropic material can be expressed in the form

$$\mathcal{C}(\mathrm{d}\kappa, 2\mu) = \mathcal{A}(\mathrm{d}\kappa, 2\mu) = \mathrm{d}\kappa\mathbf{\Lambda}_h + 2\mu\mathbf{\Lambda}_s, \quad (3.12)$$

where κ and μ are the bulk and the shear moduli of the material, respectively, $\mathcal{A}(\lambda_1, \lambda_2)$ is a symbolic notation of the arbitrary isotropic fourth-order symmetric tensor in the form

$$\{\mathcal{A}(\lambda_1, \lambda_2)\}_{ijkl} = (\lambda_1/\mathrm{d})\delta_{ij}\delta_{kl} + (\lambda_2/2)(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - (2/\mathrm{d})\delta_{ij}\delta_{kl}, \quad (3.13)$$

and

$$\mathbf{\Lambda}_h = \frac{1}{\mathrm{d}}\delta_{ij}\delta_{kl}, \quad \mathbf{\Lambda}_s = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \frac{1}{\mathrm{d}}\delta_{ij}\delta_{kl} \quad (3.14)$$

are the fourth-order projector tensors on the space of pure hydrostatic and pure shear (trace-free) fields, respectively.

By using such a notation the property tensors \mathbf{D} and \mathbf{F} of an isotropic viscoelastic material can be expressed as

$$\mathbf{D} = D^h\mathbf{\Lambda}_h + D^s\mathbf{\Lambda}_s, \quad \mathbf{F} = \mathbf{D}^{-1} = F^h\mathbf{\Lambda}_h + F^s\mathbf{\Lambda}_s, \quad (3.15)$$

where

$$D^h = \begin{bmatrix} \frac{1}{\mathrm{d}\kappa''} & \frac{\kappa'}{\kappa''} \\ \frac{\kappa'}{\kappa''} & \mathrm{d} \frac{(\kappa')^2 + (\kappa'')^2}{\kappa''} \end{bmatrix}, \quad D^s = \begin{bmatrix} \frac{1}{2\mu''} & \frac{\mu'}{\mu''} \\ \frac{\mu'}{\mu''} & 2 \frac{(\mu')^2 + (\mu'')^2}{\mu''} \end{bmatrix}, \quad (3.16)$$

$$F^h = \begin{bmatrix} \mathrm{d} \frac{(\kappa')^2 + (\kappa'')^2}{\kappa''} & -\frac{\kappa'}{\kappa''} \\ -\frac{\kappa'}{\kappa''} & \frac{1}{\mathrm{d}\kappa''} \end{bmatrix}, \quad F^s = \begin{bmatrix} 2 \frac{(\mu')^2 + (\mu'')^2}{\mu''} & -\frac{\mu'}{\mu''} \\ -\frac{\mu'}{\mu''} & \frac{1}{2\mu''} \end{bmatrix}, \quad (3.17)$$

and symbolic notation $D^h\mathbf{\Lambda}_h$ means that we multiply each element of the matrix D^h by the tensor $\mathbf{\Lambda}_h$. We will denote by $\kappa_1 = \kappa'_1 + \kappa''_1$, $\kappa_2 = \kappa'_2 + \kappa''_2$ and $\kappa_* = \kappa'_* + \kappa''_*$ the complex bulk moduli of the first and the second phases and the composite, respectively. Similarly, we will denote by $\mu_1 = \mu'_1 + \mu''_1$, $\mu_2 = \mu'_2 + \mu''_2$, and $\mu_* = \mu'_* + \mu''_*$ the corresponding complex shear moduli.

3.2. Bounds: general procedure

Let us now follow the prescription of Section 2.5 to obtain new bounds. We will address each step of the procedure giving results that are specific to the viscoelasticity complex bulk modulus bounds.

3.2.1. *Translation matrices.* Consider the matrix

$$\mathbf{T}_j = \mathbf{T}_j(t_1, t_2, t_3) = \begin{pmatrix} \mathcal{A}(-(d-1)t_1, t_1) & \mathcal{A}(-t_3, -t_3) \\ \mathcal{A}(-t_3, -t_3) & \mathcal{A}(-t_2, (d-1)t_2) \end{pmatrix} \quad (3.18)$$

and the quadratic form $\mathbf{j} : \mathbf{T}_j : \mathbf{j}$ of the vector $\mathbf{j} = (\varepsilon'' \boldsymbol{\tau}'')^T$ associated with such a matrix \mathbf{T}_j . One can show (see, e.g. Gibiansky and Milton, 1993a) that this form is quasiconvex, i.e.

$$\langle \mathbf{j} \rangle : \mathbf{T}_j : \langle \mathbf{j} \rangle \leq \langle \mathbf{j} : \mathbf{T}_j : \mathbf{j} \rangle, \quad \forall \mathbf{j} \in \mathcal{E}_j, \quad (3.19)$$

for any admissible \mathbf{j} -field, and for any positive values of the parameters $t_1 \geq 0$ and $t_2 \geq 0$ and any (positive or negative) value of the parameter t_3 . In two dimensions, this form is quasiconvex for any positive value of the parameter $t_1 \geq 0$, and any (positive or negative) values of the parameters t_2 and t_3 . Summarizing, one can state that

$$\mathbf{j} : \mathbf{T}_j : \mathbf{j} \text{ is quasiconvex for any set } \{t_1, t_2, t_3\} : \begin{cases} t_1 \geq 0, & t_2 \geq 0, & \forall t_3, & \text{if } d = 3, \\ t_1 \geq 0, & \forall t_2, & \forall t_3, & \text{if } d = 2. \end{cases} \quad (3.20)$$

Note that the translation matrix \mathbf{T}_j can be presented in the form

$$\mathbf{T}_j = \mathbf{T}_j^h \mathbf{\Lambda}_h + T_j^s \mathbf{\Lambda}_s, \quad (3.21)$$

where

$$T_j^h = \begin{pmatrix} -(d-1)t_1 & -t_3 \\ -t_3 & -t_2 \end{pmatrix}, \quad T_j^s = \begin{pmatrix} t_1 & -t_3 \\ -t_3 & (d-1)t_2 \end{pmatrix}. \quad (3.22)$$

3.2.2. *Translated composite.* Consider two composites with identical microstructure. The first one consists of isotropic viscoelastic phases \mathbf{F}_1 and \mathbf{F}_2 [see (3.16)] and possesses the effective properties \mathbf{F}_* , and the second one contains phases translated by the matrix \mathbf{T}_j , i.e. $\hat{\mathbf{F}}_1 = \mathbf{F}_1 - \mathbf{T}_j \geq 0$, $\hat{\mathbf{F}}_2 = \mathbf{F}_2 - \mathbf{T}_j \geq 0$, and possesses the effective properties \mathbf{F}_* . Note that the translated phases may not correspond to any viscoelastic material; they just need to be nonnegative.

3.2.3. *Translation inequality.* The only difficulty in evaluating the inequality (2.50) for the problem under study is the calculation of the Y -transformation of the effective properties tensor \mathbf{F}_* . Gibiansky and Milton (1993a) have shown that

$$\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) = \begin{pmatrix} \mathcal{Y}_c'' + \mathcal{Y}_c'(\mathcal{Y}_c'')^{-1}\mathcal{Y}_c' & \mathcal{Y}_c'(\mathcal{Y}_c'')^{-1} \\ (\mathcal{Y}_c'')^{-1}\mathcal{Y}_c' & (\mathcal{Y}_c'')^{-1} \end{pmatrix}, \quad (3.23)$$

