A complete characterization of the possible bulk and shear moduli of planar polycrystals.

M. Avellaneda*, A.V. Cherkaev#, L.V. Gibiansky†, G.W. Milton*#, M. Rudelson‡

* Courant Institute of Mathematical Sciences, 251 Mercer Street, New York, NY 10012, USA;

University of Utah, Department of Mathematics, Salt Lake City, Utah, 84112, USA;

† Princeton University, Department of Civil Engineering and Operations Research, Princeton, NJ 08544, USA.

‡ Hebrew University of Jerusalem, Institute of Mathematics and Computer Science, Givat Ram, 91904, Jerusalem, Israel.

Abstract

This paper investigates the range of possible elastic moduli of two-dimensional isotropic polycrystals. The polycrystals are comprised of grains obtained from a single orthotropic material. The overall elastic properties are described by an effective bulk modulus K_0 and an effective shear modulus μ_0. The pair (K_0, μ_0) is shown to be confined to a rectangle in the (K, μ) plane. Microstructures are identified which correspond to every point within the rectangle, and in particular to the corner points. Optimal bounds on the effective Poisson's ratio and Young's modulus follow immediately. Under a certain constraint on the crystal moduli, the rectangle degenerates to a line segment: the effective shear modulus of such a polycrystal is microstructure independent. This extends earlier work of Hill and Lurie and Cherkaev who established microstructure independence for polycrystals constructed from square symmetric crystals.

1 Introduction

Polycrystals are ubiquitous in nature and amongst engineered materials. These solids are composed of small crystallites forming an aggregate that appears homogeneous at the macroscopic scale. Some of the more common polycrystalline solids include commercial metals and alloys as well as ceramics. Two-dimensional polycrystals can now be manufactured using thin-film technology. Many of the mechanical properties of polycrystals, such as stiffness, plasticity and strength, can be attributed to the micromechanical interactions between crystallites. In other words, the geometric configuration of the crystallites plays a crucial role in determining macroscopic behavior.

Investigating the relation between the overall, or effective, elastic moduli of polycrystals and the associated single-crystal constants is a central theme in the theory of composite materials. This is a complex problem which requires solving the elasticity equations in an intricate geometry. Furthermore, the exact microstructure is usually not available because a polycrystal is highly disordered. For this reason, the problem has traditionally been handled by statistical methods and various approximations. The effective medium approximation (Hershey, 1954; Kneer, 1965) is based on a self-consistent calculation for a single spherical grain. Another approach, due to Hashin and Shtrikman (1962) and later perfected by Walpole (1966) uses the statistical assumption of "equiaxiality", which, roughly speaking, means that the orientations of different grains are uncorrelated. This leads to the celebrated Hashin-Shtrikman (HS) bounds (Hashin & Shtrikman (1962)) . Both the effective medium theory and the HS bounds are discussed in the review articles of Watt, Davis and O'Connell (1976) and Willis (1982). They are good predictors of the physical properties of a polycrystal provided the anisotropy of the single crystal is not too pronounced.

There are however some drawbacks to these approaches. The effective medium approximation only accounts for interactions between grains in an average way. The Hashin-Shtrikman approach is based on the equiaxiality assumption which is not universally valid (Beran, Mason, Adams, & Olsen 1995). In particular, one would expect the Hashin-Shtrikman bounds to be violated in some cases, i.e. the HS bounds to be "too narrow". This is indeed the case, as shown by Schulgasser (1977) for the conductivity of three-dimensional polycrystals and by Avellaneda & Milton (1989) and Rudelson (1989) for the bulk modulus of two-dimensional elastic polycrystals. Curiously enough, the HS bounds can be also be "too wide". In fact, for the case of polycrystals with extreme single-crystal anisotropy, Schulgasser (1977) and Helsing and Helte (1991) have shown that the HS bounds for conductivity are actually less restrictive than other bounds obtained without assuming equiaxiality. A similar observation applies to the elastic moduli of 2D polycrystals when the single crystal has square symmetry: the HS theory allows for an interval of possible shear moduli even though the effective shear modulus is determined exactly (Lurie and Cherkaev, 1984a).

In this paper, we investigate the elastic behavior of macroscopically isotropic polycrystals without making ancillary statistical assumptions on the grain orientations. Specifically, we ask following question: *What are the possible values of the elastic moduli of a macroscopically isotropic polycrystal formed from a given single-crystal ?* By solving this problem, we provide rigorous, geometry-independent bounds which determine the entire range of *realizable* elastic moduli. At present, our theory is limited to two-dimensional polycrystals, although in principle it could be extended to three-dimensional systems. For simplicity, we will treat the case where the single crystal has orthotropic symmetry, deferring the treatment of general symmetry classes to a future publication (Avellaneda and Milton, 1995).

The current paper is a non-trivial extension of previous work of Avellaneda & Milton (1989) and Rudelson (1989) who obtained the best possible bounds on the effective bulk modulus, without taking into account possible correlations between bulk and shear modes.

The mathematical approach that we use to determine the range of effective moduli (K_0, μ_0) is the *translation method.* It originated in the work of Lurie & Cherkaev (1984b) and Murat and Tartar (see Tartar, 1985) and was applied to the conductivity of three-dimensional polycrystals by Avellaneda, Cherkaev, Lurie, and Milton (1988). The translation method consists in constructing suitable comparison functionals to estimate the elastic energy stored in an isotropic polycrystal. The procedure leads to a set of bounds which, in the present case, confine the possible values of the effective moduli to a rectangle in the (K, μ)-plane. We show that this rectangle corresponds precisely to the physical range of values for K_0 and μ_0 by exhibiting microgeometries that achieve every allowed pair. This involves carrying out explicit constructions of microgeometries. These microgeometries have a hierarchical, multilayered structure.

To describe the bounds it is convenient to introduce four crystal moduli c_{1111}, c_{2222}, c_{1122}, and c_{1212} which enter the Cartesian form of the constitutive relation

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{12} \end{pmatrix} = \begin{pmatrix} c_{1111} & c_{1122} & 0 \\ c_{1122} & c_{2222} & 0 \\ 0 & 0 & 2c_{1212} \end{pmatrix} \begin{pmatrix} \epsilon_{11} \\ \epsilon_{22} \\ \epsilon_{12} \end{pmatrix}, \tag{1.1}$$

for an orthotropic crystal aligned with the co-ordinate axes. Here the σ_{ij} and the ϵ_{ij} are the Cartesian components of the stress and strain fields. The elastic response of the isotropic polycrystal is described by a constitutive relation which in Cartesian co-ordinates takes the form

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{12} \end{pmatrix} = \begin{pmatrix} K_0 + \mu_0 & K_0 - \mu_0 & 0 \\ K_0 - \mu_0 & K_0 + \mu_0 & 0 \\ 0 & 0 & 2\mu_0 \end{pmatrix} \begin{pmatrix} \epsilon_{11} \\ \epsilon_{22} \\ \epsilon_{12} \end{pmatrix}, \tag{1.2}$$

incorporating a two-dimensional bulk modulus K_0 and a two-dimensional shear modulus μ_0.

Our findings are summarized by the following statements:

(i) Any isotropic two-dimensional polycrystal possesses a pair of effective elastic moduli K_0, μ_0 which satisfy the inequalities

$$K_l \le K_0 \le K_u, \tag{1.3}$$

$$\mu_l \leq \mu_0 \leq \mu_u, \tag{1.4}$$

where

$$K_l = \frac{c_{1111}c_{2222} - c_{1122}^2}{c_{1111} + c_{2222} - 2c_{1122}}, \tag{1.5}$$

$$K_u = \frac{1}{2}\left(\sqrt{c_{1111}c_{2222}} + c_{1122}\right), \tag{1.6}$$

$$\mu_l = \min\{\mu^+, \mu^-\}, \tag{1.7}$$

$$\mu_u = \max\{\mu^+, \mu^-\}, \tag{1.8}$$

$$\mu^+ = \frac{c_{1111}c_{2222} - c_{1122}^2}{2c_{1122} - 2c_{2222} + 2\sqrt{c_{2222}[c_{1111} + c_{2222} - 2c_{1122} + (c_{1111}c_{2222} - c_{1122}^2)/c_{1212}]}}, \tag{1.9}$$

$$\mu^- = \frac{c_{1111}c_{2222} - c_{1122}^2}{2c_{1122} - 2c_{1111} + 2\sqrt{c_{1111}[c_{1111} + c_{2222} - 2c_{1122} + (c_{1111}c_{2222} - c_{1122}^2)/c_{1212}]}}. \tag{1.10}$$

(ii) For any pair (K_0, μ_0) allowed by the inequalities (1.3)-(1.4) there exists an elastically isotropic microstructure with effective bulk modulus K_0 and effective shear modulus μ_0.

The question of which quantity μ^+ or μ^- forms the upper bound on the shear modulus and which quantity forms the lower bound is dictated by the sign of the quantity

$$\Delta = c_{1111}c_{2222} - (c_{1122} + 2c_{1212})^2. \tag{1.11}$$

Under the assumption that $c_{1111} \geq c_{2222}$ (which can be ensured by interchanging the x_1 and x_2 axes if necessary), and under the conditions that $c_{1111}c_{2222} - c_{1122}^2 > 0$ and $c_{1212} > 0$ (which are valid for any positive definite stiffness matrix), we have

$$\begin{aligned} \mu^+ &\geq \mu^- \quad \text{when} \quad \Delta \geq 0, \\ \mu^+ &\leq \mu^- \quad \text{when} \quad \Delta \leq 0, \end{aligned} \tag{1.12}$$

In the special case when $\Delta = 0$ the upper and lower bounds μ_l and μ_u coincide and determine the effective shear modulus exactly,

$$\mu_l = \mu_u = c_{1212} \quad \text{when} \quad \Delta = 0. \tag{1.13}$$

By contrast, the effective bulk modulus depends on the microgeometry of the polycrystal: the rectangle determined by the bounds degenerates to a line segment, not a point, when $\Delta = 0$.

We emphasize that these bounds, since they are attained by specific microgeometries, are the best possible in the absence of additional information on the microstructure. We describe in this paper an explicit constructive procedure for generating microstructures with moduli that range over all points in the (K, μ) plane contained in the rectangle allowed by the bounds (1.3)-(1.4).

As a corollary, the new bounds imply optimal bounds on the two-dimensional effective Poisson's ratio $\nu_0 = (K_0 - \mu_0)/(K_0 + \mu_0)$:

$$\frac{K_l - \mu_u}{K_l + \mu_u} \leq \nu_0 \leq \frac{K_u - \mu_l}{K_u + \mu_l}. \tag{1.14}$$

They also imply optimal bounds on the two-dimensional effective Young's modulus $E_0 = 4K_0\mu_0/(K_0 + \mu_0)$:

$$\frac{4K_l\mu_l}{K_l + \mu_l} \leq E_0 \leq \frac{4K_u\mu_u}{K_u + \mu_u}. \tag{1.15}$$

If the original crystal has square symmetry, i.e.

$$c_{1111} = c_{2222}, \tag{1.16}$$

then the rectangle determined by the bounds degenerates to the single point

$$K_l = K_u = K, \quad \mu_l = \mu_u = \frac{K}{-1 + \sqrt{(K + \mu^{(2)})(K + \mu^{(1)})/(\mu^{(1)}\mu^{(2)})}}, \tag{1.17}$$

where K, $\mu^{(1)}$ and $\mu^{(2)}$ are the bulk and two shear moduli of the square symmetric crystal:

$$K = (c_{1111} + c_{1122})/2, \quad \mu^{(1)} = (c_{1111} - c_{1122})/2, \quad \mu^{(2)} = c_{1212}. \tag{1.18}$$

Thus the effective moduli of such a polycrystal are uniquely determined. The uniqueness of the bulk modulus is well-known and was established by Hill (1952), the uniqueness of the shear modulus was established by Lurie and Cherkaev (1984a). In this paper we establish microstructure independence of the shear modulus not only when the crystal has square symmetry but also when $c_{1111}c_{2222} = (c_{1122} + 2c_{1212})^2$: see (1.13). Such exact formulae are especially useful because they provide stringent tests of approximation formulae. Microstructure independent formulae are known for other polycrystal properties too: Dykhne (1970) derived an exact formula for the conductivity of a two-dimensional polycrystal; Hashin (1984) and Schulgasser (1989) derived exact relations linking the effective thermal expansion coefficient with the effective bulk modulus; Milgrom and Shtrikman (1989) derived exact relations amongst the thermoelectric moduli; and Benveniste (1994) derived exact relations amongst the piezoelectric moduli.

formulae

2 A representation of symmetric fourth-order tensors for planar elasticity

2.1 Equations of planar elasticity for anisotropic materials

We consider the equations of planar elasticity for an anisotropic material. Let $\boldsymbol{\sigma}$ be the stress tensor field:

$$\nabla \cdot \boldsymbol{\sigma} = 0, \quad \boldsymbol{\sigma} = \boldsymbol{\sigma}^T, \tag{2.1}$$

and let $\boldsymbol{\epsilon}$ be the strain tensor field:

$$\boldsymbol{\epsilon} = \frac{1}{2}[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T], \tag{2.2}$$

where $\boldsymbol{u}$ is the displacement vector. The stresses and strains are connected by the anisotropic generalization of Hooke's law

$$\boldsymbol{\sigma} = \boldsymbol{C} : \boldsymbol{\epsilon}, \tag{2.3}$$

which in Cartesian co-ordinates has the representation:

$$\begin{aligned} \sigma_{11} &= c_{1111}\epsilon_{11} + c_{1122}\epsilon_{22} + 2c_{1112}\epsilon_{12}, \\ \sigma_{22} &= c_{2211}\epsilon_{11} + c_{2222}\epsilon_{22} + 2c_{2212}\epsilon_{12}, \\ \sigma_{12} &= c_{1211}\epsilon_{11} + c_{1222}\epsilon_{22} + 2c_{1212}\epsilon_{12}. \end{aligned} \tag{2.4}$$

The subindices of σ and ϵ indicate the different components of these tensors. This notation is equivalent to the standard engineering representation (see Christensen, 1979). The fourth-rank tensor $\boldsymbol{C}$ is symmetric and positive-definite. Its Cartesian components c_{ijkl} posess the usual symmetries of an elasticity tensor, *viz.*,

$$c_{ijkl} = c_{klij} = c_{jikl} = c_{ijlk}, \quad i, j, k, l = 1, 2. \tag{2.5}$$

In (2.3), as elsewhere, the symbol $\boldsymbol{A} : \boldsymbol{B}$ denotes a contraction of the last two indices of tensor $\boldsymbol{A}$ with the first two indices of tensor $\boldsymbol{B}$, i.e.

$$\boldsymbol{\sigma} = \boldsymbol{C} : \boldsymbol{\epsilon}, \quad \boldsymbol{C} = \boldsymbol{A} : \boldsymbol{B} \quad \text{means} \quad \sigma_{ij} = \sum_{k=1}^{2}\sum_{l=1}^{2} c_{ijkl}\boldsymbol{\epsilon}_{lk}, \quad c_{ijkl} = \sum_{m=1}^{2}\sum_{n=1}^{2} a_{ijmn}b_{nmkl} \tag{2.6}$$