[cf. (3.9) and note a plus sign for the off-diagonal elements]. Here the complex tensor $\mathcal{Y}_c = \mathcal{Y}_c' + i\mathcal{Y}_c''$ is defined via

$$\mathcal{Y}_c = -f_2 \mathcal{C}_1 - f_1 \mathcal{C}_2 + f_1 f_2 (\mathcal{C}_1 - \mathcal{C}_2) : (f_1 \mathcal{C}_1 + f_2 \mathcal{C}_2 - \mathcal{C}_*)^{-1} : (\mathcal{C}_1 - \mathcal{C}_2), \quad (3.24)$$

where \mathcal{C}_* , \mathcal{C}_1 , and \mathcal{C}_2 are complex stiffness tensors of the composite, phase 1, and phase 2, respectively. For an isotropic composite,

$$\mathcal{Y}_c = \mathcal{A}(\mathrm{d}y_\kappa, 2y_\mu), \quad (3.25)$$

where

$$y_\kappa = y'_\kappa + iy''_\kappa = -f_1\kappa_2 - f_2\kappa_1 + \frac{f_1f_2(\kappa_1 - \kappa_2)^2}{f_1\kappa_1 + f_2\kappa_2 - \kappa_*} \quad (3.26)$$

and

$$y_\mu = y'_\mu + iy''_\mu = -f_1\mu_2 - f_2\mu_1 + \frac{f_1f_2(\mu_1 - \mu_2)^2}{f_1\mu_1 + f_2\mu_2 - \mu_*} \quad (3.27)$$

are the Y -transformations of the effective complex bulk and shear moduli, respectively. Thus, the tensors $\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2)$ of an isotropic composite can be expressed as

$$\mathbf{Y}(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) = Y_F^h \mathbf{\Lambda}_h + Y_F^s \mathbf{\Lambda}_s, \quad (3.28)$$

where

$$Y_F^h = \begin{bmatrix} \mathrm{d} \frac{(y'_\kappa)^2 + (y''_\kappa)^2}{y''_\kappa} & \frac{y'_\kappa}{y''_\kappa} \\ \frac{y'_\kappa}{y''_\kappa} & \frac{1}{\mathrm{d}y''_\kappa} \end{bmatrix}, \quad Y_F^s = \begin{bmatrix} 2 \frac{(y''_\mu)^2 + (y'_\mu)^2}{y''_\mu} & \frac{y'_\mu}{y''_\mu} \\ \frac{y'_\mu}{y''_\mu} & \frac{1}{2y''_\mu} \end{bmatrix}. \quad (3.29)$$

[cf. (3.15), (3.17)].

3.2.4. Trial fields. It remains to find an appropriate trial field to get the explicit expression for the bounds in (2.50). For the viscoelasticity problem, the necessary trial field $\mathbf{e}(\mathbf{x})$ is the pair of the stress and strain fields, i.e.

$$\mathbf{e}(\mathbf{x}) = (-\boldsymbol{\tau}(\mathbf{x}) \quad \boldsymbol{\varepsilon}(\mathbf{x}))^T, \quad (3.30)$$

where we omitted the notation for the real part of the fields. Henceforth, in this section we will deal only with real quantities.

Perturbation trial fields to bound the bulk and shear moduli of the pure elastic composite were found by Beran and Molyneux (1966) and McCoy (1970). Milton (1981b, 1982) significantly simplified the derivation. He found that the Fourier coefficients $\hat{\tau}$ and $\hat{\varepsilon}$ of the first-order terms for the hydrostatic average fields are given by the expressions

$$\hat{\tau}(\mathbf{k}) = \tau \left(\mathbf{I} - \frac{\mathbf{k}\mathbf{k}}{k^2} \right) \omega(\mathbf{k}) = \hat{\mathbf{p}}_r^*(\mathbf{k}) + \hat{\mathbf{p}}_r(\mathbf{k}), \quad (3.31)$$

$$\hat{\varepsilon}(\mathbf{k}) = \varepsilon \frac{\mathbf{k}\mathbf{k}}{k^2} \omega(\mathbf{k}) = \hat{\mathbf{p}}_e^*(\mathbf{k}) + \hat{\mathbf{P}}_e(\mathbf{k}), \quad (3.32)$$

where

$$\hat{\mathbf{p}}_\tau^*(\mathbf{k}) = \frac{d-1}{d} \tau \omega(\mathbf{k}) \mathbf{I}, \quad \hat{\mathbf{P}}_\tau(\mathbf{k}) = -\tau \frac{\mathbf{k}\mathbf{k} - \mathbf{I}k^2/d}{k^2} \omega(\mathbf{k}), \quad (3.33)$$

$$\hat{\mathbf{p}}_e^*(\mathbf{k}) = \frac{1}{d} \varepsilon \omega(\mathbf{k}) \mathbf{I}, \quad \hat{\mathbf{P}}_e(\mathbf{k}) = \varepsilon \frac{\mathbf{k}\mathbf{k} - \mathbf{I}k^2/d}{k^2} \omega(\mathbf{k}). \quad (3.34)$$

Note that the Fourier coefficients $\hat{\mathbf{p}}_\tau^*(\mathbf{k})$ and $\hat{\mathbf{p}}_e^*(\mathbf{k})$ are proportional to the unit tensor, i.e. are hydrostatic, whereas the Fourier coefficients $\hat{\mathbf{P}}_\tau(\mathbf{k})$ and $\hat{\mathbf{P}}_e(\mathbf{k})$ are pure shear (trace-free) fields in Fourier space. Now we have to show that the decompositions (3.31)–(3.34) indeed satisfy the properties required by the variational principle (2.43).

First, we note that the Fourier coefficients $\hat{\mathbf{p}}_\tau^*(\mathbf{k})$ and $\hat{\mathbf{p}}_e^*(\mathbf{k})$ are proportional to the Fourier coefficients $\omega(\mathbf{k})$ of the characteristic function $\chi_1(\mathbf{x})$ [see (3.33) and (3.34)]. Therefore, the corresponding fields $\mathbf{p}_\tau^*(\mathbf{x})$ and $\mathbf{p}_e^*(\mathbf{x})$ are constant in each of the phases. Second, for statistically homogeneous composites,

$$\langle \chi_1(\mathbf{x}) \mathbf{P}_\tau(\mathbf{x}) \rangle = -\tau \sum_{\mathbf{k} \neq 0} \frac{\mathbf{k}\mathbf{k} - \mathbf{I}k^2/d}{k^2} \omega(\mathbf{k}) \omega(-\mathbf{k}) = 0. \quad (3.35)$$

Indeed, for such composites, the Fourier coefficients $\omega(\mathbf{k})$ of the characteristic function $\chi_1(\mathbf{x})$ should not depend on the direction of the wave vector. Thus, the entire sum on the right-hand side of (3.35) should be isotropic, i.e. proportional to the unit tensor. As one can easily see, the trace of this sum is equal to zero, i.e.

$$\text{Tr} \left[\sum_{\mathbf{k} \neq 0} \frac{\mathbf{k}\mathbf{k} - \mathbf{I}k^2/d}{k^2} \omega(\mathbf{k}) \omega(-\mathbf{k}) \right] = \sum_{\mathbf{k} \neq 0} \frac{\text{Tr}[\mathbf{k}\mathbf{k}] - k^2}{k^2} \omega(k) \omega(-k) = 0, \quad (3.36)$$

thus proving (3.35). Therefore, the decompositions (3.31) and (3.32) are indeed the decompositions of the trial fields into parts proportional to fields constant in the phases, and fields with zero average in each of the phases.

For the complex bulk modulus bounds, we will use the trial field in the form

$$\mathbf{e}(\mathbf{x}) = \mathbf{e}_0 + \mathbf{p}_e^*(\mathbf{x}) + \mathbf{P}_e(\mathbf{x}), \quad \mathbf{e}_0 = \begin{pmatrix} -\tau_0 \\ \varepsilon_0 \end{pmatrix}, \quad \mathbf{p}_e^*(\mathbf{x}) = \begin{pmatrix} -\mathbf{p}_\tau^*(\mathbf{x}) \\ \mathbf{p}_e^*(\mathbf{x}) \end{pmatrix}, \quad \mathbf{P}_e(\mathbf{x}) = \begin{pmatrix} -\mathbf{P}_\tau(\mathbf{x}) \\ \mathbf{P}_e(\mathbf{x}) \end{pmatrix}, \quad (3.37)$$

where the Fourier coefficients of the fields $\mathbf{p}_e^*(\mathbf{x})$, $\mathbf{p}_\tau^*(\mathbf{x})$, $\mathbf{P}_e(\mathbf{x})$, and $\mathbf{P}_\tau(\mathbf{x})$ are given by equations (3.33) and (3.34).