It is convenient (see, for example, Lurie, Cherkaev, and Fedorov, 1982) to introduce the following orthonormal basis in the space of the second order tensors :

$$\begin{aligned} \boldsymbol{a}_1 &= (\boldsymbol{ii} + \boldsymbol{jj})/\sqrt{2}, \qquad \boldsymbol{a}_2 = (\boldsymbol{ii} - \boldsymbol{jj})/\sqrt{2}, \\ \boldsymbol{a}_3 &= (\boldsymbol{ij} + \boldsymbol{ji})/\sqrt{2}, \qquad \boldsymbol{a}_4 = (\boldsymbol{ij} - \boldsymbol{ji})/\sqrt{2}, \\ &\qquad\qquad \boldsymbol{a}_k : (\boldsymbol{a}_\ell)^T = \delta_{k\ell}, \end{aligned} \tag{2.7}$$

where $\boldsymbol{i}$ and $\boldsymbol{j}$ are the unit vectors of the Cartesian x_1 and x_2 axes, $\delta_{k\ell}$ is the Kronecker symbol, and $\boldsymbol{ij}$ represents the direct tensor product of $\boldsymbol{i}$ and $\boldsymbol{j}$. These tensors have the following matrix form

$$\begin{aligned} \boldsymbol{a}_1 &= \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad \boldsymbol{a}_2 = \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \\ \boldsymbol{a}_3 &= \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \boldsymbol{a}_4 = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} \end{aligned} \tag{2.8}$$

in the Cartesian basis $\boldsymbol{i}$, $\boldsymbol{j}$. The subspace of the symmetric second order tensors admits the orthonormal basis $\boldsymbol{a}_1,\ \boldsymbol{a}_2,\ \boldsymbol{a}_3$. Symmetric strain and stress tensors have the following representation in this basis

$$\boldsymbol{\epsilon} = \sum_{i=1}^{3} \epsilon_i \boldsymbol{a}_i \ , \quad \boldsymbol{\sigma} = \sum_{i=1}^{3} \sigma_i \boldsymbol{a}_i, \tag{2.9}$$

where the coefficients ϵ_i , σ_i are given by the equations

$$\begin{gathered}\epsilon_1 = \boldsymbol{\epsilon} : \boldsymbol{a}_1 = \frac{1}{\sqrt{2}}(\epsilon_{11} + \epsilon_{22}), \quad \epsilon_2 = \boldsymbol{\epsilon} : \boldsymbol{a}_2 = \frac{1}{\sqrt{2}}(\epsilon_{11} - \epsilon_{22}), \quad \epsilon_3 = \boldsymbol{\epsilon} : \boldsymbol{a}_3 = \sqrt{2}\epsilon_{12}, \\ \sigma_1 = \boldsymbol{\sigma} : \boldsymbol{a}_1 = \frac{1}{\sqrt{2}}(\sigma_{11} + \sigma_{22}), \quad \sigma_2 = \boldsymbol{\sigma} : \boldsymbol{a}_2 = \frac{1}{\sqrt{2}}(\sigma_{11} - \sigma_{22}), \quad \sigma_3 = \boldsymbol{\sigma} : \boldsymbol{a}_3 = \sqrt{2}\sigma_{12}.\end{gathered} \tag{2.10}$$

In this representation Hooke's law (2.3) has the form

$$\sigma_j = \sum_{i=1}^{3} c_{ji}\epsilon_i \ , \quad j = 1, 2, 3, \tag{2.11}$$

where the coefficients c_{ij} equal

$$\begin{gathered}c_{11} = \frac{1}{2}(c_{1111} + c_{2222} + 2c_{1122}), \quad c_{12} = \frac{1}{2}(c_{1111} - c_{2222}), \quad c_{13} = c_{1112} + c_{2212}, \\ c_{22} = \frac{1}{2}(c_{1111} + c_{2222} - 2c_{1122}), \quad c_{23} = c_{1112} - c_{2212}, \quad c_{33} = 2c_{1212},\end{gathered} \tag{2.12}$$

when expressed in terms of the Cartesian coefficients c_{ijkl}. The anisotropic stiffness tensor $\boldsymbol{C}$ is represented by the symmetric positive definite matrix

$$\boldsymbol{C} = \begin{pmatrix} c_{11} & c_{12} & c_{13} \\ c_{12} & c_{22} & c_{23} \\ c_{13} & c_{23} & c_{33} \end{pmatrix}. \tag{2.13}$$

Aside from a constant factor 1/2, which we choose to omit, the elastic energy density can be represented either as a quadratic form in the strains

$$W_\epsilon = \boldsymbol{\epsilon} : \boldsymbol{C} : \boldsymbol{\epsilon}, \tag{2.14}$$

or as a quadratic form in the stresses

$$W_\sigma = \boldsymbol{\sigma} : \boldsymbol{S} : \boldsymbol{\sigma}, \tag{2.15}$$

where $\boldsymbol{S} = \boldsymbol{C}^{-1}$ is the compliance tensor.

2.2 The action of the rotation group on fourth-order tensors

Here we discuss the transformation of the elements of the fourth-order tensor due to a rotation of the Cartesian axes. Consider two different systems of axes with orthonormal basis vectors $\boldsymbol{i}, \boldsymbol{j}$ and $\boldsymbol{i}', \boldsymbol{j}'$ and let ϕ denote the angle between $\boldsymbol{i}$ and $\boldsymbol{i}'$, i.e.,

$$\begin{aligned} \boldsymbol{i}' &= \boldsymbol{i}\ \cos\phi + \boldsymbol{j}\sin\phi, \\ \boldsymbol{j}' &= -\boldsymbol{i}\sin\phi + \boldsymbol{j}\cos\phi, \end{aligned} \tag{2.16}$$

or, in matrix form:

$$\begin{pmatrix} \boldsymbol{i}' \\ \boldsymbol{j}' \end{pmatrix} = \begin{pmatrix} \cos\phi & \sin\phi \\ -\sin\phi & \cos\phi \end{pmatrix} \begin{pmatrix} \boldsymbol{i} \\ \boldsymbol{j} \end{pmatrix}. \tag{2.17}$$

We associate the tensor basis $\boldsymbol{a}_1', \boldsymbol{a}_2', \boldsymbol{a}_3'$ with the vectors $\boldsymbol{i}', \boldsymbol{j}'$ in the same way as (2.7) associates the basis $\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3$ with the vectors $\boldsymbol{i}, \boldsymbol{j}$. It is easy to verify that the two bases are connected by the relationships

$$\begin{aligned} \boldsymbol{a}_1' &= \boldsymbol{a}_1, \\ \boldsymbol{a}_2' &= \quad \cos 2\phi\ \boldsymbol{a}_2 + \sin 2\phi\ \boldsymbol{a}_3, \\ \boldsymbol{a}_3' &= -\sin 2\phi\ \boldsymbol{a}_3 + \cos 2\phi\ \boldsymbol{a}_3, \end{aligned} \tag{2.18}$$

or, in matrix form,

$$\boldsymbol{a}' = \boldsymbol{R} : \boldsymbol{a}, \tag{2.19}$$

where $\boldsymbol{R} = \boldsymbol{R}(\phi)$ is the rotation matrix

$$\boldsymbol{R} = \boldsymbol{R}(\phi) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos 2\phi & \sin 2\phi \\ 0 & -\sin 2\phi & \cos 2\phi \end{pmatrix}, \tag{2.20}$$

$\boldsymbol{a}' = (\boldsymbol{a}_1', \boldsymbol{a}_2', \boldsymbol{a}_3')$, and $\boldsymbol{a} = (\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3)$. Under this change of basis, the matrix $\boldsymbol{C}$ transforms to a new matrix

$$\boldsymbol{C}' = \boldsymbol{R} : \boldsymbol{C} : \boldsymbol{R}^T. \tag{2.21}$$

This transformation can be rewritten either as

$$\begin{aligned} c_{11}' &= c_{11}, \\ c_{12}' &= c_{12}\cos 2\phi + c_{13}\sin 2\phi, \\ c_{13}' &= -c_{12}\sin 2\phi + c_{13}\cos 2\phi, \\ c_{22}' &= c_{22}\cos^2 2\phi + c_{33}\cos^2 2\phi + 2c_{23}\sin 2\phi\cos 2\phi, \\ c_{23}' &= (c_{33} - c_{22})\cos 2\phi\sin 2\phi + c_{23}(\cos^2 2\phi - \sin^2 2\phi), \\ c_{33}' &= c_{33}\cos^2 2\phi + c_{22}\cos^2 2\phi - 2c_{23}\sin 2\phi\cos 2\phi, \end{aligned} \tag{2.22}$$

$$\tag{2.23}$$

or, more concisely, as

$$c_{11}' = c_{11}, \tag{2.24}$$

$$\begin{pmatrix} c'_{12} \\ c'_{13} \end{pmatrix} = \begin{pmatrix} \cos 2\phi & \sin 2\phi \\ -\sin 2\phi & \cos 2\phi \end{pmatrix} \begin{pmatrix} c_{12} \\ c_{13} \end{pmatrix}, \tag{2.25}$$

$$\begin{pmatrix} c'_{22} & c'_{23} \\ c'_{23} & c'_{33} \end{pmatrix} = \begin{pmatrix} \cos 2\phi & \sin 2\phi \\ -\sin 2\phi & \cos 2\phi \end{pmatrix} \begin{pmatrix} c_{22} & c_{23} \\ c_{23} & c_{33} \end{pmatrix} \begin{pmatrix} \cos 2\phi & \sin 2\phi \\ -\sin 2\phi & \cos 2\phi \end{pmatrix}^T. \tag{2.26}$$

Observe that the action of the rotation group on symmetric fourth-order tensors $\boldsymbol{C}$ can be understood in terms of the action of the rotation group on the scalar

$$c_s = c_{11}, \tag{2.27}$$

the 2−vector

$$\boldsymbol{c}_v = (c_{12}, c_{13}), \tag{2.28}$$

and the 2×2 matrix

$$\boldsymbol{C}_m = \begin{pmatrix} c_{22} & c_{23} \\ c_{23} & c_{33} \end{pmatrix}. \tag{2.29}$$

It is clear now that a fourth-order elasticity tensor possesses five rotational invariants. Namely, it has the two linear invariants

$$\begin{aligned} I_1 &= c_{11}, \\ I_2 &= Tr\ \boldsymbol{C}_m = c_{22} + c_{33}, \end{aligned} \tag{2.30}$$

two quadratic invariants

$$\begin{aligned} I_3 &= ||\boldsymbol{c}_v|| = c_{12}^2 + c_{13}^2, \\ I_4 &= \det \boldsymbol{C}_m = c_{22}c_{33} - c_{23}^2, \end{aligned} \tag{2.31}$$

and the cubic invariant

$$I_5 = \det \boldsymbol{C}. \tag{2.32}$$

2.3 Orthotropy, square symmetry, and isotropy

It is convenient to use the preceding representation to characterize various special classes of material symmetry for the single crystal.

Orthotropic symmetry corresponds to the case when the vector $\boldsymbol{c}_v$ is directed along one of the principal axes of the matrix $\boldsymbol{C}_m$:

$$\boldsymbol{C}_m \cdot \boldsymbol{c}_v = \alpha \boldsymbol{c}_v, \quad \text{for some } \alpha \in \mathbb{R}. \tag{2.33}$$

This means that in the coordinates which diagonalize the matrix $\boldsymbol{C}_m$ the stiffness matrix $\boldsymbol{C}$ has the form

$$\boldsymbol{C} = \begin{pmatrix} c_{11} & c_{12} & 0 \\ c_{12} & c_{22} & 0 \\ 0 & 0 & c_{33} \end{pmatrix}, \tag{2.34}$$

and the associated compliance matrix $\boldsymbol{S} = \boldsymbol{C}^{-1}$ has a similar form,

$$\boldsymbol{S} = \begin{pmatrix} s_{11} & s_{12} & 0 \\ s_{12} & s_{22} & 0 \\ 0 & 0 & s_{33} \end{pmatrix}, \tag{2.35}$$

where

$$s_{11} = \frac{c_{22}}{c_{11}c_{22} - (c_12)^2}, \quad s_{22} = \frac{c_{11}}{c_{11}c_{22} - (c_12)^2}, \quad s_{12} = \frac{-c_{12}}{c_{11}c_{22} - (c_12)^2}, \quad s_{33} = \frac{1}{c_{33}}. \tag{2.36}$$

In other words, the principal axes can be chosen in such a way that the trace-free basis tensor $\boldsymbol{a}_3$ becomes an eigenvector of the matrix $\boldsymbol{C}$. We will assume that the principal axes $\boldsymbol{i}$ and $\boldsymbol{j}$ are chosen so that

$$c_{12} \geq 0. \tag{2.37}$$

The class of anisotropic materials with square symmetry (the two-dimensional analogue of cubic symmetry) corresponds to an orthotropic medium in which the amplitude of the vector $\boldsymbol{c}_v$ vanishes, i.e.,

$$c_{12} = c_{13} = 0. \tag{2.38}$$

The tensor $\boldsymbol{a}_1$ then is also an eigenvector of the matrix $\boldsymbol{C}$ which becomes diagonal in this basis (2.7), viz.,

$$\boldsymbol{C} = \begin{pmatrix} c_{11} & 0 & 0 \\ 0 & c_{22} & 0 \\ 0 & 0 & c_{33} \end{pmatrix} = \begin{pmatrix} 2K & 0 & 0 \\ 0 & 2\mu^{(1)} & 0 \\ 0 & 0 & 2\mu^{(2)} \end{pmatrix}, \tag{2.39}$$

where $K = c_{11}/2$ is the two-dimensional bulk modulus, $\mu^{(1)} = c_{22}/2$ and $\mu^{(2)} = c_{33}/2$ are the shear moduli.

Finally, isotropy is a special class of square symmetry in which the matrix $\boldsymbol{C}_m$ is proportional to the identity:

$$c_{22} = c_{33} = 2\mu \;\rightarrow\; \boldsymbol{C}_m = 2\mu\boldsymbol{I}. \tag{2.40}$$

The stiffness tensor of an isotropic medium

$$\boldsymbol{C}(K,\mu) = \begin{pmatrix} c_{11} & 0 & 0 \\ 0 & c_{22} & 0 \\ 0 & 0 & c_{22} \end{pmatrix} = \begin{pmatrix} 2K & 0 & 0 \\ 0 & 2\mu & 0 \\ 0 & 0 & 2\mu \end{pmatrix}. \tag{2.41}$$

is defined by two constants: the bulk modulus $K = c_{11}/2$ and the shear modulus $\mu = c_{22}/2 = c_{33}/2$.

3 Applying the translation method to obtain the bounds

Consider a polycrystal which is an assemblage of grains of the same homogeneous anisotropic material, having different orientations with respect to the laboratory frame. At each point $\boldsymbol{x}$ within the material the elasticity tensor field is given by

$$\boldsymbol{C}(\boldsymbol{x}) = \boldsymbol{C}(\phi(\boldsymbol{x})) = \boldsymbol{R}(\phi(\boldsymbol{x})) : \boldsymbol{C} : \boldsymbol{R}^T(\phi(\boldsymbol{x})), \tag{3.1}$$

where $\phi(\boldsymbol{x})$ describes the orientation of the grain containing $\boldsymbol{x}$, and $\boldsymbol{R}(\phi)$ is given by (2.20).