3.2.5. Evaluation of the bound. We will evaluate separately the right-hand and left-hand sides of the bound (2.50). Substituting the expressions (3.33), (3.34) for the Fourier coefficients $\hat{\mathbf{p}}_e^*(\mathbf{k})$ of the field $\mathbf{p}_e^*(\mathbf{x})$, one finds that

$$\begin{aligned}
\langle \mathbf{p}_e^*(\mathbf{x}) : (\mathbf{Y}_*(\mathbf{F}_*, \mathbf{F}_1, \mathbf{F}_2) + \mathbf{T}_j)^{-1} : \mathbf{p}_e^*(\mathbf{x}) \rangle &= \mathbf{I} : \mathbf{\Lambda}_h : \mathbf{I} \sum_{\mathbf{k} \neq 0} \omega(\mathbf{k}) \omega(-\mathbf{k}) \\
&\times \begin{bmatrix} -\frac{d-1}{d} \tau \\ \frac{1}{d} \varepsilon \end{bmatrix} \left[\begin{bmatrix} d \frac{(y''_{\kappa})^2 + (y'_{\kappa})^2}{y''_{\kappa}} & \frac{y'_{\kappa}}{y''_{\kappa}} \\ \frac{y'_{\kappa}}{y''_{\kappa}} & \frac{1}{dy''_{\kappa}} \end{bmatrix} - \begin{pmatrix} (d-1)t_1 & t_3 \\ t_3 & t_2 \end{pmatrix} \right]^{-1} \begin{bmatrix} -\frac{d-1}{d} \tau \\ \frac{1}{d} \varepsilon \end{bmatrix} \\
&= f_1 f_2 \begin{pmatrix} \tau \\ \varepsilon \end{pmatrix} \left[\begin{bmatrix} \frac{d^2}{(d-1)^2} \frac{(y''_{\kappa})^2 + (y'_{\kappa})^2}{y''_{\kappa}} & -\frac{d}{d-1} \frac{y'_{\kappa}}{y''_{\kappa}} \\ -\frac{d}{d-1} \frac{y'_{\kappa}}{y''_{\kappa}} & \frac{1}{y''_{\kappa}} \end{bmatrix} \right. \\
&\quad \left. - \begin{bmatrix} \frac{d}{d-1} t_1 & -\frac{d}{d-1} t_1 \\ -\frac{d}{d-1} t_1 & dt_1 \end{bmatrix} \right]^{-1} \begin{pmatrix} \tau \\ \varepsilon \end{pmatrix}, \quad (3.38)
\end{aligned}$$

where we used the relations

$$\sum_{\mathbf{k} \neq 0} \omega(\mathbf{k}) \omega(-\mathbf{k}) = f_1 f_2, \quad \mathbf{I} : \mathbf{\Lambda}_h : \mathbf{I} = d \quad (3.39)$$

and the notation y_{κ} for the Y -transformation of the effective complex bulk modulus κ_* [see (3.26)].

To evaluate the right-hand side of the inequality (2.50) we mention that for any $\mathbf{P}(\mathbf{x})$ with the Fourier coefficients given by

$$\hat{\mathbf{P}}(\mathbf{k}) = \frac{\mathbf{k}\mathbf{k} - \mathbf{I}k^2/d}{k^2} \omega(\mathbf{k}) \quad (3.40)$$

[compare with (3.33) and (3.34)], the following equality holds

$$\langle \chi_i(\mathbf{x}) \mathbf{P}(\mathbf{x}) : \mathbf{\Lambda}_s : \mathbf{P}(\mathbf{x}) \rangle = f_1 f_2 \frac{d-1}{d} \zeta_i, \quad i = 1, 2, \quad (3.41)$$

where ζ_1 and $\zeta_2 = 1 - \zeta_1$ are the three-point geometrical parameters [see Milton (1982)].

Then one can deduce that

$$\langle \mathbf{P}_e(\mathbf{x}) : (\mathbf{F}(\mathbf{x}) - \mathbf{T}_j)^{-1} : \mathbf{P}_e(\mathbf{x}) \rangle = f_1 f_2 \frac{d-1}{d} \begin{pmatrix} \tau \\ \varepsilon \end{pmatrix} [\zeta_1 (F_1^s - T_j^s)^{-1} + \zeta_2 (F_2^s - T_j^s)^{-1}] \begin{pmatrix} \tau \\ \varepsilon \end{pmatrix}, \quad (3.42)$$

where F_1^s and F_2^s are defined similar to (3.17), and T_j^s is given by (3.22).

Now we combine our findings, i.e. expressions (3.38) for the left-hand side of the bound (2.50), and expression (3.42) for the right-hand side of the bound. The ampli-

tudes τ and ε of the strains and stress fields are arbitrary. Therefore, one can use the bounds for the quadratic forms to get (after some simple calculations) the matrix inequality

$$(Y_\kappa(\kappa_*) - T_\kappa)^{-1} \leq \zeta_1(Y_1 - T_\kappa)^{-1} + \zeta_2(Y_2 - T_\kappa)^{-1}, \quad (3.43)$$

where

$$Y_\kappa = \begin{bmatrix} \frac{(y''_\kappa)^2 + (y'_\kappa)^2}{y''_\kappa} & -\frac{y'_\kappa}{y''_\kappa} \\ -\frac{y'_\kappa}{y''_\kappa} & \frac{1}{y''_\kappa} \end{bmatrix}, \quad T_\kappa = \begin{pmatrix} \frac{d-1}{d} t_1 & -t_3 \\ -t_3 & dt_2 \end{pmatrix}, \quad (3.44)$$

and

$$Y_i = \begin{bmatrix} \frac{2(d-1)}{d} \frac{(\mu'_i)^2 + (\mu''_i)^2}{\mu''_i} & -\frac{\mu'_i}{\mu''_i} \\ -\frac{\mu'_i}{\mu''_i} & \frac{d}{2(d-1)} \frac{1}{\mu''_i} \end{bmatrix}, \quad i = 1, 2. \quad (3.45)$$

Remark: Note that the “origin” of the matrix T_κ on the left-hand and right-hand sides of equation (3.43) differ. Namely, on the left-hand side of (3.43), the matrix T_κ appears from the *bulk part* of the translation matrix \mathbf{T}_j , whereas on the right-hand side it appeared from the *shear part* of the matrix \mathbf{T}_j . Although formally the inequality (3.43) is very similar to the translation inequality (2.17), it is not as general. Indeed, in the translation inequality, the same translation matrix enters on both sides of the inequality. On the contrary, in (3.43) the matrices $T_\kappa^r = T_\kappa$ on the right-hand side and $T_\kappa^l = T_\kappa$ on the left-hand side are equal but not identical in the sense that they came from different projections of the translation matrix \mathbf{T}_j . It is not clear at the moment whether the equality $T_\kappa^r = T_\kappa^l$ is a general rule or a specific feature of some class of problems.

3.2.6. Simplification of the bound via Z-transformation. The bound (3.43) has the same form as a translation inequality (2.17) with the only difference being that the volume fractions f_1 and f_2 are replaced by the geometrical parameters ζ_1 and ζ_2 . Therefore, it makes sense to introduce the function

$$Z(Y_\kappa, Y_1, Y_2) = -\zeta_2 Y_1 - \zeta_1 Y_2 + \zeta_1 \zeta_2 (Y_1 - Y_2) : (\zeta_1 Y_1 + \zeta_2 Y_2 - Y_\kappa)^{-1} : (Y_1 - Y_2), \quad (3.46)$$

which can be seen to be similar to the Y -transformation (2.31). Here $:$ denotes matrix multiplication. Then, the inequality (3.43) reduces to

$$Z_\kappa + T_\kappa \geq 0, \quad (3.47)$$

where the matrix T_κ is given by (3.44) and

$$Z_{\kappa} = \begin{bmatrix} \frac{(z''_{\kappa})^2 + (z'_{\kappa})^2}{z''_{\kappa}} & \frac{z'_{\kappa}}{z''_{\kappa}} \\ \frac{z'_{\kappa}}{z''_{\kappa}} & \frac{1}{z''_{\kappa}} \end{bmatrix}. \quad (3.48)$$

Here

$$z_{\kappa} = z'_{\kappa} + iz''_{\kappa} \quad (3.49)$$

in the Z -transformation of the complex scalar quantity y_{κ}

$$z_{\kappa} = -\zeta_1 y_2 - \zeta_2 y_1 + \frac{\zeta_1 \zeta_2 (y_1 - y_2)^2}{\zeta_1 y_1 + \zeta_2 y_2 - y_{\kappa}}, \quad (3.50)$$

$$y_1 = \frac{2(d-1)}{d} \mu_1, \quad y_2 = \frac{2(d-1)}{d} \mu_2 \quad (3.51)$$

and y_{κ} , defined by (3.26), is the Y -transformation of the effective complex bulk modulus κ_* .

The inequality (3.47) holds for any matrix T_{κ} corresponding to the admissible translation matrix \mathbf{T} that depends on three parameters t_1 , t_2 and t_3 . These parameters are subject to the restriction (3.20) of the quasiconvexity, and restrictions (2.10) in each of the phases. In terms of the matrix T_{κ} the inequalities (2.10) can be expressed as

$$\begin{bmatrix} \frac{(\kappa'_i)^2 + (\kappa''_i)^2}{\kappa''_i} & -\frac{\kappa'_i}{\kappa''_i} \\ -\frac{\kappa'_i}{\kappa''_i} & \frac{1}{\kappa''_i} \end{bmatrix} + \begin{pmatrix} \frac{d-1}{d} t_1 & t_3 \\ t_3 & dt_2 \end{pmatrix} \geq 0, \quad i = 1, 2, \quad (3.52)$$

and

$$\begin{bmatrix} \frac{2(d-1)}{d} \frac{(\mu''_i)^2 + (\mu'_i)^2}{\mu''_i} & -\frac{\mu'_i}{\mu''_i} \\ -\frac{\mu'_i}{\mu''_i} & \frac{1}{\mu''_i} \end{bmatrix} - \begin{pmatrix} \frac{d-1}{d} t_1 & -t_3 \\ -t_3 & dt_2 \end{pmatrix} \geq 0, \quad i = 1, 2. \quad (3.53)$$

Now we are ready to interpret the bounds and the restrictions in geometrical terms as was done by Gibiansky and Milton (1993a).

3.3. Geometrical interpretation of the bulk modulus bounds

It is useful to summarize the findings of the previous section. The bound on the Z -transformation of the effective complex bulk modulus is given by the matrix inequality (3.47) which is valid for any values of the parameters t_1 , t_2 and t_3 subject to the restrictions of the quasiconvexity (3.20) and additional matrix inequalities (3.52) and (3.53). It is convenient to introduce the new notation

$$g_1 = \frac{d-1}{d} t_1, \quad g_2 = dt_2, \quad g_3 = t_3. \quad (3.54)$$

In terms of these new parameters, the bounds are given by the matrix inequality

$$\begin{bmatrix} \frac{(z'_\kappa)^2 + (z''_\kappa)^2}{z''_\kappa} + g_1 & \frac{z'_\kappa}{z''_\kappa} - g_3 \\ \frac{z'_\kappa}{z''_\kappa} - g_3 & \frac{1}{z''_\kappa} + g_2 \end{bmatrix} \geq 0 \quad (3.55)$$

which is valid for any values of the parameters g_1 , g_2 and g_3 that satisfy the matrix inequalities

$$\begin{bmatrix} \frac{(\kappa'_i)^2 + (\kappa''_i)^2}{\kappa''_i} + g_1 & -\frac{\kappa'_i}{\kappa''_i} + g_3 \\ -\frac{\kappa'_i}{\kappa''_i} + g_3 & \frac{1}{\kappa''_i} + g_2 \end{bmatrix} \geq 0, \quad i = 1, 2, \quad (3.56)$$

and

$$\begin{bmatrix} \frac{(y'_i)^2 + (y''_i)^2}{y''_i} - g_1 & -\frac{y'_i}{y''_i} + g_3 \\ -\frac{y'_i}{y''_i} + g_3 & \frac{1}{y''_i} - g_2 \end{bmatrix} \geq 0, \quad i = 2, 2, \quad (3.57)$$

and the quasiconvexity conditions

$$\{g_1, g_2, g_3\} : \begin{cases} g_1 \geq 0, & g_2 \geq 0, & \forall g_3, & \text{if } d = 3 \\ g_1 \geq 0, & \forall g_2, & \forall g_3, & \text{if } d = 2 \end{cases} \quad (3.58)$$

Note that the inequalities (3.55)–(3.57) are very similar, thus allowing for a simple geometrical interpretation of the bounds.

The determinant of the positive definite matrix (3.55) must be positive, i.e.

$$(g_2/z'')[(z' - z'_c)^2 + (z'' - z'_c)^2 - R^2] \geq 0, \quad (3.59)$$

where

$$z'_c = -\frac{g_3}{g_2}, \quad z''_c = -\frac{1 + g_1 g_2 - g_3^2}{2g_2}, \quad R = \left| \frac{1 - g_1 g_2 + g_3^2}{2g_2} \right|. \quad (3.60)$$

Therefore, for any composite, the Z -transformation of the complex bulk modulus κ_* lies outside of the circle in the complex Z -plane with the center $z_c = z'_c + iz''_c$ and the radius R , if $g_2 \geq 0$, and inside this circle if $g_2 \leq 0$. By changing the parameters g_1 , g_2 , and g_3 we can move and resize this circle. Note that we always use nonstrict inequalities in our bounds, so that when we say that the point lies outside (inside) of

the circle, this means that it either lies strictly outside (inside) or on the boundary of this circle.

Analyzing the restrictions (3.56)–(3.58) for the positive values of the parameters g_1 and g_2 (which is required if $d \geq 3$), one can show that the Z -transformation z_κ of the effective complex bulk modulus κ_* lies *outside* any circle that:

(i) does not contain the origin O of the complex Z -plane, due to the inequality;

$$(z'_c)^2 + (z''_c)^2 - R^2 = g_1/g_2 \geq 0, \quad \text{if } g_1 \geq 0, \quad g_2 \geq 0. \quad (3.61)$$

(ii) does not contain the points κ_1 and κ_2 , due to the inequalities (3.56) if $g_1 \geq 0$, and $g_2 \geq 0$;

(iii) contains the points $-y_1 = -2(d-1)/d \mu_1$ and $-y_2 = -2(d-1)/d \mu_2$ due to the inequalities (3.57) if $g_1 \geq 0$, and $g_2 \geq 0$.

Observe that the circles may degenerate into the straight lines that separate the complex Z -plane into two parts. One of these parts must contain the points $-y_1$ and $-y_2$. The other part must contain the points κ_1 , κ_2 and O . Any straight line can be considered as a circle of infinite radius that passes through the infinity point $z_\infty = \infty$. Therefore, one can show that any circle (or line) that corresponds to the requirements (i)–(iii) corresponds to some values of the parameters $g_1 \geq 0$, $g_2 \geq 0$, and g_3 [see Gibiansky and Milton (1993a)]. By changing the values of the parameters g_1 , g_2 , and g_3 one can move and resize these circles.

Thus, in three-dimensions there are six important points that define the bounds: the complex bulk moduli of the phases κ_1 , κ_2 , the points $-y_1$, $-y_2$, the origin O of the complex Z -plane, and the infinity point z_∞ of this plane. The bounds are defined by the intersection of the exteriors of the circles that do not contain the points O , κ_1 , κ_2 and z_∞ but contain the points $-y_1$, $-y_2$. We emphasize that any of these six characteristic points may lie on the boundary of the circle. Now one can reformulate the bounds as follows:

Statement 1a: The Z -transformation of the effective bulk modulus of a three-dimensional composite lies outside any circle that contains the points $-y_1$, $-y_2$ and does not contain the points κ_1 , κ_2 , O , and z_∞ .

In two dimensions, however, the parameter g_2 need not be positive; it can have any negative value as well. For negative values of this parameter, the bound (3.59) and the restrictions (3.56), and (3.57), show that the Z -transformation of the effective bulk modulus of the planar composite lies *inside* any circle in the complex Z -plane that:

(i) contains the origin of the complex Z -plane, due to the inequality

$$(z'_c)^2 + (z''_c)^2 - R^2 = g_1/g_2 \leq 0, \quad \text{if } g_1 \geq 0, \quad g_2 \leq 0; \quad (3.62)$$

(ii) contains the points κ_1 and κ_2 due to the inequalities (3.56) if $g_1 \geq 0$, and $g_2 \leq 0$;

(iii) does not contain the points $-y_1 = -2(d-1)/d \mu_1$ and $-y_2 = -2(d-1)/d \mu_2$, due to the inequalities (3.57) if $g_1 \geq 0$, and $g_2 \leq 0$.