We are interested in the *homogenization problem*, i.e. in describing the effective stiffness properties of such a material. As customary, we assume that the sizes of the grains are much smaller than the sample size and the wavelength of the applied field. Under these assumptions, the material behaves as a homogeneous continuum with an effective elasticity tensor $\boldsymbol{C}_0$ which describes the relation between averaged stress and strain fields, i.e.

$$\begin{aligned} \langle \boldsymbol{\sigma} \rangle &= \boldsymbol{C}_0 : \langle \boldsymbol{\epsilon} \rangle \ , \\ \langle \boldsymbol{\epsilon} \rangle &= \frac{1}{2}[\langle \nabla \boldsymbol{u} \rangle + \langle \nabla \boldsymbol{u} \rangle^T]. \end{aligned} \tag{3.2}$$

Here the symbol $\langle \cdot \rangle$ denotes volume averaging. In general, this tensor is anisotropic, positive definite and symmetric. It depends on the single-crystal stiffness tensor $\boldsymbol{C}$ as well as on geometrical structure of the polycrystal (the arrangement of the crystallites). However, due to our assumption of macroscopic homogeneity, the effective tensor is independent of the particular loading mode.

To derive the bounds characterizing the range of feasible effective properties, it is convenient to use the representation (2.34) for the stiffness tensor of the crystal instead of the Cartesian representation used in the introduction. The bounds (1.5)-(**??**) can be rewritten in terms of the coefficients c_{ij} as follows:

$$K_l = \frac{c_{11}c_{22} - c_{12}^2}{2c_{22}}, \tag{3.3}$$

$$K_u = \frac{c_{11}c_{22} - c_{12}^2}{c_{22} - c_{11} + \sqrt{(c_{11} + c_{22})^2 - 4c_{12}^2}}, \tag{3.4}$$

$$\mu^+ = \frac{c_{11}c_{22} - c_{12}^2}{-2c_{22} + 2c_{12} + 2\sqrt{[c_{22} + (c_{11}c_{22} - c_{12}^2)/c_{33}][c_{11} + c_{22} - 2c_{12}]}}, \tag{3.5}$$

$$\mu^- = \frac{c_{11}c_{22} - c_{12}^2}{-2c_{22} - 2c_{12} + 2\sqrt{[c_{22} + (c_{11}c_{22} - c_{12}^2)/c_{33}][c_{11} + c_{22} + 2c_{12}]}}. \tag{3.6}$$

Also

$$\Delta_1 = (c_{11} + c_{33})(c_{22} - c_{33}) - c_{12}^2, \tag{3.7}$$

$$\Delta_2 = c_{33}(c_{22} - c_{11})/2 + c_{11}c_{22} - c_{12}^2, \tag{3.8}$$

and the assumption $c_{1111} \geq c_{2222}$ is equivalent to the condition $c_{12} \geq 0$.

We now proceed to give a description of the translation method in the form which will be used to obtain bounds on the tensor $\boldsymbol{C}_0$.

3.1 Preliminaries: what quantities do we need to bound ?

The notion of effective stiffness tensor can also be derived ¿from (stored) elastic energy considerations: the effective tensor should correspond to an " equivalent " energy density storing the same amount of energy under any given applied load. In fact, the energy stored the composite material under an applied load is known (again omitting the factor of 1/2) to be equal to

$$W^* = \langle \boldsymbol{\epsilon} \rangle \ : \boldsymbol{C}_0 : \ \langle \boldsymbol{\epsilon} \rangle = \langle \boldsymbol{\sigma} \rangle \ : \boldsymbol{S}_0 : \ \langle \boldsymbol{\sigma} \ \rangle, \tag{3.9}$$

where $\boldsymbol{\epsilon}$ and $\boldsymbol{\sigma}$ are the actual strain and stress microfields in the composite and the effective compliance tensor $\boldsymbol{S}_0$ is the inverse of the effective elasticity tensor $\boldsymbol{C}_0$, i.e. $\boldsymbol{S}_0 = \boldsymbol{C}_0^{-1}$ (see, e.g., Christensen, 1979; Sanchez - Palencia, 1980).

The variational principles of minimum energy

$$W_\epsilon^* = \boldsymbol{\epsilon}_0 : \boldsymbol{C}_0 : \boldsymbol{\epsilon}_0 = \inf_{\substack{\boldsymbol{\epsilon} \\ \boldsymbol{\epsilon} = \frac{1}{2}[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T], \\ \langle \boldsymbol{\epsilon} \rangle = \boldsymbol{\epsilon}_0}} \langle \boldsymbol{\epsilon} : \boldsymbol{C}(\boldsymbol{x}) : \boldsymbol{\epsilon} \rangle \tag{3.10}$$

and minimum complimentary energy

$$W_\sigma^* = \boldsymbol{\sigma}_0 : \boldsymbol{S}_0 : \boldsymbol{\sigma}_0 = \inf_{\substack{\boldsymbol{\sigma} \\ \nabla \cdot \boldsymbol{\sigma} = 0, \boldsymbol{\sigma} = \boldsymbol{\sigma}^T, \\ \langle \boldsymbol{\sigma} \rangle = \boldsymbol{\sigma}_0}} \langle \boldsymbol{\sigma} : \boldsymbol{S}(\boldsymbol{x}) : \boldsymbol{\sigma} \rangle \tag{3.11}$$

can be used to estimate the energy stored in the composite and hence to obtain bounds on the effective tensor $\boldsymbol{C}_0 = \boldsymbol{S}_0^{-1}$. Unlike in (3.9), the fields $\boldsymbol{\epsilon}$ and $\boldsymbol{\sigma}$ entering these variational principles can be any kinematically admissible strain and stress fields. The actual fields are those which deliver the minimum values to the functionals (3.10) and (3.11).

The translation method is based on the lower bound of the energy functional J

$$J = \sum_{i=1}^{N} W_i^*, \tag{3.12}$$

which is equal to the sum of the values $W_i^* = \boldsymbol{\epsilon}_i : \boldsymbol{C}_0 : \boldsymbol{\epsilon}_i$ or $W_i^* = \boldsymbol{\sigma}_i : \boldsymbol{S}_0 : \boldsymbol{\sigma}_i$ of the average elastic energy stored in the polycrystal when it is exposed sequentially to N linearly independent external strain or stress fields with fixed mean values $\boldsymbol{\epsilon}_i$ and $\boldsymbol{\sigma}_i$ (see, e.g., Cherkaev and Gibiansky, 1993).

To obtain a *lower* bound on the effective bulk modulus K_0 one can subject the composite to the external hydrostatic *strain* field

$$\langle \boldsymbol{\epsilon} \rangle = \boldsymbol{\epsilon}_h = \epsilon_h \boldsymbol{a}_1, \tag{3.13}$$

where ϵ_h is a given constant. Indeed, the energy W_h^* of an isotropic composite exposed in this field is proportional to the effective bulk modulus K_0 (see (2.41)):

$$W_h^* = \boldsymbol{\epsilon}_h : \boldsymbol{C}_0 : \boldsymbol{\epsilon}_h = 2K_0 \epsilon_h^2. \tag{3.14}$$

The corresponding functional has the form:

$$J^{\epsilon} = \inf_{\substack{\boldsymbol{\epsilon} \\ \boldsymbol{\epsilon} = \frac{1}{2}[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T], \\ \langle \boldsymbol{\epsilon} \rangle = \boldsymbol{\epsilon}_h}} \langle\, \boldsymbol{\epsilon}(\boldsymbol{x}) : \boldsymbol{C}(\boldsymbol{x}) : \boldsymbol{\epsilon}(\boldsymbol{x}) \,\rangle = 2K_0 \epsilon_h^2, \tag{3.15}$$

(see (3.10)). Any lower bound on the functional (3.15) gives us a lower bound on the effective bulk modulus K_0, because the amplitude ϵ_h of the hydrostatic strain field $\boldsymbol{\epsilon}_h$ is fixed.

Remark: It is convenient for us (see Cherkaev and Gibiansky, 1993) to rewrite the energy as a quadratic form of the gradient of displacement $\boldsymbol{\zeta}$ instead of strain $\boldsymbol{\epsilon}$:

$$\boldsymbol{\zeta} = \nabla \boldsymbol{u}, \quad \boldsymbol{\epsilon} = \tfrac{1}{2}(\boldsymbol{\zeta} + \boldsymbol{\zeta}^T)\,. \tag{3.16}$$

This quadratic form is associated with a four by four matrix C' which has the representation

$$C' = \begin{pmatrix} c_{11} & c_{12} & c_{13} & 0 \\ c_{12} & c_{22} & c_{23} & 0 \\ c_{13} & c_{23} & c_{33} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \tag{3.17}$$

in the basis (2.7).

To obtain an *upper* bound on the bulk modulus we need to subject the composite to a hydrostatic *stress* field $\boldsymbol{\sigma}_h = \sigma_h\, \boldsymbol{a}_1$. Any lower bound on the corresponding functional

$$J^{\sigma} = \inf_{\substack{\boldsymbol{\sigma} \\ \nabla \cdot \boldsymbol{\sigma} = 0, \\ \boldsymbol{\sigma} = \boldsymbol{\sigma}^T, \langle \boldsymbol{\sigma} \rangle = \boldsymbol{\sigma}_h}} \langle\, \boldsymbol{\sigma}(\boldsymbol{x}) : \boldsymbol{S}(\boldsymbol{x}) : \boldsymbol{\sigma}(\boldsymbol{x}) \,\rangle = (2K_0)^{-1} \sigma_h^2 \tag{3.18}$$

then gives us an upper bound on K_0.

Similarly, to obtain a lower bound on the shear modulus of the mixture one can examine the energy stored in the composite when it is subjected to a shear-type trial strain field. We emphasize here that this procedure of using functionals also applies to anisotropic composites, and in particular to composites with square symmetry which have two effective shear moduli. A functional that incorporates only one shear-type strain field delivers information on only the lowest of the two shear moduli when the composite is allowed to rotate relative to the applied field. Such a functional is therefore not sensitive to the value of the other shear modulus. One might expect that the lower of the two shear moduli, taken over all composites with square symmetry, would be less than the minimum value of the effective shear modulus of isotropic composites, which are necessarily restricted to have two *equal* effective shear moduli. In other words, we anticipate that the composite would "prefer" to have square symmetry (or perhaps orthotropic symmetry) to minimize the energy associated with a single shear field. To take into account the assumption made on the the isotropy of the mixture, we should also consider the response of the composite to the orthogonal shear mode. Thus, to find a suitable lower bound on the shear modulus of an isotropic composite, we should a functional corresponding to the sum of the elastic energies stored by the material under the action of two trial orthogonal shear fields $\boldsymbol{\epsilon}_s^{(1)} = \epsilon_s^{(1)}\boldsymbol{a}_2$ and $\boldsymbol{\epsilon}_s^{(2)} = \epsilon_s^{(2)}\boldsymbol{a}_3$:

$$J^{\epsilon\epsilon} = \boldsymbol{\epsilon}_s^{(1)} : \boldsymbol{C}_0 : \boldsymbol{\epsilon}_s^{(1)} + \boldsymbol{\epsilon}_s^{(2)} : \boldsymbol{C}_0 : \boldsymbol{\epsilon}_s^{(2)} = 2\mu_0[(\epsilon_s^{(1)})^2 + (\epsilon_s^{(2)})^2], \tag{3.19}$$

where $\epsilon_s^{(1)}$ and $\epsilon_s^{(2)}$ are fixed constants.

To find an upper bound of the shear modulus we use the functional equal to the sum of energies stored by the composite exposed to two orthogonal shear fields of stresses $\boldsymbol{\sigma}_s^{(1)} = \sigma_s^{(1)}\boldsymbol{a}_2$ and $\boldsymbol{\sigma}_s^{(2)} = \sigma_s^{(2)}\boldsymbol{a}_3$:

$$J^{\sigma\sigma} = \boldsymbol{\sigma}_s^{(1)} : \boldsymbol{C}_0^{-1} : \boldsymbol{\sigma}_s^{(1)} + \boldsymbol{\sigma}_s^{(2)} : \boldsymbol{C}_0^{-1} : \boldsymbol{\sigma}_s^{(2)} = ((\sigma_s^{(1)})^2 + (\sigma_s^{(2)})^2)/2\mu_0, \tag{3.20}$$

where $\sigma_s^{(1)}$ and $\sigma_s^{(2)}$ are given constants.

Remark: In general, in order to get coupled shear-bulk bounds one needs to expose the composite to three different fields: one hydrostatic and two orthogonal shear ones, each chosen to be a stress or strain as appropriate. This procedure was employed in the paper by Cherkaev and Gibiansky (1993) to obtain coupled bounds for the effective shear and bulk moduli of isotropic composites of two isotropic phases. As we will see below, and as is evident from the description in the Introduction, the best possible bounds on the effective bulk and shear moduli of an isotropic polycrystal are uncoupled, i.e. they form a rectangle in the (K_0, μ_0) plane. That is why this consideration is unnecessary.

Remark: It has been rigorously proved [see Remark 3.7 in Milton (1994) and also Francfort and Milton (1994)] that knowledge of the minimal values of such functionals (3.12) for all combinations of applied stress and strain fields, allows one to construct the boundary of the entire G-closure, i.e. the set of all effective elasticity tensors which are realized by some microstrucure. In the same way that convex sets can be characterized through their Legendre transforms, G-closures be characterized through the minimal values of these functionals. The G-closure is not convex but it has sufficient convexity properties (due to its stability under lamination) to ensure that it can be characterized by the minimal values of these functionals.

3.2 Derivation of the bounds

Note, that each of the functionals described above is a quadratic form of the fields. Any such functional can be rewritten in the compact form

$$J = \boldsymbol{E}_0 \cdot \boldsymbol{D}_0 \cdot \boldsymbol{E}_0 = \inf_{\substack{\boldsymbol{E} \\ \boldsymbol{E} \in \mathcal{F},\ \langle \boldsymbol{E} \rangle = \boldsymbol{E}_0}} \langle\ \boldsymbol{E} \cdot \boldsymbol{D}(\boldsymbol{x}) \cdot \boldsymbol{E}\ \rangle, \tag{3.21}$$

where $\boldsymbol{D}_0$ and $\boldsymbol{D}(\boldsymbol{x})$ are appropriate matrices, describing respectively the effective and local properties. The matrix $\boldsymbol{D}(\boldsymbol{x})$ either coincides with the stiffness matrix $\boldsymbol{C}(x)$ (functional (3.15)), or with the compliance matrix $\boldsymbol{S}(x)$ (functional (3.18)), or it has the block-diagonal form

$$\boldsymbol{D}(x) = \begin{pmatrix} \boldsymbol{C}(\boldsymbol{x}) & 0 \\ 0 & \boldsymbol{C}(\boldsymbol{x}) \end{pmatrix} \tag{3.22}$$

(functional (3.19)), or the form:

$$\boldsymbol{D}(x) = \begin{pmatrix} \boldsymbol{S}(\boldsymbol{x}) & 0 \\ 0 & \boldsymbol{S}(\boldsymbol{x}) \end{pmatrix} \tag{3.23}$$

(functional (3.20)). The components of the vector $\boldsymbol{E}$ consist of strain or stress tensors, according to the nature of $\boldsymbol{D}(\boldsymbol{x})$. The set $\mathcal{F}$ is a set of Ω- periodic vector fields with prescribed mean values and satisfying some differential restrictions. For the components of $\boldsymbol{E}$ which are stresses these restrictions express the equilibrium equations

$$\nabla \cdot \boldsymbol{\sigma} = 0, \quad \boldsymbol{\sigma}^T = \boldsymbol{\sigma}, \tag{3.24}$$

and for the components of $\boldsymbol{E}$ which are strains they express the infinitesimal strain compatibility conditions

$$\mathrm{curl}(\mathrm{curl}\ \boldsymbol{\epsilon})^T = 0, \quad \text{or} \quad \boldsymbol{\epsilon} = \frac{1}{2}[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T] \tag{3.25}$$

(Atkin and Fox, 1990). The idea of the translation method is to use these differential restrictions to bound (3.21).