Thus we see that in two dimensions the infinity point of the complex Z -plane does not play any role. By changing the parameters g_1 , g_2 , and g_3 , we move and resize the corresponding circle in the Z -plane, and for any composite the value $z_\kappa(\kappa_*)$ lies on

the same side of the circle (inside or outside) as the points O , κ_1 and κ_2 , and on different sides of the point $-y_1$ and $-y_2$. One should not consider the circles that "separate" the points $-y_1$ and $-y_2$ (i.e. place one of them inside the circle and the other of them outside) or separate the points O , κ_1 and κ_2 . In light of the above, one can formulate the following result:

Statement 1b: Consider any circle that separates the complex Z -plane into two parts (interior and exterior) such that one of them contains the points $-y_1$ and $-y_2$, and the other contains the points O , κ_1 and κ_2 . Then, the Z -transformation $z_\kappa(\kappa_*)$ of the effective bulk modulus of a two-dimensional composite belongs to the part that contains the points O , κ_1 and κ_2 .

4. EXPLICIT FORMS OF THE COMPLEX BULK MODULUS BOUNDS

In this section, we find explicit forms of the bounds obtained in the previous section. There are three different ways to express the bounds corresponding to the three different planes: the Z_κ -plane, Y_κ -plane, or κ_* plane. Each representation has its own advantages. Bounds in terms of the complex bulk modulus itself allow one to see the limits of changing the effective bulk modulus for fixed volume fractions and ζ -parameters. Bounds in terms of the Y -transformations do not depend explicitly on the phase volume fractions. This dependence is hidden in the definition of the Y -transformation. Thus in such a form one can compare properties of composites with different phase volume fractions but fixed ζ -parameters. Finally, the Z -transformation eliminates the dependence of the bounds on the ζ -parameters. In terms of Z -transformations, the bounds depend only on the phase moduli but not on the phase volume fractions or other geometrical parameters. This allows one to compare the properties of composites with different volume fractions and ζ -parameters.

First, we construct the bounds in the complex Z_κ -plane, and then map them into the complex Y_κ -plane, and finally into the complex κ_* -plane. The bounds are given by arcs of circles. Let $\text{Arc}(z_1, z_2, z_3)$ denote an arc in the complex Z -plane that passes through the points z_1 and z_2 and while extended to circle passes through the point z_3 . It is given by the points

$$z = \gamma z_1 + (1-\gamma)z_2 - \frac{\gamma(1-\gamma)(z_1 - z_2)^2}{\gamma z_2 + (1-\gamma)z_1 - z_3}, \quad \gamma \in [0, 1]. \quad (4.1)$$

Let also $\text{Arc}(z_1, z_\infty, z_3)$ denote the half-line $[z_1, z_\infty)$ that when extended to line passes through the point z_3 .

In Figs 1–3 that illustrate the results of this section we assume that the phase bulk moduli κ_1, κ_2 , shear moduli μ_1, μ_2 volume fraction $f_1, f_2 = 1 - f_1$, and the three-point parameters $\zeta_1, \zeta_2 = 1 - \zeta_1$ are equal to

$$\begin{aligned} \kappa_1 &= 10 + i2, & \mu_1 &= 6 + i1, & f_1 &= 0.5, & \zeta_1 &= 0.5, \\ \kappa_2 &= 1 + i8, & \mu_2 &= 1 + i4, & f_2 &= 0.5, & \zeta_2 &= 0.5, \end{aligned} \quad (4.2)$$

for a three-dimensional problem or equal to

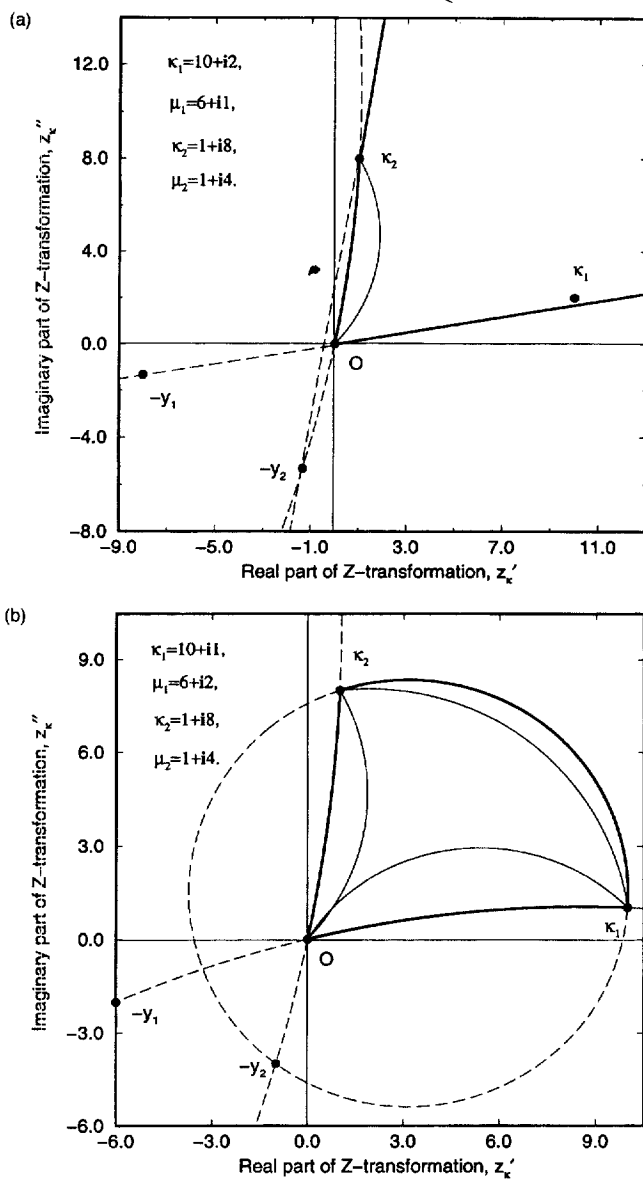


Fig. 1. Bounds on the Z-transformation of the complex bulk modulus of an isotropic viscoelastic three-dimensional (a) and planar (b) composite. The bold curves describe our bounds. The thin arcs illustrate additional arcs described by Statements 1a and 1b.

$$\begin{aligned} \kappa_1 &= 10 + i1, & \mu_1 &= 6 + i2, & f_1 &= 0.5, & \zeta_1 &= 0.5, \\ \kappa_2 &= 1 + i8, & \mu_2 &= 1 + i4, & f_2 &= 0.5, & \zeta_2 &= 0.5, \end{aligned} \quad (4.3)$$

for a planar composite. Such values of the moduli are chosen for the illustration only and do not represent any particular materials.

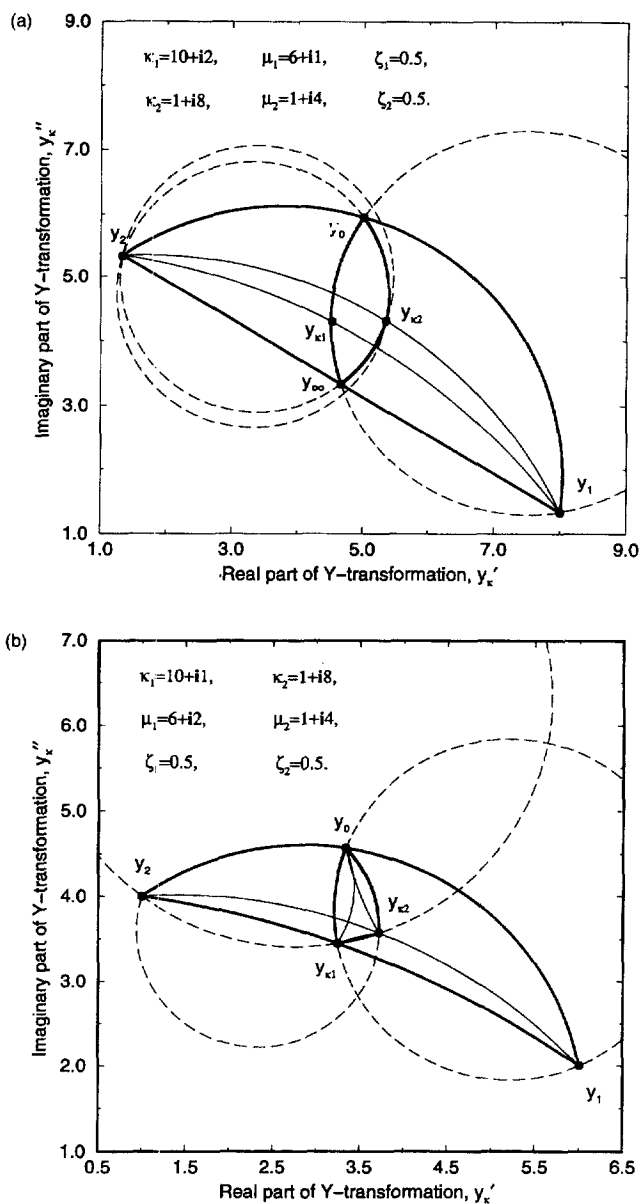


Fig. 2. Bounds on the Y-transformation of the complex bulk modulus of an isotropic viscoelastic three-dimensional (a) and planar (b) composite. The smallest bold curvilinear sets describe the new bounds of Statements 2a and 2b. The thin arcs illustrate additional arcs described by Statements 2a and 2b. The larger lens-shaped region represents the Gibiansky–Milton (1993a) bounds that do not depend on the ζ -parameters. The marked points are defined in Table 1.