As an example, the functional (3.15) is associated with the set $\mathcal{F}$:

$$\mathcal{F} = \{\boldsymbol{E} \mid \ \boldsymbol{E} = \frac{1}{2}[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^T], \quad \boldsymbol{E} \ \text{ is } \ \Omega - \text{periodic}\}. \tag{3.26}$$

The matrix field $\boldsymbol{D}(\boldsymbol{x})$ in the polycrystal has the form as the matrix field $\boldsymbol{C}(\boldsymbol{x})$, i.e. it can be expressed in terms of the moduli of the crystal through the equation

$$\boldsymbol{D}(\boldsymbol{x}) = \boldsymbol{D}(\phi(\boldsymbol{x})) = \boldsymbol{\mathcal{R}}(\phi(\boldsymbol{x})) : \boldsymbol{D} : \boldsymbol{\mathcal{R}}^T(\phi(\boldsymbol{x})), \tag{3.27}$$

where $\boldsymbol{\mathcal{R}}(\phi)$) either coincides with $\boldsymbol{R}(\phi)$ given by (2.20) (functionals (3.15) and (3.18)) or has the block diagonal form

$$\boldsymbol{\mathcal{R}}(\phi) = \begin{pmatrix} \boldsymbol{R}(\phi) & 0 \\ 0 & \boldsymbol{R}(\phi) \end{pmatrix} \tag{3.28}$$

(functionals (3.19) and (3.20)).

Our goal is to bound the infimum of the functional J over all fields $\boldsymbol{E} \in \mathcal{F}$ with given average value $\boldsymbol{E}_0$ and over all microstructures, i.e. over all orientation fields $\phi(\boldsymbol{x})$:

$$\inf_{\phi(\boldsymbol{x})} \quad \inf_{\substack{\boldsymbol{E} \\ \boldsymbol{E} \in \mathcal{F}, \ \langle \boldsymbol{E} \rangle = \boldsymbol{E}_0}} \quad \langle \boldsymbol{E} \cdot \boldsymbol{D}(\phi) \cdot \boldsymbol{E} \rangle. \tag{3.29}$$

To obtain a bound on this infimum we will use the translation method introduced by Lurie and Cherkaev (1984b, 1986a), Murat and Tartar (1985), Tartar (1978,1985). The detailed description of this method can be found in the papers by Lurie and Cherkaev (1986b), Cherkaev and Gibiansky (1993), Milton (1990a,b). Here we adapt the method to the elastic polycrystal problem in a similar way to what was done by Avellaneda, Cherkaev, Lurie, and Milton (1988) for the conducting polycrystal.

The translation method hinges on the existence of *quasiconvex* (Dacorogna 1982) quadratic functions of the field $\boldsymbol{E}$

$$\Phi(\boldsymbol{E}) = \boldsymbol{E} \cdot \boldsymbol{T} \cdot \boldsymbol{E}. \tag{3.30}$$

These are functions that, like convex functions, satisfy

$$\langle \boldsymbol{E} \cdot \boldsymbol{T} \cdot \boldsymbol{E} \rangle \geq \langle \boldsymbol{E} \rangle \cdot \boldsymbol{T} \cdot \langle \boldsymbol{E} \rangle \tag{3.31}$$

for all $\boldsymbol{E} \in \mathcal{F}$. Of course, any positive semidefinite matrix $\boldsymbol{T}$ generates a quasiconvex quadratic form. The main interest of quasiconvex functions lies, however, in the fact that $\boldsymbol{T}$ need not be definite, due to the differential (kinematic) constraints implicit in the definition $\mathcal{F}$. A matrix $\boldsymbol{T}$ associated to a quasiconvex function will be referred to as a *translation matrix*.

Suppose we have found a quasiconvex function associated with the translation matrix $\boldsymbol{T}$. Let us add the corresponding function $\Phi(\boldsymbol{E})$ to the functional J and then subtract it:

$$J = \langle \boldsymbol{E} \cdot (\boldsymbol{D}(\boldsymbol{x}) - \boldsymbol{T}) \cdot \boldsymbol{E} \rangle + \langle \boldsymbol{E} \cdot \boldsymbol{T} \cdot \boldsymbol{E} \rangle, \tag{3.32}$$

implying

$$\langle \boldsymbol{E} \rangle \cdot (\boldsymbol{D}_0 - \boldsymbol{T}) \cdot \langle \boldsymbol{E} \rangle \geq \langle \boldsymbol{E} \cdot (\boldsymbol{\mathcal{R}}^T(\phi(\boldsymbol{x})) \cdot \boldsymbol{D} \cdot \boldsymbol{\mathcal{R}}(\phi(\boldsymbol{x})) - \boldsymbol{T}) \cdot \boldsymbol{E} \rangle. \tag{3.33}$$

We choose the translation tensor $\boldsymbol{T}$ in such a way that the matrix $[\boldsymbol{\mathcal{R}}^T(\phi(\boldsymbol{x})) \cdot \boldsymbol{D} \cdot \boldsymbol{\mathcal{R}}(\phi(\boldsymbol{x})) - \boldsymbol{T}]$ is positive definite, or perhaps positive semidefinite, for all $\boldsymbol{x}$:

$$\boldsymbol{\mathcal{R}}^T(\phi(\boldsymbol{x})) \cdot \boldsymbol{D} \cdot \boldsymbol{\mathcal{R}}(\phi(\boldsymbol{x})) - \boldsymbol{T} \geq 0, \quad \forall \ \ \boldsymbol{x}. \tag{3.34}$$

Then the right hand side of (3.33) is also nonnegative, which means that

$$\boldsymbol{D}_0 - \boldsymbol{T} \geq 0. \tag{3.35}$$

The inequality (3.34) serves to restrict the set of matrices $\boldsymbol{T}$ that can be used to yield bounds. The inequality (3.35) gives us the bounds on the tensor $\boldsymbol{D}_0$. It is valid with any admissible quasiconvex matrix $\boldsymbol{T}$. Clearly, the inequality (3.35) restricts the set of possible effective tensors independently of their microstructure and provides bounds which apply to anisotropic effective tensors as well as to isotropic ones.

For the purpose of bounding the effective moduli of isotropic polycrystals we will see that it is sufficient to restrict ourselves to isotropic translations $\boldsymbol{T}_0$ such that

$$\boldsymbol{\mathcal{R}}^T(\phi(\boldsymbol{x})) \cdot \boldsymbol{T}_0 \cdot \boldsymbol{\mathcal{R}}(\phi(\boldsymbol{x})) = \boldsymbol{T}_0. \tag{3.36}$$

In this case the requirement (3.34) is rotationally invariant and reduces to the simple constraint that

$$\boldsymbol{D} - \boldsymbol{T}_0 \geq 0. \tag{3.37}$$

Let us summarize *the bounds*: for any effective stiffness tensor $\boldsymbol{C}_0$ of a polycrystal the following inequality holds

$$\boldsymbol{D}_0 - \boldsymbol{T}_0 \geq 0 \tag{3.38}$$

for any matrix $\boldsymbol{T}_0$ having a quasiconvex isotropic quadratic form such that

$$\boldsymbol{D} - \boldsymbol{T}_0 \geq 0. \tag{3.39}$$

Here $\boldsymbol{D}$ and $\boldsymbol{D}_0$ are the matrices associated with the functional (3.15), (3.18)-(3.20) as was discussed earlier. We get four inequalities by studying the four functionals (3.15), (3.18)-(3.20).

3.3 Construction of translation matrices using bilinear quasiaffine functions

With the exception of lower bound on the bulk modulus, the translation matrices are constructed from bilinear quasiaffine functions.

A bilinear function of two *gradients* $\boldsymbol{\zeta}^{(1)} = \nabla \boldsymbol{u}^{(1)}$ and $\boldsymbol{\zeta}^{(2)} = \nabla \boldsymbol{u}^{(2)}$ given by

$$g(\boldsymbol{\zeta}^{(1)},\ \boldsymbol{\zeta}^{(2)}) = \boldsymbol{\zeta}^{(1)} \cdot \boldsymbol{\Phi}^{\zeta\zeta} \cdot \boldsymbol{\zeta}^{(2)} \tag{3.40}$$

is quasiaffine if it possesses the property of affine functions

$$\langle g(\boldsymbol{\zeta}^{(1)},\ \boldsymbol{\zeta}^{(2)}) \rangle = g(\langle \boldsymbol{\zeta}^{(1)} \rangle,\ \langle \boldsymbol{\zeta}^{(2)} \rangle) \tag{3.41}$$

for all periodic *gradients* $\boldsymbol{\zeta}^{(1)}$ and $\boldsymbol{\zeta}^{(2)}$. Similarly, a bilinear function of two divergence-free *stress fields* $\boldsymbol{\sigma}^{(1)}$ and $\boldsymbol{\sigma}^{(2)}$ given by

$$g(\boldsymbol{\sigma}^{(1)}, \boldsymbol{\sigma}^{(2)}) = \boldsymbol{\sigma}^{(1)} \cdot \boldsymbol{\Phi}^{\sigma\sigma} \cdot \boldsymbol{\sigma}^{(2)} \tag{3.42}$$

is quasiaffine if it likewise possesses the property of affine functions

$$\langle g(\boldsymbol{\sigma}^{(1)},\ \boldsymbol{\sigma}^{(2)})\rangle = g(\langle\boldsymbol{\sigma}^{(1)}\rangle,\ \langle\boldsymbol{\sigma}^{(2)}\rangle) \tag{3.43}$$

for all periodic divergence-free *stress fields* $\boldsymbol{\sigma}^{(1)}$ and $\boldsymbol{\sigma}^{(2)}$.

Clearly if $\boldsymbol{\Phi}^{\zeta\zeta}$ or $\boldsymbol{\Phi}^{\sigma\sigma}$ is self-adjoint then such a bilinear form possesses the required properties of a translation. The associated quadratic form is then called quasiaffine. Non self-adjoint bilinear forms are useful in constructing self-adjoint translations $\boldsymbol{T}$ for functionals involving sums of several energies: we can use them in the off-diagonal blocks of the matrix $\boldsymbol{T}$ and still obtain a translation which is self-adjoint with a quasiaffine quadratic form. This will become clearer in Sections 3.4 - 3.7.

The specific bilinear and quadratic quasiaffine forms we need are well known, see e.g. Lurie and Cherkaev (1986b), Cherkaev and Gibiansky (1993). Here we follow Cherkaev and Gibiansky (1993), only mentioning those bilinear forms that we use to prove the bounds. Namely, for gradients we will use the bilinear form with matrix

$$\boldsymbol{\Phi}^{\zeta\zeta}(t_1,\ t_2) = \begin{pmatrix} t_1 & 0 & 0 & t_2 \\ 0 & -t_1 & t_2 & 0 \\ 0 & -t_2 & -t_1 & 0 \\ -t_2 & 0 & 0 & t_1 \end{pmatrix} \tag{3.44}$$

in the basis $\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3, \boldsymbol{a}_4$ (see (2.7)), and for stresses we will use the bilinear form with matrix

$$\boldsymbol{\Phi}^{\sigma\sigma}(t_1,\ t_2) = \begin{pmatrix} t_1 & 0 & 0 \\ 0 & -t_1 & t_2 \\ 0 & -t_2 & -t_1 \end{pmatrix} \tag{3.45}$$

in the basis $\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3$. Each of these bilinear form contains two parameters t_1 and t_2 which can be chosen freely. Notice that these matrices are self-adjoint when we choose $t_2 = 0$, and for values of t_2 not equal to zero we have

$$[\boldsymbol{\Phi}^{\zeta\zeta}(t_1,\ t_2)]^T = \boldsymbol{\Phi}^{\zeta\zeta}(t_1, -t_2), \qquad [\boldsymbol{\Phi}^{\sigma\sigma}(t_1,\ t_2)]^T = \boldsymbol{\Phi}^{\sigma\sigma}(t_1,\ -t_2). \tag{3.46}$$

3.4 The upper bound on the bulk modulus

Following Avellaneda and Milton (1989) and Rudelson (1989), the upper bound for the bulk modulus can be obtained by using the functional J^σ and the quasiaffine function associated with the translation matrix

$$\boldsymbol{T}^\sigma(t_1) = \boldsymbol{\Phi}^{\sigma\sigma}(t_1,\ 0). \tag{3.47}$$

This matrix is the symmetric part of the matrix $\boldsymbol{\Phi}^{\sigma\sigma}(t_1,\ t_2)$. For this functional we identify the matrices $\boldsymbol{D}$ and $\boldsymbol{D}_0$ with the compliance tensors $\boldsymbol{S}$ and $\boldsymbol{S}_0$ respectively. The bound (3.38) for an isotropic effective tensor $\boldsymbol{S}_0$ is presented by the following inequality

$$\begin{pmatrix} \frac{1}{2K_0} - t_1 & 0 & 0 \\ 0 & \frac{1}{2\mu_0} + t_1 & 0 \\ 0 & 0 & \frac{1}{2\mu_0} + t_1 \end{pmatrix} \geq 0, \tag{3.48}$$

where, according to (3.39), the parameter t_1 must satisfy the restriction

$$\begin{pmatrix} s_{11}-t_1 & s_{12} & 0 \\ s_{12} & s_{22}+t_1 & 0 \\ 0 & 0 & s_{33}+t_1 \end{pmatrix} \geq 0. \tag{3.49}$$

The inequality for bulk modulus K_0

$$\frac{1}{2K_0} - t_1 \geq 0, \tag{3.50}$$

implied by (3.48) is most restrictive when the parameter t_1 is chosen as large as possible, i.e. when it is chosen as the positive root

$$t_1 = \frac{1}{2}\left[s_{11} - s_{22} + \sqrt{(s_{11}+s_{22})^2 - 4s_{12}^2}\right] \tag{3.51}$$

of the quadratic equation

$$(s_{11}-t_1)(s_{22}+t_1) - s_{12}^2 = 0. \tag{3.52}$$

This condition follows from (3.49) treated as an equality. When t_1 is chosen in this way, one of the eigenvalues of $\boldsymbol{D}-\boldsymbol{T}$ is equal to zero, while the remaining eigenvalues are positive. By combining (3.50) and (3.51) and expressing the result in terms of the coefficients c_{ij} of the stiffness matrix we arrive at the upper bound (1.3),(1.6) on the bulk modulus of the polycrystal.

3.5 The lower bound on the bulk modulus.

The lower bound on the bulk modulus can be obtained by using the functional J^ϵ and the quasiconvex quadratic function associated with the translation matrix

$$\boldsymbol{T}^\epsilon(t_1) = \begin{pmatrix} t_1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad \text{with } t_1 \geq 0. \tag{3.53}$$

The positive semidefiniteness of this matrix ensures the quasiconvexity (and in fact the convexity) of the associated quadratic form.