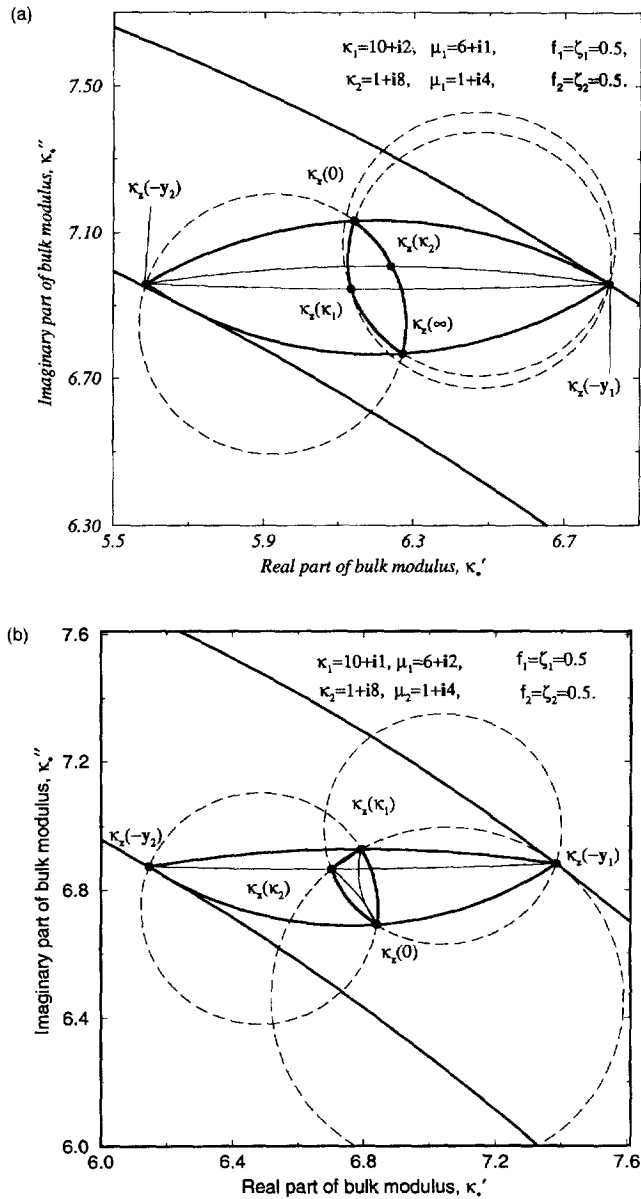


Fig. 3. Bounds on the complex bulk modulus of an isotropic viscoelastic three-dimensional (a) and planar (b) composite. The smallest curvilinear triangular sets are the new bounds given by the Statements 5a and 5b. The thin arcs illustrate additional arcs described by the Statement 5b. The dashed circles illustrate the procedure. The intermediate-size set represents the Gibiansky–Milton (1993a) fixed-volume-fraction bounds. The largest arcs show the arbitrary-volume-fraction bounds of Gibiansky and Lakes (1993, 1997).

The marked points are defined in Table 1.

4.1. Bounds in the Z -plane

4.1.1. *Three-dimensional composites.* Statement 1a implicitly describes the complex bulk modulus bounds for a three-dimensional composite in the Z -plane. It is an easy task to construct explicit bounds for any given phase properties. For example, such bounds are presented on Fig. 1(a) for the values of the parameters given by (4.2).

There are six important points that define the bounds: the complex bulk moduli of the phases κ_1, κ_2 , the points $-y_1, -y_2$, the origin O of the complex Z_κ -plane, and the infinity point z_∞ of this plane [see Fig. 1(a)]. The bounds are described by the line $\text{Arc}(\kappa_2, z_\infty, -y_2)$, by the line $\text{Arc}(O, z_\infty, -y_1)$, and (in the interval $[O, \kappa_2]$) by the outermost of the arcs $\text{Arc}(O, \kappa_2, -y_1)$ and $\text{Arc}(O, \kappa_2, -y_2)$. For the given values (4.2) of the parameters, it is an $\text{Arc}(O, \kappa_2, -y_2)$. The Appendix describes the procedure leading to Fig. 1(a) in more detail.

Let us consider some other specific values of the phase moduli. The simplest example is when the phases have real Poisson's ratios. Then the ratios κ_1/μ_1 and κ_2/μ_2 are real, the points $-y_1 = -2(d-1)\mu_1/d$, O , and κ_1 lie on the same line, the points $-y_2 = 2(d-1)\mu_2/d$, O , and κ_2 lie on the other line, and the bounds are given by the cone with the vertex O and the sides containing the points κ_1 and κ_2 .

If the phases have real moduli, then the bounds degenerate to the half-line $[O, \infty)$ and coincide with previously known bounds [Beran and Molyneux (1966), McCoy (1970), Milton (1981a, 1982)]

$$\kappa_- \leq \kappa_* \leq \kappa_+, \quad (4.4)$$

where

$$\kappa_\pm = f_1\kappa_1 + f_2\kappa_2 - \frac{f_1f_2(\kappa_1 - \kappa_2)^2}{f_2\kappa_1 + f_1\kappa_2 + y_\pm}, \quad (4.5)$$

and

$$y_- = \frac{2(d-1)}{d} \left(\frac{\zeta_1}{\mu_1} + \frac{\zeta_2}{\mu_2} \right)^{-1}, \quad y_+ = \frac{2(d-1)}{d} (\zeta_1\mu_1 + \zeta_2\mu_2). \quad (4.6)$$

Indeed, as was shown by Gibiansky and Torquato (1995b), such bounds can be written in the form

$$y_- \leq y_k \leq y_+ \quad (4.7)$$

where y_k is the Y -transformation of the effective bulk modulus, or in the form

$$0 \leq z_k \leq \infty, \quad (4.8)$$

where z_k is the Z -transformation of the effective bulk modulus.

4.1.2. *Planar composites.* Statement 1b implicitly describes the bounds on the bulk modulus for planar viscoelastic composites. One can show that it is equivalent to the following description:

Statement 2: The Z -transformation of the effective bulk modulus of a viscoelastic

two-phase composite lies in the curvilinear triangle (O, κ_1, κ_2) given by the outermost three of the six arcs

$$\begin{aligned} &\text{Arc}(O, \kappa_1, -Y_1), \quad \text{Arc}(O, \kappa_1, -Y_2), \\ &\text{Arc}(O, \kappa_2, -Y_1), \quad \text{Arc}(O, \kappa_2, -Y_2), \\ &\text{Arc}(\kappa_1, \kappa_2, -Y_1), \quad \text{Arc}(\kappa_1, \kappa_2, -Y_2), \end{aligned}$$

in the complex Z -plane.

This result is illustrated in Fig. 1(b) for the parameters given by (4.3). The arcs described by Statement 2 are given in the complex z_κ -plane. The outermost arcs $\text{Arc}(O, \kappa_1, -Y_1)$, $\text{Arc}(O, \kappa_2, -Y_1)$, and $\text{Arc}(\kappa_1, \kappa_2, -Y_2)$ are marked by bold curves.

In the particular case of a composite containing phases with real Poisson's ratios, the sides (O, κ_1) and (O, κ_2) of the curvilinear triangle (O, κ_1, κ_2) degenerate into a straight line connecting the vertices of the triangle.