For the functional J^ϵ we identify the matrices $\boldsymbol{D}$ and $\boldsymbol{D}_0$ with the elasticity tensors $\boldsymbol{C}$ and $\boldsymbol{C}_0$ respectively. The bound $\boldsymbol{C}_0 - \boldsymbol{T}^\epsilon \geq 0$ for an isotropic elasticity tensor $\boldsymbol{C}_0$ is equivalent to the matrix inequality

$$\begin{pmatrix} 2K_0-t_1 & 0 & 0 \\ 0 & 2\mu_0 & 0 \\ 0 & 0 & 2\mu_0 \end{pmatrix} \geq 0, \tag{3.54}$$

where to ensure the positive semidefiniteness of $\boldsymbol{C} - \boldsymbol{T}^{\epsilon}$ the parameter t_1 must satisfy the additional restriction that

$$\begin{pmatrix} c_{11} - t_1 & c_{12} & 0 \\ c_{12} & c_{22} & 0 \\ 0 & 0 & c_{22} \end{pmatrix} \geq 0. \tag{3.55}$$

The best possible choice of t_1 is that positive number which makes the matrix (3.55) positive semidefinite, namely

$$t_1 = \frac{c_{11}c_{22} - c_{12}^2}{c_{22}} > 0, \tag{3.56}$$

which through (3.54) gives the desired bound

$$K_0 \geq \frac{t_1}{2} = \frac{c_{11}c_{22} - c_{12}^2}{2c_{22}} = K_l. \tag{3.57}$$

This bound, in fact, coincides with the well-known harmonic mean bound by Hill (1952)

$$\boldsymbol{a}_1 : \boldsymbol{C}_0^{-1} : \boldsymbol{a}_1 \quad = \quad \frac{1}{2K_0} \quad \leq \quad \boldsymbol{a}_1 : \langle \boldsymbol{R}(\boldsymbol{x}) : \boldsymbol{C}^{-1} : \boldsymbol{R}(\boldsymbol{x}) \rangle : \boldsymbol{a}_1. \tag{3.58}$$

One can check that the expression on the right hand side (3.58) does not depend on the rotation $\boldsymbol{R}(\boldsymbol{x})$ and equals the coefficient s_{11} of the compliance matrix $\boldsymbol{S} = \boldsymbol{C}^{-1}$ of the crystal. Therefore we arrive at the bound

$$\frac{1}{2K_0} \leq s_{11} = \frac{c_{22}}{c_{11}c_{22} - c_{12}^2}, \tag{3.59}$$

which is obviously equivalent to (3.57). The coincidence of these bounds reflects the fact that the matrix $\boldsymbol{T}$ given by (3.53) does not incorporate any of the differential properties of the fields. We introduced this $\boldsymbol{T}$ to demonstrate that we can recover the harmonic mean bound (3.58) within the framework of the translation method as presented in this paper.

3.6 The upper bound on the shear modulus

To obtain the upper bound on the shear modulus of the polycrystal we bound the sum of the energies stored by the composite in two orthogonal shear-type stress fields, i.e. we bound the functional (3.19). The matrix $\boldsymbol{D}^{\sigma\sigma}$, associated with this functional has a form

$$\boldsymbol{D}^{\sigma\sigma} = \begin{pmatrix} s_{11} & s_{12} & 0 & 0 & 0 & 0 \\ s_{12} & s_{22} & 0 & 0 & 0 & 0 \\ 0 & 0 & s_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & s_{11} & s_{12} & 0 \\ 0 & 0 & 0 & s_{12} & s_{22} & 0 \\ 0 & 0 & 0 & 0 & 0 & s_{33} \end{pmatrix}. \tag{3.60}$$

We use the translation matrix

$$\boldsymbol{T}^{\sigma\sigma}(t_1,\ t_2) = \begin{pmatrix} \boldsymbol{\Phi}^{\sigma\sigma}(-t_1, 0) & \boldsymbol{\Phi}^{\sigma\sigma}(0, -t_2) \\ \boldsymbol{\Phi}^{\sigma\sigma}(0, t_2) & \boldsymbol{\Phi}^{\sigma\sigma}(-t_1, 0) \end{pmatrix} \tag{3.61}$$

which depends on the two parameters t_1 and t_2. Upon making the substitution (3.45) we see that $\boldsymbol{T}^{\sigma\sigma}$ is represented by the six by six matrix

$$\boldsymbol{T}^{\sigma\sigma}(t_1,\ t_2) = \begin{pmatrix} -t_1 & 0 & 0 & 0 & 0 & 0 \\ 0 & t_1 & 0 & 0 & 0 & -t_2 \\ 0 & 0 & t_1 & 0 & t_2 & 0 \\ 0 & 0 & 0 & -t_1 & 0 & 0 \\ 0 & 0 & t_2 & 0 & t_1 & 0 \\ 0 & -t_2 & 0 & 0 & 0 & t_1 \end{pmatrix}. \tag{3.62}$$

The bounds come from the inequality

$$\boldsymbol{D}_0^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}(t_1,\ t_2) \geq 0, \tag{3.63}$$

which is valid for any choice of the parameters $t_1,\ t_2$ satisfying the equation

$$\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}(t_1,\ t_2) \geq 0. \tag{3.64}$$

Here the diagonal matrix $\boldsymbol{D}_0^{\sigma\sigma}$ is defined as

$$\boldsymbol{D}_0^{\sigma\sigma} = \begin{pmatrix} \boldsymbol{S}_0 & 0 \\ 0 & \boldsymbol{S}_0 \end{pmatrix}, \tag{3.65}$$

and is the "direct sum" of two copies of effective compliance tensor $\boldsymbol{S}_0$ of a polycrystal.

Let us find the critical values of the parameters $t_1,\ t_2$ which make the bound (3.63) the most restrictive. We mention first, that each of the matrices $\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}$ and $\boldsymbol{D}_0^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}$ is the direct sum of two blocks

$$\begin{aligned} \boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma} &= \boldsymbol{A}^{1,\ 2,\ 6} \oplus \boldsymbol{A}^{3,\ 4,\ 5}, \\ \boldsymbol{D}_0^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma} &= \boldsymbol{A}_0^{1,\ 2,\ 6} \oplus \boldsymbol{A}_0^{3,\ 4,\ 5}, \end{aligned} \tag{3.66}$$

The matrix $\boldsymbol{A}^{a,b,c}$ here and below denotes a diagonal minor of the matrix $\boldsymbol{D} - \boldsymbol{T}$; it is the 3×3 matrix which remains when one strikes out from $\boldsymbol{D} - \boldsymbol{T}$ those columns and rows with numbers not equal to a, b, or c.

The matrices on the right hand side (3.66) have the following form:

$$\boldsymbol{A}^{1,\ 2,\ 6} = \begin{pmatrix} s_{11} + t_1 & s_{12} & 0 \\ s_{12} & s_{22} - t_1 & t_2 \\ 0 & t_2 & s_{33} - t_1 \end{pmatrix}, \tag{3.67}$$

$$\boldsymbol{A}_0^{1,\ 2,\ 6} = \begin{pmatrix} \frac{1}{2K_0} + t_1 & 0 & 0 \\ 0 & \frac{1}{2\mu_0} - t_1 & t_2 \\ 0 & t_2 & \frac{1}{2\mu_0} - t_1 \end{pmatrix}, \tag{3.68}$$

$$\boldsymbol{A}^{3,\ 4,\ 5} = \begin{pmatrix} s_{33} - t_1 & 0 & -t_2 \\ 0 & s_{11} + t_1 & s_{12} \\ -t_2 & s_{12} & s_{22} - t_1 \end{pmatrix}, \tag{3.69}$$

$$\boldsymbol{A}_0^{3,\ 4,\ 5} = \begin{pmatrix} \frac{1}{2\mu_0} - t_1 & 0 & -t_2 \\ 0 & \frac{1}{2K_0} + t_1 & 0 \\ -t_2 & 0 & \frac{1}{2\mu_0} - t_1 \end{pmatrix}. \tag{3.70}$$

One can easily see that the pair of matrices $\boldsymbol{A}^{1,\ 2,\ 6}$ and $\boldsymbol{A}^{3,\ 4,\ 5}$ are both simultaneously positive semidefinite whenever one of the matrices in the pair is positive semidefinite. The same is true for the matrices $\boldsymbol{A}_0^{1,\ 2,\ 6}$ and $\boldsymbol{A}_0^{3,\ 4,\ 5}$. Hence it suffices to analyze only one matrix out of each pair.

Now the bounds (3.63) imply $\boldsymbol{A}_0^{1,\ 2,\ 6}$ is positive definite, or equivalently that

$$\frac{1}{2K_0} + t_1 \geq 0, \qquad \frac{1}{2\mu_0} - t_1 - t_2 \geq 0, \qquad \frac{1}{2\mu_0} - t_1 + t_2 \geq 0, \tag{3.71}$$

and the restrictions (3.64) are satisfied when the leading diagonal subdeterminants of the matrix $\boldsymbol{A}^{1,\ 2,\ 6}$ are non-negative, i.e. when

$$\begin{gathered} s_{11} + t_1 \geq 0, \\ (s_{11} + t_1)(s_{22} - t_1) - s_{12}^2 \geq 0, \\ f(t_1, t_2) = \det(\boldsymbol{A}^{1,\ 2,\ 6}) = (s_{33} - t_1)[(s_{11} + t_1)(s_{22} - t_1) - s_{12}^2] - (s_{11} + t_1)\ t_2^2 \geq 0, \end{gathered} \tag{3.72}$$

These restrictions define a region $\mathcal{T}$ of the plane comprised of all points $(t_1,\ t_2)$ compatible with the inequalities (3.72). The matrix $\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}$ is positive-semidefinite if and only if $(t_1,\ t_2)$ is in the set $\mathcal{T}$. In particular, the positive definiteness of $\boldsymbol{D}^{\sigma\sigma}$ implies that $\mathcal{T}$ contains the origin $t_1 = t_2 = 0$. Now let us introduce polar coordinates, setting

$$\boldsymbol{t} = (t_1, t_2) = (r\cos\theta, r\sin\theta). \tag{3.73}$$

By rewriting the inequality $\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}(t_1, t_2) \geq 0$ in the form

$$\frac{1}{r}\boldsymbol{I} \geq (\boldsymbol{D}^{\sigma\sigma})^{-1/2}\boldsymbol{T}^{\sigma\sigma}(\cos\theta, \sin\theta)(\boldsymbol{D}^{\sigma\sigma})^{-1/2} \tag{3.74}$$

it is clear that $\boldsymbol{t}$ is in $\mathcal{T}$ if and only if $1/r$ is greater than or equal to the largest eigenvalue $\lambda^+(\theta)$ of the matrix $(\boldsymbol{D}^{\sigma\sigma})^{-1/2}\boldsymbol{T}^{\sigma\sigma}(\cos\theta, \sin\theta)(\boldsymbol{D}^{\sigma\sigma})^{-1/2}$. Notice that $\lambda^+(\theta)$ is necessarily positive because the quasiconvexity property implies that $\boldsymbol{T}^{\sigma\sigma}(\cos\theta, \sin\theta)$ has at least one non-negative eigenvalue. For values of r less than $1/\lambda^+(\theta)$ the matrix $\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}$ is strictly positive definite and $f(t_1, t_2)$ is strictly positive. For $r = 1/\lambda^+(\theta)$ the matrix $\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}$ has a zero eigenvalue and $f(t_1, t_2) = 0$.

By combining these facts we get a simple prescription for finding $\mathcal{T}$: cut the plane along all curves where $f(t_1, t_2) = 0$ and pick the piece containing the origin. This set $\mathcal{T}$ is necessarily convex. Indeed, if $\boldsymbol{t} = (t_1, t_2)$ and $\boldsymbol{t}' = (t_1', t_2')$ are two points in $\mathcal{T}$ then

$$\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}(t_1, t_2) \geq 0 \ \text{ and } \ \boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}(t_1', t_2') \geq 0, \tag{3.75}$$

and by taking a weighted sum of these equations we see that

$$\boldsymbol{D}^{\sigma\sigma} - \boldsymbol{T}^{\sigma\sigma}(pt_1 + (1-p)t_1', pt_2 + (1-p)t_2') \geq 0 \quad \forall p \in [0,1], \tag{3.76}$$

implying that $p\boldsymbol{t} + (1-p)\boldsymbol{t}' \in \mathcal{T}$.

The bounds (3.71) imply

$$\frac{1}{2\mu_0} \geq \max_{(t_1,\ t_2)\in\mathcal{T}} (t_1 + t_2). \tag{3.77}$$

The solution to this maximization problem leads to the expression for the upper bound given by (1.10). These calculations and a more complete discussion are placed in Appendix A.

3.7 The lower bound on the shear modulus

To obtain a lower bound on the shear modulus of the polycrystal, we bound the sum of the energies stored by the composite in two orthogonal shear-type strain fields, i.e. we bound the functional (3.20). The matrix $\boldsymbol{D}^{\zeta\zeta}$ associated with this functional has a form

$$\boldsymbol{D}^{\zeta\zeta} = \begin{pmatrix} c_{11} & c_{12} & 0 & 0 & 0 & 0 & 0 & 0 \\ c_{12} & c_{22} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & c_{33} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{11} & c_{12} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{12} & c_{22} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & c_{33} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}. \tag{3.78}$$

We use the translation matrix

$$\boldsymbol{T}^{\zeta\zeta}(t_1,\ t_2) = \begin{pmatrix} \boldsymbol{\Phi}^{\zeta\zeta}(-t_1, 0) & \boldsymbol{\Phi}^{\zeta\zeta}(0, -t_2) \\ \boldsymbol{\Phi}^{\zeta\zeta}(0,\ t_2) & \boldsymbol{\Phi}^{\zeta\zeta}(-t_1, 0) \end{pmatrix}, \tag{3.79}$$

which depends upon the two parameters t_1 and t_2. By making the substitution (3.44) we see that $\boldsymbol{T}^{\zeta,\zeta}$ is represented by the 8×8 matrix

$$\boldsymbol{T}^{\zeta\zeta}(t_1,\ t_2) = \begin{pmatrix} -t_1 & 0 & 0 & 0 & 0 & 0 & 0 & -t_2 \\ 0 & t_1 & 0 & 0 & 0 & 0 & -t_2 & 0 \\ 0 & 0 & t_1 & 0 & 0 & t_2 & 0 & 0 \\ 0 & 0 & 0 & -t_1 & t_2 & 0 & 0 & 0 \\ 0 & 0 & 0 & t_2 & -t_1 & 0 & 0 & 0 \\ 0 & 0 & t_2 & 0 & 0 & t_1 & 0 & 0 \\ 0 & -t_2 & 0 & 0 & 0 & 0 & t_1 & 0 \\ -t_2 & 0 & 0 & 0 & 0 & 0 & 0 & -t_1 \end{pmatrix}. \tag{3.80}$$

The bounds come from the inequality

$$\boldsymbol{D}_0^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta}(t_1,\ t_2) \geq 0, \tag{3.81}$$

which holds for any choice of the parameters $t_1,\ t_2$ such that

$$\boldsymbol{D}^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta}(t_1,\ t_2) \geq 0. \tag{3.82}$$

Here the diagonal matrix $\boldsymbol{D}_0^{\zeta\zeta}$ is defined as

$$\boldsymbol{D}_0^{\zeta\zeta} = \begin{pmatrix} \boldsymbol{C}'_0 & 0 \\ 0 & \boldsymbol{C}'_0 \end{pmatrix}. \tag{3.83}$$

It is the direct of sum of two copies of the effective stiffness tensor $\boldsymbol{C}'_0$ of the polycrystal, each represented in the form (3.17) of a four by four matrix.

The next step is to find the values of the parameters $t_1,\ t_2$ which give the most restrictive bounds in (3.81). Note that each of the two matrices $\boldsymbol{D}^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta}$ and $\boldsymbol{D}_0^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta}$ is the direct sum of two blocks

$$\boldsymbol{D}^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta} = \boldsymbol{A}^{1,\ 2,\ 7,\ 8} \oplus \boldsymbol{A}^{3,\ 4,\ 5,\ 6}, \qquad \boldsymbol{D}_0^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta} = \boldsymbol{A}_0^{1,\ 2,\ 7,\ 8} \oplus \boldsymbol{A}_0^{3,\ 4,\ 5,\ 6}. \tag{3.84}$$

(Recall that the matrix $\boldsymbol{A}^{a,b,c,d}$ denotes a diagonal minor of the matrix $\boldsymbol{D} - \boldsymbol{T}$; it is the four by four matrix which remains when one strikes out from $\boldsymbol{D} - \boldsymbol{T}$ those columns and rows with numbers not equal to a, b, c or d.)

The matrices in the right hand sides of (3.84) have the following form:

$$\boldsymbol{A}^{1,\ 2,\ 7,\ 8} = \begin{pmatrix} c_{11}+t_1 & c_{12} & 0 & t_2 \\ c_{12} & c_{22}-t_1 & t_2 & 0 \\ 0 & t_2 & c_{33}-t_1 & 0 \\ t_2 & 0 & 0 & t_1 \end{pmatrix}, \tag{3.85}$$

$$\boldsymbol{A}_0^{1,\ 2,\ 7,\ 8} = \begin{pmatrix} 2K_0+t_1 & 0 & 0 & t_2 \\ 0 & 2\mu_0-t_1 & t_2 & 0 \\ 0 & t_2 & 2\mu_0-t_1 & 0 \\ t_2 & 0 & 0 & t_1 \end{pmatrix}, \tag{3.86}$$

$$\boldsymbol{A}^{3,\ 4,\ 5,\ 6} = \begin{pmatrix} c_{33}-t_1 & 0 & 0 & -t_2 \\ 0 & t_1 & -t_2 & 0 \\ 0 & -t_2 & c_{11}+t_1 & c_{12} \\ -t_2 & 0 & c_{12} & c_{22}-t_1 \end{pmatrix}, \tag{3.87}$$

$$\boldsymbol{A}_0^{3,\ 4,\ 5,\ 6} = \begin{pmatrix} 2\mu_0-t_1 & 0 & 0 & -t_2 \\ 0 & t_1 & -t_2 & 0 \\ 0 & -t_2 & 2K_0+t_1 & 0 \\ -t_2 & 0 & 0 & 2\mu_0-t_1 \end{pmatrix}. \tag{3.88}$$

One can easily see that the pair of matrices $\boldsymbol{A}^{1,\ 2,\ 7,\ 8}$ and $\boldsymbol{A}^{3,\ 4,\ 5,\ 6}$ are simultaneously positive semidefinite when one matrix in the pair is positive semidefinite. The same is true for the pair of matrices $\boldsymbol{A}_0^{1,\ 2,\ 7,\ 8}$ and $\boldsymbol{A}_0^{3,\ 4,\ 5,\ 6}$. Hence, it suffices to analyze only one matrix out of each pair.

The bounds (3.81) and the restrictions (3.82) imply $\boldsymbol{A}_0^{1,\ 2,\ 7,\ 8}$ is positive definite, or equivalently that

$$t_1 \geq 0, \quad (2K_0 + t_1)t_1 - t_2^2 \geq 0, \quad 2\mu_0 - t_1 - t_2 \geq 0, \quad 2\mu_0 - t_1 + t_2 \geq 0, \tag{3.89}$$

when (t_1, t_2) is in a region $\mathcal{T}$, now obtained by cutting the plane along all curves where

$$\begin{aligned} f(t_1, t_2) &= \det(\boldsymbol{A}^{1,\ 2,\ 7,\ 8}) \\ &= [(c_{33} - t_1)(c_{22} - t_1) - t_2^2][t_1(c_{11} + t_1) - t_2^2] - t_1 c_{12}^2 (c_{33} - t_1) = 0, \end{aligned} \tag{3.90}$$

and picking the piece containing the origin. The analysis of the previous section shows that $\boldsymbol{D}^{\zeta\zeta} - \boldsymbol{T}^{\zeta\zeta}$ is positive-definite or positive-semidefinite if and only if $(t_1,\ t_2)$ is in this set $\mathcal{T}$.

¿From the bounds (3.89) we see that

$$2\mu_0 \geq \max_{(t_1,\ t_2) \in \mathcal{T}} (t_1 + t_2). \tag{3.91}$$

The solution of the maximization problem (3.91) provides the lower bound (1.4), (1.7) on the effective shear modulus. The proof of this is a straightforward calculation that we relegate to Appendix B.

4 Attainability of the bounds: description of the optimal microstructures

4.1 Extreme points

The bounds (1.3)-(1.4) form a rectangle $ABCD$ in the plane of effective constants $(K_0,\ \mu_0)$. To prove the attainability of each point inside this rectangle, it is enough to exhibit microstructures with isotropic effective tensors that correspond to the corner points $A = (K_l, \mu^+)$, $B = (K_u, \mu^+)$, $C = (K_u, \mu^-)$, and $D = (K_l, \mu^-)$ (see Figure 1).

To prove this point, consider the class of isotropic composites assembled from two given isotropic materials sharing a common bulk modulus $K_1 = K_2$ but having different shear moduli μ_1 and μ_2. It is well-known (Hill, 1963) that all such composites will have the same bulk modulus, i.e.,$K_0 = K_1 = K_2$, and will have a shear modulus μ_0 that can lie anywhere in the interval $[\mu_1, \mu_2]$. Similarly (Hill, 1964), composites assembled from isotropic materials with a common shear modulus but having different bulk moduli, are such that $\mu_0 = \mu_1 = \mu_2$ and have a bulk modulus K_0 that can range anywhere in the interval $[K_1, K_2]$. In particular, if we start with the isotropic polycrystals A, with moduli (K_l, μ^+), and B, with moduli (K_u, μ^+), which share a common shear modulus

μ^+, one can assemble isotropic materials filling the interval AB. These materials possess the moduli

$$K_{AB} \in [K_l, K_u], \quad \mu_{AB} = \mu^+ . \tag{4.1}$$

One obtains in the same way isotropic composites which fill the interval CD by mixing two isotropic polycrystals C, with moduli (K_u, μ^-), and D, with moduli (K_l, μ^-). These composites possess moduli

$$K_{CD} \in [K_l, K_u], \quad \mu_{CD} = \mu^- . \tag{4.2}$$

Then, one can choose a pair of materials, one along the interval AB and the other along the interval CD, so that they share the same bulk modulus ($K_{AB} = K_{CD}$) and mix them. This generates all the materials in the interval

$$K_0 = K_{AB}, \quad \mu_0 \in [\mu^-,\ \mu^+] = [\mu_l,\ \mu_u] . \tag{4.3}$$

Finally, upon recalling that K_{AB} can be chosen to lie anywhere in the interval $[K_l, K_u]$, we conclude that all points within the rectangle can be achieved by a mixture of the materials A, B, C, D, see Figure 1.

In the following sections we will demonstrate that the corner points are the effective tensors of some special elastically isotropic microstructures. We will use laminate microstructures because they are natural candidates for optimal microstructures and because formulas are available giving their effective elasticity tensors.

4.2 Microstructures that correspond to the corner points B and C

The corner points B and C can be realized using a laminate composite material of infinite rank. Such structures were studied in many papers (Norris, 1985; Milton, 1986; Avellaneda, Cherkaev, Lurie, Milton 1988) and are known to correspond to the self-consistent scheme (Milton, 1986).

To describe the construction we start with the relation

$$\boldsymbol{S}_0 = f_a \boldsymbol{S}_a + f_b \boldsymbol{S}_b - f_a f_b \frac{(\boldsymbol{S}_b - \boldsymbol{S}_a) : \boldsymbol{t}_0\boldsymbol{t}_0\boldsymbol{t}_0\boldsymbol{t}_0 : (\boldsymbol{S}_b - \boldsymbol{S}_a)}{\boldsymbol{t}_0\boldsymbol{t}_0 : (f_b \boldsymbol{S}_a + f_a \boldsymbol{S}_b) : \boldsymbol{t}_0\boldsymbol{t}_0}, \tag{4.4}$$

giving the effective compliance tensor $\boldsymbol{S}_0$ of a laminate combined from two materials with compliance tensors $\boldsymbol{S}_a$ and $\boldsymbol{S}_b$ mixed in proportions f_a and f_b with $\boldsymbol{t}_0$ being a unit vector tangential to the layers. Suppose now that the volume fraction f_b of the second material tends to zero. Then the effective tensor $\boldsymbol{S}_0$ will differ only slightly from $\boldsymbol{S}_a$, and we can regard (4.4) as defining a differential process. Specifically, the relation (4.4) leads to the differential equation

$$\frac{d\boldsymbol{S}_0}{df} = (\boldsymbol{S}_b - \boldsymbol{S}_0) - \frac{(\boldsymbol{S}_b - \boldsymbol{S}_0) : \boldsymbol{t}_0\boldsymbol{t}_0\boldsymbol{t}_0\boldsymbol{t}_0 : (\boldsymbol{S}_b - \boldsymbol{S}_0)}{\boldsymbol{t}_0\boldsymbol{t}_0 : \boldsymbol{S}_b : \boldsymbol{t}_0\boldsymbol{t}_0} \tag{4.5}$$

which describes the evolution of the effective tensor $\boldsymbol{S}_0$ (now taking the role of $\boldsymbol{S}_a$) when we add a portion df of the material $\boldsymbol{S}_b(f)$ to it. Here f parameterizes the evolution: as f changes we are free to select different materials to add to the composite, these materials being chosen from the trajectory $\boldsymbol{S}_b(f)$ in matrix space. At each stage in this differential process the material $\boldsymbol{S}_b(f)$ should be layered with the composite, with the layers being tangential to $\boldsymbol{t}_0 = \boldsymbol{t}_0(f)$ (which may also depend upon f) and with the layer width being much larger than the existing layer widths in the composite, (to allow us to treat the composite as a homogeneous medium with compliance tensor $\boldsymbol{S}_0$ when calculating the effective tensor at the next stage).

Let us now consider the following process of building a polycrystal. We suppose some isotropic seed material with tensor $\boldsymbol{S}_0$ has been given. Here $\boldsymbol{S}_0$ should not yet be thought of as an effective tensor, rather it should be regarded as the starting material for the differential scheme described in the preceding paragraph. The only constraint on $\boldsymbol{S}_0$ besides isotropy is that it should have bulk and shear moduli which are non-zero, positive, and finite.

We want to add an infinitesimal proportion df of the given crystal to this seed material, layering it equal proportions $df/4$ in four different directions $\boldsymbol{t}_1$, $\boldsymbol{t}_2$, $\boldsymbol{t}_3$ and $\boldsymbol{t}_4$ each 45° apart, with the crystal in each layer oriented so that the layering direction is a principle axis of the crystal in that layer.

First we add the tensor $\boldsymbol{S}_1 = \boldsymbol{S} = \boldsymbol{C}^{-1}$ (i.e. the original crystal),

$$\boldsymbol{S}_1 = \begin{pmatrix} s_{11} & s_{12} & 0 \\ s_{12} & s_{22} & 0 \\ 0 & 0 & s_{33} \end{pmatrix}, \tag{4.6}$$

in layers with tangential vector $\boldsymbol{t}_1 = \boldsymbol{i}$.

Then, we add the tensor $\boldsymbol{S}_2 = \boldsymbol{R}(\pi/4) : \boldsymbol{S} : \boldsymbol{R}^T(\pi/4)$ (i.e. the crystal rotated by the angle $\pi/4$),

$$\boldsymbol{S}_2 = \begin{pmatrix} s_{11} & 0 & s_{12} \\ 0 & s_{33} & 0 \\ s_{12} & 0 & s_{22} \end{pmatrix}, \tag{4.7}$$

in layers that are rotated by the same angle with tangential vector $\boldsymbol{t}_2 = (\boldsymbol{i} + \boldsymbol{j})/\sqrt{2}$.

Then, we add the tensor $\boldsymbol{S}_3 = \boldsymbol{R}(\pi/2) : \boldsymbol{S} : \boldsymbol{R}^T(\pi/2)$ (i.e. the original crystal rotated by the angle $\pi/2$),

$$\boldsymbol{S}_3 = \begin{pmatrix} s_{11} & -s_{12} & 0 \\ -s_{12} & s_{22} & 0 \\ 0 & 0 & s_{33} \end{pmatrix}, \tag{4.8}$$

in layers with tangential vector $\boldsymbol{t}_3 = \boldsymbol{j}$.

Finally, we add the crystal $\boldsymbol{S}_4 = \boldsymbol{R}(3\pi/4) : \boldsymbol{S} : \boldsymbol{R}^T(3\pi/4)$ (i.e. original crystal rotated by the angle $3\pi/4$),

$$\boldsymbol{S}_4 = \begin{pmatrix} s_{11} & 0 & -s_{12} \\ 0 & s_{33} & 0 \\ -s_{12} & 0 & s_{22} \end{pmatrix}, \tag{4.9}$$

in layers that are rotated by the same angle with the tangential vector $\boldsymbol{t}_4 = (-\boldsymbol{i}+\boldsymbol{j})/\sqrt{2}$.

The differential equation describing this process has the form

$$\frac{d\boldsymbol{S}_0}{df} = \frac{1}{4}\sum_{i=1}^{4}\left[\boldsymbol{S}_i - \boldsymbol{S}_0 - \frac{(\boldsymbol{S}_i - \boldsymbol{S}_0) : \boldsymbol{t}_i\boldsymbol{t}_i\boldsymbol{t}_i\boldsymbol{t}_i : (\boldsymbol{S}_i - \boldsymbol{S}_0)}{\boldsymbol{t}_i\boldsymbol{t}_i : \boldsymbol{S}_i : \boldsymbol{t}_i\boldsymbol{t}_i}\right]. \tag{4.10}$$

The isotropy of the original seed tensor $\boldsymbol{S}_0$ and the eightfold symmetry of the differential process ensures that $\boldsymbol{S}_0(f)$ remains isotropic for all f. We iterate this process until the seed material occupies an infinitesimal portion of the composite. Continuing the process further only diminishes the already infinitesimal portion of the seed material, which by that stage can have little influence on the overall effective properties of the composite. In other words the differential process must converge to a unique fixed point $\boldsymbol{S}_0$, defined by the relation

$$\sum_{i=1}^{4}\left[\boldsymbol{S}_i - \boldsymbol{S}_0 - \frac{(\boldsymbol{S}_i - \boldsymbol{S}_0) : \boldsymbol{t}_i\boldsymbol{t}_i\boldsymbol{t}_i\boldsymbol{t}_i : (\boldsymbol{S}_i - \boldsymbol{S}_0)}{\boldsymbol{t}_i\boldsymbol{t}_i : \boldsymbol{S}_i : \boldsymbol{t}_i\boldsymbol{t}_i}\right] = 0. \tag{4.11}$$

At this stage we can remove the seed material entirely, since it occupies an infinitesimal volume fraction, replacing it with pure crystal. In other words, we can regard the positive definite tensor $\boldsymbol{S}_0$ which solves (4.11) as being the effective tensor of an infinite rank polycrystal. For a rigorous proof of this see Milton (1986). As already mentioned, the tensor on the left hand side of (4.11) is isotropic due to the eight-fold symmetry of the construction procedure and due to the isotropy of the tensor

$$\boldsymbol{S}_0 = \frac{1}{2K_0}\boldsymbol{a}_1\boldsymbol{a}_1 + \frac{1}{2\mu_0}(\boldsymbol{a}_2\boldsymbol{a}_2 + \boldsymbol{a}_3\boldsymbol{a}_3). \tag{4.12}$$

Therefore it suffices to take the trace of (4.11), and to take its contraction on the left and right with respect to the isotropic basis vector $\boldsymbol{a}_1$. In this way we obtain two scalar equations for the bulk and shear moduli of the polycrystal, namely,

$$(s_{11} - 1/2K_0)(s_{11} + s_{22} - 2s_{12}) = (s_{11} - s_{12} - 1/2K_0)^2, \tag{4.13}$$

$$(s_{22} + s_{33} - 1/\mu_0)(s_{11} + s_{22} - 2s_{12}) = (s_{12} - s_{22} + 1/2\mu_0)^2, \tag{4.14}$$

(see Appendix C). These equations have a unique solution with $K_0 \geq 0$ and $\mu_0 \geq 0$, given by

$$\begin{aligned} \frac{1}{K_0} &= s_{11} - s_{22} + \sqrt{(s_{11} + s_{22})^2 - 4s_{12}^2}, \\ \frac{1}{\mu_0} &= 2\Big(- s_{11} + s_{12} + \sqrt{(s_{11} + s_{33})(s_{11} + s_{22} - 2s_{12})}\ \Big). \end{aligned} \tag{4.15}$$

One can check (by substituting the expressions for the compliance matrix coefficients in terms of the stiffness matrix coefficients) that the effective bulk and shear moduli of this polycrystal are equal to

$$K_0 = K_u, \quad \mu_0 = \mu^-, \tag{4.16}$$

i.e. correspond to the point C, see (1.3)-(1.10) and Fig.1.