For the elastic composite with pure real bulk and shear moduli the triangular (O, κ_1, κ_2) degenerates into the interval $[0, \kappa_{\max}]$ of the real axes. Here $\kappa_{\max} = \max\{\kappa_1, \kappa_2\}$ is maximal of the two phase bulk moduli (that are real for this example). Then the bulk modulus bounds can be presented in the form (4.4)–(4.5) where the lower bound y_- is given by (4.6) but the upper bound is equal to

$$y_+ = \zeta_1 y_1 + \zeta_2 y_2 - \frac{\zeta_1 \zeta_2 (y_1 - y_2)}{\zeta_1 y_2 + \zeta_2 y_1 + \kappa_{\max}}, \quad (4.9)$$

or, equivalently

$$0 \leq z_\kappa \leq \kappa_{\max} \quad (4.10)$$

in agreement with the results by Silnutzer (1972) and Gibiansky and Torquato (1995b).

4.2. Bounds in the Y -plane

One can easily map the bounds from the Z_κ -plane into the Y_κ -plane. To do it one just needs to find the Y_κ -plane image of the boundary circles in the Z_κ -plane. The Z -transformation is a fractional-linear one. Under such transformation, any circle transforms into the circle (one may consider a line as a circle that passes through the infinity point). A circle in the complex plane is defined by three points that it passes through. Thus, to find the image of the circle it is sufficient to find the images of the three points on this circle. We will be interested in the images of the six points $O = 0$, $z_\infty = \infty$, $z_{\kappa_1} = \kappa_1$, $z_{\kappa_2} = \kappa_2$, $z_{y_1} = -2(d-1)\mu_1/d$, and $z_{y_2} = -2(d-1)\mu_2/d$. The following Table 1 describes the images of these characteristic points in the Y_κ -plane (and in the κ_* -plane that we will discuss in the next section).

The bounds in the complex Y_κ -plane are given by the Y -images of the extremal curves in the Z_κ -plane (arcs and lines). One can formulate these bounds as follows:

Statement 3a: The Y -transformation y_κ of the effective complex bulk modulus κ_* of a d -dimensional ($d \geq 3$) isotropic viscoelastic two-phase composite lies in the curvi-

Table 1. Transformation of the characteristic points from Z_κ -plane into Y_κ -plane and κ_* -plane. Here $y_1 = 2(d-1)\mu_1/d$ and $y_2 = 2(d-1)\mu_2/d$

$z_\kappa(y_\kappa, y_1, y_2)$	$y_\kappa(\kappa_*, \kappa_1, \kappa_2)$	κ_*
$0 = 0,$	$y_0 = \left[\frac{\zeta_1}{y_1} + \frac{\zeta_2}{y_2} \right]^{-1}$	$\kappa_z(0) = \left[\frac{f_1}{\kappa_1 + y_0} + \frac{f_2}{\kappa_2 + y_0} \right]^{-1} - y_0$
$z_\infty = \infty$	$y_\infty = \zeta_1 y_1 + \zeta_2 y_2$	$\kappa_z(\infty) = \left[\frac{f_1}{\kappa_1 + y_\infty} + \frac{f_2}{\kappa_2 + y_\infty} \right]^{-1} - y_\infty$
$z_{\kappa_1} = \kappa_1$	$y_{\kappa_1} = \left[\frac{\zeta_1}{y_1 + \kappa_1} + \frac{\zeta_2}{y_2 + \kappa_1} \right]^{-1} - \kappa_1$	$\kappa_z(\kappa_1) = \left[\frac{f_1}{\kappa_1 + y_{\kappa_1}} + \frac{f_2}{\kappa_2 + y_{\kappa_1}} \right]^{-1} - y_{\kappa_1}$
$z_{\kappa_2} = \kappa_2$	$y_{\kappa_2} = \left[\frac{\zeta_1}{y_1 + \kappa_2} + \frac{\zeta_2}{y_2 + \kappa_2} \right]^{-1} - \kappa_2$	$\kappa_z(\kappa_2) = \left[\frac{f_1}{\kappa_1 + y_{\kappa_2}} + \frac{f_2}{\kappa_2 + y_{\kappa_2}} \right]^{-1} - y_{\kappa_2}$
$z_{y_1} = -\frac{2(d-1)}{d}\mu_1$	$y_1 = \frac{2(d-1)}{d}\mu_1$	$\kappa_z(-y_1) = \left[\frac{f_1}{\kappa_1 + y_1} + \frac{f_2}{\kappa_2 + y_1} \right]^{-1} - y_1$
$z_{y_2} = -\frac{2(d-1)}{d}\mu_2$	$y_2 = \frac{2(d-1)}{d}\mu_2$	$\kappa_z(-y_2) = \left[\frac{f_1}{\kappa_1 + y_2} + \frac{f_2}{\kappa_2 + y_2} \right]^{-1} - y_2$

linear set $(y_{\kappa_1}, y_0, y_{\kappa_2}, y_\infty)$ given in the complex y_κ -plane by the outermost of the 12 arcs

$$\begin{aligned}
 &\text{Arc}(y_0, y_{\kappa_1}, y_1), \quad \text{Arc}(y_0, y_{\kappa_1}, y_2), \\
 &\text{Arc}(y_0, y_{\kappa_2}, y_1), \quad \text{Arc}(y_0, y_{\kappa_2}, y_2), \\
 &\text{Arc}(y_{\kappa_1}, y_{\kappa_2}, y_1), \quad \text{Arc}(y_{\kappa_1}, y_{\kappa_2}, y_2), \\
 &\text{Arc}(y_\infty, y_{\kappa_1}, y_1), \quad \text{Arc}(y_\infty, y_{\kappa_1}, y_2), \\
 &\text{Arc}(y_\infty, y_{\kappa_2}, y_1), \quad \text{Arc}(y_\infty, y_{\kappa_2}, y_2), \\
 &\text{Arc}(y_0, y_\infty, y_1), \quad \text{Arc}(y_0, y_\infty, y_2).
 \end{aligned}$$

Statement 3b: The Y -transformation y_κ of the effective complex bulk modulus κ_* of a planar ($d = 2$) isotropic viscoelastic two-phase composite lies in the curvilinear triangle $(y_{\kappa_1}, y_0, y_{\kappa_2})$ given in the complex y_κ -plane by the outermost three of the six arcs

$$\begin{aligned}
 &\text{Arc}(y_0, y_{\kappa_1}, y_1), \quad \text{Arc}(y_0, y_{\kappa_1}, y_2), \\
 &\text{Arc}(y_0, y_{\kappa_2}, y_1), \quad \text{Arc}(y_0, y_{\kappa_2}, y_2), \\
 &\text{Arc}(y_{\kappa_1}, y_{\kappa_2}, y_1), \quad \text{Arc}(y_{\kappa_1}, y_{\kappa_2}, y_2).
 \end{aligned}$$

The points used in the Statements 3a and 3b are defined in Table 1.

Figures 2(a) and 2(b) illustrate the bounds on the effective bulk modulus of three-dimensional and planar composites, respectively, in the Y_κ -plane. The values of the phase moduli and ζ -parameters are given by (4.2) or (4.3). The larger circular arcs correspond to the bounds obtained by Gibiansky and Milton (1993a) without specifying the values of the geometrical parameters ζ_1 and ζ_2 . The smallest regions are our new bounds. The characteristic points that were used in the construction of the bounds are marked on each of these figures. In such a form (in the Y_κ -plane) the bounds do not depend on the volume fractions. This dependence is "hidden" in the definition of the Y -transformation.

For some values of the parameters, the curvilinear region $(y_{\kappa 1}, y_0, y_{\kappa 2}, y_\infty)$ described by the Statement 3a may degenerate into the curvilinear triangle or even lens-shaped region. For example, in Fig. 3(a) we see that the point $y_{\kappa 2}$ lies within the curvilinear triangle $(y_0, y_{\kappa 1}, y_\infty)$ outlined by the arcs $\text{Arc}(y_0, y_\infty, y_2)$, $\text{Arc}(y_0, y_{\kappa 1}, y_1)$, and $\text{Arc}(y_\infty, y_{\kappa 1}, y_1)$, and thus do not influence the bounds. Similarly, the curvilinear triangle described by the Statement 3b may degenerate into the lens-shaped region, if one of the three points $y_{\kappa 1}$, y_0 , and $y_{\kappa 2}$ lies inside of the arcs of the circles connecting the other two points and the points y_1, y_2 . Note that the points y_1 and y_2 correspond to the Hashin-Shtrikman (1963) assemblages of coated spheres (circles in two dimensions).