To get the remaining point B we consider a similar construction but with the crystals in the laminates rotated by 90°. This means that instead of the tensor $\boldsymbol{S}$ we now work with the tensor $\boldsymbol{S}'$:

$$\boldsymbol{S}' = \begin{pmatrix} s_{11} & -s_{12} & 0 \\ -s_{12} & s_{22} & 0 \\ 0 & 0 & s_{33} \end{pmatrix}. \tag{4.17}$$

Notice that the crystal in each layer is still oriented so that the layering direction is a principle axis of the crystal in that layer. By comparing the tensors $\boldsymbol{S}'$ and $\boldsymbol{S}$ it is clear that all the previous formulas apply but with s_{12} replaced by $-s_{12}$. In particular, the effective tensor $\boldsymbol{S}_0'$ of the structure has a bulk modulus K_0' and a shear modulus μ_0' given by

$$\begin{aligned} \frac{1}{K_0'} &= s_{11} - s_{22} + \sqrt{(s_{11}+s_{22})^2 - 4s_{12}^2}, \\ \frac{1}{\mu_0'} &= 2\Big(- s_{11} - s_{12} + \sqrt{(s_{11}+s_{33})(s_{11}+s_{22}+2s_{12})}\ \Big), \end{aligned} \tag{4.18}$$

i.e. they correspond to the point B :

$$K_0 = K_u, \qquad \mu_0 = \mu^+, \tag{4.19}$$

see (1.3)-(1.10) and Fig.1. In summary, we have found the microstructures that correspond to both corner points B and C of the rectangular box defined by the bounds.

4.3 Microstructures that correspond to the corner points A and D

In this section we describe two possible types of microstructures corresponding to the points A and D of the bound rectangle. The first is a laminate composite of infinite rank. The second is a composite of second rank. Either of the two constructions realizes the bounds associated with the points A and D. We start with the infinite rank procedure because it seems to be more naturally motivated.

4.3.1 Laminates of infinite rank

We want to build a sequence of orthotropic materials which tend to an isotropic material with bulk and shear moduli corresponding to the extremal point A (or to the extremal point D). First let us specify the basic step of the procedure. Given an arbitrary orthotropic single crystal, there corresponds a rectangle $ABCD$ defining the bounds on K_0 and μ_0 for the associated isotropic polycrystal. Clearly, if we construct by homogenization an orthotropic effective tensor $\boldsymbol{S}^{(1)}$ starting from $\boldsymbol{S}$, it will have a corresponding rectangle $A_1B_1C_1D_1$. Because $\boldsymbol{S}^{(1)}$ is in the G-closure associated with $\boldsymbol{S}$, the new rectangle must lie inside $ABCD$. The idea of the proof of realizability of the bound associated with the point A is to construct, starting from $\boldsymbol{S}$, a sequence of orthotropic effective tensors $\boldsymbol{S}^{(n)}$, $n = 1, 2, 3, \ldots$, such that $A_n = A$ for all n and such that that the tensors in the sequence become progressively less anisotropic tending to an isotropic effective medium as n tends to infinity. This medium will then be such that $(K_0, \mu^+) = A$. The associated rectangles $A_n B_n C_n D_n$ form a nested sequence with common vertex A.

To construct $\boldsymbol{S}^{(1)}$ we use a pair of materials $\boldsymbol{S}^+$ and $\boldsymbol{S}^-$ obtained by rotating $\boldsymbol{S}$ by the angles ϕ and $-\phi$ respectively,

$$\boldsymbol{S}^+ = \boldsymbol{R}(\phi) : \boldsymbol{S} : \boldsymbol{R}^T(\phi), \qquad \boldsymbol{S}^- = \boldsymbol{R}(-\phi) : \boldsymbol{S} : \boldsymbol{R}^T(-\phi), \tag{4.20}$$

and we laminate them in equal proportions (i.e. with $f_1 = f_2 = 1/2$), with the layers being tangential to the vector $\boldsymbol{i}$. The resulting effective tensor $\boldsymbol{S}^{(1)}$ of this laminate is orthotropic tensor with coefficients s'_{ij} given by the relations

$$\begin{gathered} s'_{11} = s_{11}, \quad s'_{12} = s_{12}\cos 2\phi, \quad s'_{22} = s_{22} + (s_{33} - s_{22})(\sin 2\phi)^2, \quad s_{13} = s_{23} = 0, \\ s'_{33} = s_{33} + (s_{22} - s_{33})(\sin 2\phi)^2 - \frac{[s_{12} + (s_{22} - s_{33})\cos 2\phi]^2 (\sin 2\phi)^2}{s_{11} + s_{22}(\cos 2\phi)^2 + s_{33}(\sin 2\phi)^2 + 2s_{12}\cos 2\phi}. \end{gathered} \tag{4.21}$$

(see (4.4)). By using the results of the previous sections one can calculate the bounds on the effective bulk and shear moduli of any isotropic polycrystal assembled from this new orthotropic material. The resulting lower bound on the bulk modulus, $K_l = 1/(2s'_{11})$, obviously coincides with the corresponding bound, $K_l = 1/(2s_{11})$, associated with the original crystal (see (3.59)). We would like to choose the angle ϕ in such a way as to preserve one of the shear modulus bounds, namely μ^+. Let us choose the angle so that

$$\begin{gathered} -s_{11} - s_{12} + \sqrt{(s_{11} + s_{33})(s_{11} + s_{22} + 2s_{12})} = \\ -s'_{11} + s'_{12} + \sqrt{(s'_{11} + s'_{33})(s'_{11} + s'_{22} - 2s'_{12})} = \frac{1}{2\mu^+} \end{gathered} \tag{4.22}$$

If this equation has a solution, it will guarantee that the rectangle $A_1 B_1 C_1 D_1$ associated with $\boldsymbol{S}^{(1)}$ has the same corner $A_1 = A$ as the rectangle $ABCD$ associated with $\boldsymbol{S}$. A calculation shows that the equation (4.22) has the solution

$$\cos 2\phi = c_* = \frac{-s_{11} - s_{33} + \sqrt{(s_{11} + s_{33})(s_{11} + s_{22} + 2s_{12})}}{s_{22} - s_{33} + 2s_{12}}. \tag{4.23}$$

Because the value c_* should be the cosine of some angle, we still need to check that $|c_*| \leq 1$. Indeed, one can see that

$$c_* = \frac{\sqrt{pq} - p}{q - p} = \frac{\sqrt{p}}{\sqrt{q} + \sqrt{p}}, \tag{4.24}$$

where

$$p = s_{11} + s_{33} > 0, \quad q = s_{11} + s_{22} + 2s_{12} > 0 \tag{4.25}$$

are both strictly positive for any positive definite matrix $\boldsymbol{S}$. Hence,

$$0 < c_* < 1. \tag{4.26}$$

Therefore, we can construct an orthotropic material from the original crystal in such a way that "preserves" the corner point A. Notice that (4.23) has two solutions for ϕ, differing by 180°. This non-uniqueness reflects the fact that elasticity tensors are invariant under 180° rotations. Thus there is no intrinsic difference between the two solutions.

Alternatively we could choose the angle ϕ so that

$$\begin{aligned} -s_{11} + s_{12} + \sqrt{(s_{11}+s_{33})(s_{11}+s_{22}-2s_{12})} &= \\ -s'_{11} - s'_{12} + \sqrt{(s'_{11}+s'_{33})(s'_{11}+s'_{22}+2s'_{12})} &= \frac{1}{2\mu^-}. \end{aligned} \tag{4.27}$$

Calculations show that this has the solution

$$\cos 2\phi = c_{**} = \frac{-s_{11} - s_{33} + \sqrt{(s_{11}+s_{33})(s_{11}+s_{22}-2s_{12})}}{s_{22} - s_{33} - 2s_{12}}, \tag{4.28}$$

which in turn provides a solution for ϕ because the positive definiteness of $\boldsymbol{S}$ implies

$$0 < c_{**} < 1. \tag{4.29}$$

Therefore we can construct an orthotropic material from the original crystal in such way that "preserves" the other corner point $D = (K_l, \mu^-)$.

We claim that by repeating this process (at the nth stage laminating $\boldsymbol{S}^{(n-1)}$ in two different orientations to obtain an orthotropic material $\boldsymbol{S}^{(n)}$) we end up with an isotropic material. Indeed, the absolute values of both c_* and c_{**} are strictly less than one and a consequence we see from the relations (4.21) that the absolute value of s_{12} must decrease, unless it is already zero. After infinite number of steps we end up with a material which has a compliance tensor $\boldsymbol{S}_0$ with at least square symmetry, i.e. with $s_{12} = 0$. For such a square symmetric material the bound rectangle collapses to a single point, and by construction this point coincides with either the point A or D. In other words, any isotropic material built from this square symmetric material has bulk and shear moduli corresponding to point A or to point D, according to which corner point we decide to preserve in the construction procedure.

4.3.2 Laminates of second rank

We propose yet another construction which attains these bounds, specifically, there exist elastically isotropic *laminates of second rank* with moduli that correspond to the corner points A and D. Here we only describe these materials. The explicit calculations are contained in Appendix D.

To motivate the construction, consider the laminate $\boldsymbol{S}^{(1)}$ that we constructed by laminating $\boldsymbol{S}^{+}$ and $\boldsymbol{S}^{-}$ in equal proportions. Clearly there must be something special about this microstructure for the associated lower bulk modulus bound and upper (or lower) shear modulus bound to be preserved when we choose the special value of the angle ϕ (given by (4.23) or (4.28) as appropriate). Without going into a detailed explanation, the reason that the bounds are preserved is that the field $\boldsymbol{E}(\boldsymbol{x})$ takes values lying in the null-space of $\boldsymbol{D}(\boldsymbol{x}) - \boldsymbol{T}$ for all $\boldsymbol{x}$. This field optimality criterion (Avellaneda and Milton, 1995) will remain satisfied if we take the laminate $\boldsymbol{S}^{(1)}$ and change the width of the layers while preserving the field in each phase. In other words we can still satisfy the field optimality conditions with the materials $\boldsymbol{S}^{+}$ and $\boldsymbol{S}^{-}$ mixed in unequal proportions f_1 and f_2. The resulting laminate is potentially a useful building block in the construction of isotropic polycrystals corresponding to the point A. The effective tensor of this laminate, which we denote as $\boldsymbol{S}^{0+}$ is not in general orthotropic.

The next step is to prepare the material $\boldsymbol{S}^{0-}$ that is a mirror image of the material $\boldsymbol{S}^{0+}$ about the x_1-axis, i.e. about a line parallel to the vector $\boldsymbol{i}$. Now we do the following. We rotate the material $\boldsymbol{S}^{0+}$ by the angle ψ and we rotate the material $\boldsymbol{S}^{0-}$ by the angle $-\psi$ and we laminate them together in equal proportions with the vector $\boldsymbol{i}$ being tangential to the layers. By the symmetry in the microstructure, the resulting effective tensor $\boldsymbol{S}_0$ is at least orthotropic. One can easily check that it has the same compressibility s_{11} as the original crystal, viz.,

$$\boldsymbol{a}_1 : \boldsymbol{S}_0 : \boldsymbol{a}_1 = s_{11}, \quad \boldsymbol{a}_1 : \boldsymbol{S}_0 : \boldsymbol{a}_3 = 0, \quad \boldsymbol{a}_2 : \boldsymbol{S}_0 : \boldsymbol{a}_3 = 0. \tag{4.30}$$

The composite obtained in this way still has two free parameters, namely the volume fraction f_1 and the angle ψ. In Appendix D we will see that the appropriate values for these parameters are given by the formulae

$$\cos 2\psi = c_{\#} = \frac{\sqrt{(s_{11}+s_{33})(s_{11}+s_{22}+2s_{12})} - (s_{11}+s_{22}+2s_{12})}{s_{22}-s_{33}+2s_{12}} = -\frac{\sqrt{q}}{\sqrt{q}+\sqrt{p}}, \tag{4.31}$$

$$f_1 = \frac{1}{2} - \frac{(\cos 2\phi)(\cos 2\psi)}{2(\sin 2\phi)(\sin 2\psi)}, \tag{4.32}$$

in which ϕ is given by (4.23), p and q are given by (4.25).

This parameter choice ensures that the composite is isotropic with bulk and shear moduli coinciding with the point A. One can directly check that the positive definiteness of $\boldsymbol{S}$ implies that $c_{\#}$ and f_1 given by these formulae belong to the required intervals

$$c_{\#} \in [-1, 1], \quad f_1 \in [0, 1]. \tag{4.33}$$

This completes the proof that the corner point A is attainable by laminates of the second rank. In order to construct the second rank laminate corresponding to the remaining corner point D, we should first rotate the crystal by 90° to change the sign of s_{12} and then apply the same construction procedure.