4.3. *Bounds in the complex bulk modulus plane*

The next step is to further transform the bounds into the κ_* -plane. It is done in precisely the same way as in the previous section. The necessary images of the characteristic points are given in Table 1. Let $\kappa_z(z)$ be a complex function of complex variable z

$$\kappa_z(z) = f_1 \kappa_1 + f_2 \kappa_2 - \frac{f_1 f_2 (\kappa_1 - \kappa_2)^2}{f_2 \kappa_1 + f_1 \kappa_2 + y_\kappa(z)}, \quad (4.11)$$

where

$$y_\kappa(z) = \zeta_1 y_1 + \zeta_2 y_2 - \frac{\zeta_1 \zeta_2 (y_1 - y_2)^2}{\zeta_2 y_1 + \zeta_1 y_2 + z}. \quad (4.12)$$

Then the following Statements describe the bounds on the bulk modulus of an isotropic viscoelastic composite consisting of two isotropic phases with given values of the phase volume fractions $f_1 = 1 - f_2$ and three-point geometrical parameters $\zeta_1 = 1 - \zeta_2$. They follow directly from the corresponding results in the Y -plane.

Statement 4a: The effective complex bulk modulus κ_* of a d -dimensional ($d \geq 3$) isotropic viscoelastic two-phase composite lies within any circle in the complex bulk moduli plane that contains the points $\kappa_z(0)$, $\kappa_z(\infty)$, $\kappa_z(\kappa_1)$, and $\kappa_z(\kappa_2)$, but does not contain the points $\kappa_z(-y_1)$ and $\kappa_z(-y_2)$.

Statement 4b: The effective complex bulk modulus κ_* of a planar ($d = 2$) isotropic viscoelastic two phase composite lies within any circle in the complex bulk moduli

plane that contains the points $\kappa_z(0)$, $\kappa_z(\kappa_1)$, and $\kappa_z(\kappa_2)$, but does not contain the points $\kappa_z(-y_1)$ and $\kappa_z(-y_2)$.

Therefore, the bounds are given by the intersection of all such circles described by the Statements 4a and 4b. Statements 5a and 5b give constructive way to find these intersections:

Statement 5a: The effective complex bulk modulus κ_* of a d -dimensional ($d \geq 3$) isotropic viscoelastic two-phase composite lies in the curvilinear set $(\kappa_z(\kappa_1), \kappa_z(0), \kappa_z(\kappa_2), \kappa_z(\infty))$ given by the outermost of the 12 arcs

$$\begin{aligned} & \text{Arc}(\kappa_z(0), \kappa_z(\kappa_1), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(0), \kappa_z(\kappa_1), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(0), \kappa_z(\kappa_2), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(0), \kappa_z(\kappa_2), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(\kappa_1), \kappa_z(\kappa_2), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(\kappa_1), \kappa_z(\kappa_2), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(\infty), \kappa_z(\kappa_1), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(\infty), \kappa_z(\kappa_1), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(\infty), \kappa_z(\kappa_2), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(\infty), \kappa_z(\kappa_2), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(0), \kappa_z(\infty), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(0), \kappa_z(\infty), \kappa_z(-y_2)), \end{aligned}$$

in the complex bulk modulus plane.

Statement 5b: The effective complex bulk modulus κ_* of a planar ($d = 2$) isotropic viscoelastic two-phase composite lies in the curvilinear triangular $(\kappa_z(\kappa_1), \kappa_z(0), \kappa_z(\kappa_2))$ given by the outermost of the six arcs

$$\begin{aligned} & \text{Arc}(\kappa_z(0), \kappa_z(\kappa_1), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(0), \kappa_z(\kappa_1), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(0), \kappa_z(\kappa_2), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(0), \kappa_z(\kappa_2), \kappa_z(-y_2)), \\ & \text{Arc}(\kappa_z(\kappa_1), \kappa_z(\kappa_2), \kappa_z(-y_1)), \quad \text{Arc}(\kappa_z(\kappa_1), \kappa_z(\kappa_2), \kappa_z(-y_2)), \end{aligned}$$

in the complex bulk modulus plane.

Bounds on the effective complex bulk modulus of the viscoelastic composite (Statement 5a and 5b) are depicted in Figs 3(a) and 3(b). In these figures, the smallest regions correspond to the bounds of Statements 5a and 5b, the small thin arcs in the Fig. 3(b) illustrate the other arcs mentioned in the Statement 5b. For the three-dimensional problem, Fig. 3(a), we marked only the arcs that actually give the bounds.

The intermediate size lens-shaped regions correspond to the bounds on the complex bulk modulus of a composite when only the volume fraction is given. These bounds were found by Gibiansky and Milton (1993a). The largest curves are the bounds by Gibiansky and Lakes (1993, 1997) that do not contain any geometrical information; they can be used when even the phase volume fractions are unknown. Considered together with our new bounds, they form nested sets of the complex bulk modulus bounds. The two largest bold arcs represent the bounds that do not contain any information about the composite, except that of the phase moduli. If one knows the phase volume fractions, then one can use the bounds of Gibiansky and Milton (1993a). If one can measure not only the phase volume fractions but also three-point

correlation functions and calculate three-point ζ -parameters, then one can use our new geometrical-parameter bounds.

By using the new bounds, one can generate fixed-volume-fraction bounds as the union of all curvilinear sets corresponding to the new bounds for $\zeta_1 = 1 - \zeta_2 \in [0, 1]$. It can be done by using the procedure suggested by Gibiansky and Lakes (1993) who generated volume-fraction-independent bounds by using the fixed-volume-fraction bounds.

Observe that there is a similarity between this picture and the results by Milton (1981b,c, 1982) [see also Bergman (1982)], who derived the nested sequence of complex conductivity bounds. They used the analytical method, entirely different from our procedure. If applied to the complex conductivity problem, our method would produce the first three levels of the nested sequence of the best bounds on the complex conductivity of an isotropic planar composite obtained by Milton (1981a,b) and would improve upon Milton (1981a,b) bounds in three dimensions.

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APPENDIX: Z_κ -PLANE BOUNDS IN THREE DIMENSIONS

Here we describe the procedure to construct the bounds on the complex bulk modulus of a three-dimensional composite. First, we consider the complex Z_κ -plane and mark the points O , κ_1 , κ_2 , $-y_1$, and $-y_2$. The imaginary parts of the complex stiffness moduli are positive, because dissipation is positive. Therefore, the points κ_1 , κ_2 lie in the upper half-plane of the Z_κ -plane, and the points $-y_1 = -2(d-1)\mu_1/d$, $-y_2 = -2(d-1)\mu_2/d$ lie in the lower half-plane. Thus, one can imagine a line that separates the complex Z -plane into two half-planes; one of them contains the points O , κ_1 , and κ_2 and the other one contains the points $-y_1$ and $-y_2$.

Now one needs to rotate this imaginary line clockwise until it passes through one of the points O , κ_1 , or κ_2 and one of the points $-y_1$ and $-y_2$ like line $(-y_2, \kappa_2)$ in Fig. 1(a). According to Statement 1a, only the points in the half-plane containing the points O , κ_1 , or κ_2 may correspond to some composite. Thus, the boundary $(-y_2, \kappa_2)$ of this half-plane may provide part of the bounds. This is true in the case depicted in Fig. 1(a). Then one has to repeat the procedure rotating the initial line anti-clockwise. The extremal line $(-y_1, O)$ in Fig. 1(a) has been obtained by this process. Observe that the lines $(-y_2, \kappa_2)$ and $(-y_1, O)$ illustrates two possibilities: one [illustrated by the line $(-y_2, \kappa_2)$ in Fig. 1(a)] is when the extremal line passes through one of the points κ_1 or κ_2 on one side, and one of the points $-y_1$ or $-y_2$ on the other

side. Then the bound is given by the part $\text{Arc}(\kappa_2, z_x, -y_2)$ of this line, it is marked bold in Fig. 1(a). The other possible case is when the extremal line passes through one of the points $-y_1$ or $-y_2$ and the origin of the Z -plane O , as it is for the line $(-y_1, O)$. Then the bound is given by the segment $\text{Arc}(O, \infty, y_1)$ of this line marked bold in Fig. 1(a).

The next step is to check whether one can further transform the aforementioned lines into the circles that still contain the points $-y_1$ and $-y_2$ but does not contain O , κ_1 , and κ_2 . It is obviously possible if the extremal line does not pass through the origin O . If the extremal line does not pass through the origin O , e.g. as the line $(-y_2, \kappa_2)$ in Fig. 1(a), then the bound is given by the outermost of the two arcs $\text{Arc}(O, \kappa_2, -y_1)$ and $\text{Arc}(O, \kappa_2, -y_2)$.