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A Appendix A: Finding the optimal translation for the upper shear modulus bound

In this appendix we solve the maximization problem (3.77). As explained in Section 3.6, $\mathcal{T}$ is found by cutting the plane along all curves where

$$f(t_1,t_2) = (s_{33}-t_1)[(s_{11}+t_1)(s_{22}-t_1)-s_{12}^2]-(s_{11}+t_1)\,t_2^2 = 0 \tag{A.1}$$

and picking the piece containing the origin. So a natural first step is to investigate the nature of the curves along which $f(t_1,t_2)=0$.
Since we are ultimately interested in maximizing t_1+t_2, let us introduce two new variables

$$x = s_{11}+t_1, \qquad y = s_{11}+t_1+t_2, \tag{A.2}$$

in terms of which the equation $f(t_1,t_2)=0$ becomes

$$\begin{aligned} F(x,y) &= (s_{11}+s_{33}-x)[x(s_{11}+s_{22}-x)-s_{12}^2]-x(y-x)^2 \\ &= Ax^2+Bx+C=0, \end{aligned} \tag{A.3}$$

where

$$\begin{aligned} A &= 2y-2s_{11}-s_{22}-s_{33}, \\ B &= (s_{11}+s_{22})(s_{11}+s_{33})+s_{12}^2-y^2, \\ C &= -s_{12}^2(s_{11}+s_{33}). \end{aligned} \tag{A.4}$$

Since $F(x,y)$ is a quadratic function of x the equation $F(x,y) = 0$ has two, one, or no solutions for x for a fixed value of y according to whether the discriminant

$$\begin{aligned}
\Delta(y) = B^2 - 4AC &= y^4 - 2[(s_{11}+s_{22})(s_{11}+s_{33}) + s_{12}^2]y^2 + 8(s_{11}+s_{33})s_{12}^2 y \\
&\quad +[(s_{11}+s_{22})(s_{11}+s_{33}) - s_{12}^2]^2 - 4(s_{11}+s_{33})^2 s_{12}^2 \\
&= [y^2 - 2s_{12}y - (s_{11}+s_{22}-2s_{12})(s_{11}+s_{33}) + s_{12}^2] \times \\
&\quad [y^2 + 2s_{12}y - (s_{11}+s_{22}+2s_{12})(s_{11}+s_{33}) + s_{12}^2] \qquad \text{(A.5)}
\end{aligned}$$

is positive, zero, or negative. The positive definiteness of $\boldsymbol{S}$ ensures that the quartic equation $\Delta(y) = 0$ has four real roots:

$$y = y(\gamma_1,\gamma_2) = \gamma_1 s_{12} + \gamma_2\sqrt{(s_{11}+s_{33})(s_{11}+s_{22}-2\gamma_1 s_{12})}, \gamma_1 = \pm 1, \quad \gamma_2 = \pm 1. \qquad \text{(A.6)}$$

When y is negative and sufficiently large there are two solutions for x to the equation $F(x,y) = 0$ because $\Delta(y)$ is approximately equal to y^4 and is therefore positive. Increasing y we sequentially encounter situations when the equation $F(x,y) = 0$ has only one (double) root x, has no solutions at all, has only one (double) root, and again has two solutions. The case illustrated in figure 2 represents the generic situation. Notice there is a special positive value for y namely $y = (2s_{11} + s_{22} + s_{33})/2$ at which $A = 0$. As y passes through this special value one of the roots diverges to infinity and changes sign. Therefore the curves $F(x,y) = 0$ consist of a single closed loop and three open ended curves. These divide the four regions where $F(x,y) \geq 0$ from the singly connected region where $F(x,y) \leq 0$.

When we cut the $x - y$ plane around the loop and along these curves, the set $\mathcal{T}$ is the piece with $F(x,y) \geq 0$ that contains the point (s_{11}, s_{11}), i.e. the origin in the $t_1 - t_2$ plane The set $\mathcal{T}$ is therefore the region enclosed by the loop. The interval obtained by the projection this loop onto the y-axis has endpoints which are roots of the equation $\Delta(y) = 0$. The maximum value of y taken over all points in $\mathcal{T}$ is then clearly the root of the equation $\Delta(y) = 0$ which is greater than s_{11} yet smaller than all other roots having this property.

Using the positive definiteness of $\boldsymbol{S}$ we see after some algebraic manipulations that when

$$(s_{11}+s_{33})(s_{22}-s_{33}) - s_{12}^2 \geq 0 \quad \text{or} \quad s_{11} + 2s_{33} - s_{22} \leq 0 \qquad \text{(A.7)}$$

this root is $y(1,\ 1)$ and (3.77) implies the bound

$$\frac{1}{2\mu_0} \geq -s_{11} + s_{12} + \sqrt{(s_{11}+s_{33})(s_{11}+s_{22}-2s_{12})} = \frac{1}{2\mu^-}, \qquad \text{(A.8)}$$

while when

$$(s_{11}+s_{33})(s_{22}-s_{33}) - s_{12}^2 \leq 0 \quad \text{and} \quad s_{11} + 2s_{33} - s_{22} \geq 0 \qquad \text{(A.9)}$$

this root is $y(-1,\ 1)$ and we have the bound

$$\frac{1}{2\mu_0} \geq -s_{11} - s_{12} + \sqrt{(s_{11}+s_{33})(s_{11}+s_{22}+2s_{12})} = \frac{1}{2\mu^+}. \qquad \text{(A.10)}$$

Note, that the conditions (A.9) are equivalent to (1.12). We also used the condition $s_{12} \leq 0$ that is equivalent to (2.37). This completes the proof of the upper shear modulus bound (1.10).

B Appendix B: Finding the optimal translation for the lower shear modulus bound

Here we solve the maximization problem (3.91) that provides the lower bound on the shear modulus of the polycrystal. We follow a procedure similar to that described in Appendix A, and for this reason we do not provide detailed explanations since these can be found in Appendix A. We introduce the new variables

$$x = t_1, \qquad y = t_1 + t_2, \tag{B.1}$$

in terms of which the equation $f(t_1, f_2) = 0$, where now $f(t_1, t_2)$ is given by (3.90), becomes

$$\begin{aligned} F(x,y) &= [(c_{33} - x)(c_{22} - x) - (y - x)^2][x(c_{11} + x) - (y - x)^2] - xc_{12}^2(c_{33} - x) \\ &= Ax^2 + Bx + C = 0, \end{aligned} \tag{B.2}$$

where

$$\begin{aligned} A &= 4y^2 + 2y(c_{11} - c_{22} - c_{33}) + c_{12}^2 - c_{11}(c_{22} + c_{33}), \\ B &= -4y^3 - y^2(c_{11} - c_{22} - c_{33}) + 2yc_{22}c_{33} + c_{33}(c_{11}c_{22} - c_{12}^2), \\ C &= y^4 - y^2 c_{22}c_{33}. \end{aligned} \tag{B.3}$$

As for the upper shear modulus bound, $F(x, y)$ is a quadratic function of x for any given value of y. The four real roots of the discriminant

$$\begin{aligned} \Delta(y) &= B^2 - 4AC \\ &= [(c_{11} + c_{22} + c_{33})^2 - 4c_{12}^2]y^4 - 4c_{33}[c_{22}(c_{11} + c_{22} + c_{33}) - 2c_{12}^2]y^3 \\ &\quad +2c_{33}[(c_{11}c_{22} - c_{12}^2)(c_{33} - c_{11} - c_{22}) + 2c_{22}c_{33}(c_{22} - c_{11})]y^2 \\ &\quad +4c_{22}c_{33}^2(c_{11}c_{22} - c_{12}^2)y + c_{33}^2(c_{11}c_{22} - c_{12}^2)^2 \\ &= [y^2(c_{11} + c_{22} + c_{33} - 2c_{12}) - 2c_{33}(c_{22} - c_{12})y - c_{33}(c_{11}c_{22} - c_{12}^2)] \times \\ &\quad [y^2(c_{11} + c_{22} + c_{33} + 2c_{12}) - 2c_{33}(c_{22} + c_{12})y - c_{33}(c_{11}c_{22} - c_{12}^2)] \end{aligned} \tag{B.4}$$

can be found explicitly because it is the product of two quadratics. It is convenient to express the formulas for these roots in terms of the coefficients of compliance tensor S:

$$\frac{1}{y} = \frac{1}{y(\gamma_1, \gamma_2)} = -(s_{11} + \gamma_1 s_{12}) + \gamma_2 \sqrt{(s_{11} + s_{33})(s_{11} + s_{22} - 2\gamma_1 s_{12})}, \quad \text{where } \gamma_1 = \pm 1, \quad \gamma_2 = \pm 1. \tag{B.5}$$

Of these four roots $y(1,1),\ y(1,-1),\ y(-1,1)$ and $y(-1,-1)$ the desired root is that one which is greater than zero yet smaller than all other roots having this property, i.e. the root with the largest inverse.
Using the positive definiteness of $\boldsymbol{S}$ we see after some algebraic manipulations that when

$$(s_{11}+s_{33})(s_{22}-s_{33})-s_{12}^2 \geq 0 \quad \text{or} \quad s_{11}+2s_{33}-s_{22} \leq 0 \tag{B.6}$$

this root is $y(-1,\ 1)$ and (3.91) implies the bound

$$\frac{1}{2\mu_0} \leq -s_{11}-s_{12}+\sqrt{(s_{11}+s_{33})(s_{11}+s_{22}+2s_{12})} = \frac{1}{2\mu^+}, \tag{B.7}$$

while when

$$(s_{11}+s_{33})(s_{22}-s_{33})-s_{12}^2 \leq 0 \quad \text{and} \quad s_{11}+2s_{33}-s_{22} \leq 0 \tag{B.8}$$

this root is $y(1,\ 1)$ and we have the bound

$$\frac{1}{2\mu_0} \leq -s_{11}+s_{12}+\sqrt{(s_{11}+s_{33})(s_{11}+s_{22}-2s_{12})} = \frac{1}{2\mu^-}. \tag{B.9}$$

This completes the proof of the lower shear modulus bound.

C Appendix C: The differential scheme for attaining the corner points B and C

We want to reduce the matrix equation (4.11) to the pair of scalar equations (4.13) and (4.14) for the bulk and shear moduli, K_0 and μ_0, of the tensor $\boldsymbol{S}_0$. Because of the eight-fold symmetry of the problem it suffices to take the trace of this equation, and to take its contraction on the left and right with respect to the isotropic basis vector $\boldsymbol{a}_1$. By rotational symmetry all four terms in the sum must yield the same trace. So we only need calculate the trace of the first of the four terms in the sum and set it to zero. Making the substitution

$$\boldsymbol{t}_1\boldsymbol{t}_1 = \frac{1}{\sqrt{2}}(\boldsymbol{a}_1-\boldsymbol{a}_2) \tag{C.1}$$

we see that

$$\begin{aligned}
0 &= \mathrm{Tr}\left[\boldsymbol{S}_1-\boldsymbol{S}_0-\frac{(\boldsymbol{S}_1-\boldsymbol{S}_0):\boldsymbol{t}_1\boldsymbol{t}_1\boldsymbol{t}_1\boldsymbol{t}_1:(\boldsymbol{S}_1-\boldsymbol{S}_0)}{\boldsymbol{t}_1\boldsymbol{t}_1:\boldsymbol{S}_1:\boldsymbol{t}_1\boldsymbol{t}_1}\right] \\
&= s_{11}+s_{22}+s_{33}-1/(2K_0)-1/\mu_0-\frac{(\boldsymbol{a}_1-\boldsymbol{a}_2):(\boldsymbol{S}_1-\boldsymbol{S}_0)^2:(\boldsymbol{a}_1-\boldsymbol{a}_2)}{(\boldsymbol{a}_1-\boldsymbol{a}_2):\boldsymbol{S}_1:(\boldsymbol{a}_1-\boldsymbol{a}_2)} \\
&= s_{11}+s_{22}+s_{33}-1/(2K_0)-1/\mu_0-\frac{(s_{11}-s_{12}-1/2K_0)^2+(s_{12}-s_{22}+1/2\mu_0)^2}{s_{11}+s_{22}-2s_{12}}.
\end{aligned} \tag{C.2}$$

Also by rotational symmetry all four terms in the sum must yield the same contraction with respect to the basis vector $\boldsymbol{a}_1$. So we only need calculate the contraction of the first of the four terms in the sum and set it to zero. In this way we obtain the equation

$$\begin{aligned} 0 &= \boldsymbol{a}_1 : \left[\boldsymbol{S}_1 - \boldsymbol{S}_0 - \frac{(\boldsymbol{S}_1 - \boldsymbol{S}_0) : \boldsymbol{t}_1\boldsymbol{t}_1\boldsymbol{t}_1\boldsymbol{t}_1 : (\boldsymbol{S}_1 - \boldsymbol{S}_0)}{\boldsymbol{t}_1\boldsymbol{t}_1 : \boldsymbol{S}_1 : \boldsymbol{t}_1\boldsymbol{t}_1} \right] : \boldsymbol{a}_1 \\ &= s_{11} - 1/(2K_0) - \frac{[(\boldsymbol{a}_1 - \boldsymbol{a}_2) : (\boldsymbol{S}_1 - \boldsymbol{S}_0) : \boldsymbol{a}_1]^2}{(\boldsymbol{a}_1 - \boldsymbol{a}_2) : \boldsymbol{S}_1 : (\boldsymbol{a}_1 - \boldsymbol{a}_2)} \\ &= s_{11} - 1/(2K_0) - \frac{(s_{11} - s_{12} - 1/2K_0)^2}{s_{11} + s_{22} - 2s_{12}}. \end{aligned} \tag{C.3}$$

This last equation is clearly equivalent to (4.13), and by subtracting (C.3) from (C.2) we obtain (4.14).

D Appendix D: Details of the construction of the second rank laminates attaining the points A

A straightforward application of the layering formula (4.4) shows that the effective tensor $\boldsymbol{S}^{0+}$ of the laminate described in Section 4.3.2 has coefficients

$$\begin{aligned} S_{11}^{0+} &= s_{11}, \\ S_{12}^{0+} &= s_{12}c_*, \\ S_{13}^{0+} &= (f_1 - f_2)s_{12}s_*, \\ S_{22}^{0+} &= s_{22} + (s_{33} - s_{22})(s_*)^2, \\ S_{23}^{0+} &= (f_1 - f_2)(s_{22} - s_{33})c_*s_*, \\ S_{33}^{0+} &= s_{33} + (s_{22} - s_{33})(s_*)^2 - \frac{4f_1f_2(s_{12} + (s_{22} - s_{33})c_*)^2(s_*)^2}{s_{11} + s_{22}(c_*)^2 + s_{33}(s_*)^2 + 2s_{12}c_*}, \end{aligned} \tag{D.1}$$

where

$$s_* = \sin 2\phi, \quad c_* = \cos 2\phi, \tag{D.2}$$

and ϕ is given by (4.23). When we reflect this tensor about the x_1 axis we get a tensor $\boldsymbol{S}^{0-}$ with coefficients

$$\begin{aligned} S_{11}^{0-} &= S_{11}^{0+}, & S_{12}^{0-} &= S_{12}^{0+}, & S_{13}^{0-} &= -S_{13}^{0+}, \\ S_{22}^{0-} &= S_{22}^{0+}, & S_{23}^{0-} &= -S_{23}^{0+}, & S_{33}^{0-} &= S_{33}^{0+}. \end{aligned} \tag{D.3}$$

Next we rotate the material $\boldsymbol{S}^{0+}$ by the angle ψ to get the tensor $\boldsymbol{S}^{1+}$ and we rotate the material $\boldsymbol{S}^{0-}$ by the angle $-\psi$ to get the tensor $\boldsymbol{S}^{1-}$. Let us introduce the parameters

$$c = \cos 2\psi, \qquad s = \sin 2\psi. \tag{D.4}$$

With this simplified notation the tensor

$$\boldsymbol{S}^{1+} = \boldsymbol{R}(\phi) : \boldsymbol{S}^{0+} : \boldsymbol{R}^T(\phi), \tag{D.5}$$

has coefficients given by the equations

$$\begin{aligned}
S_{11}^{1+} &= s_{11}, \\
S_{12}^{1+} &= S_{12}^{0+} c + S_{13}^{0+} s, \\
S_{13}^{1+} &= -S_{12}^{0+} s + S_{13}^{0+} c, \\
S_{22}^{1+} &= S_{22}^{0+} c^2 + S_{33}^{0+} s^2 + 2S_{23}^{0+} cs, \\
S_{23}^{1+} &= (S_{33}^{0+} - S_{22}^{0+}) cs + S_{23}^{0+} (c^2 - s^2), \\
S_{33}^{1+} &= S_{22}^{0+} s^2 + S_{33}^{0+} c^2 - 2S_{23}^{0+} cs.
\end{aligned} \tag{D.6}$$

The tensor

$$\boldsymbol{S}^{1-} = \boldsymbol{R}(-\phi) : \boldsymbol{S}^{0-} : \boldsymbol{R}^T(-\phi) \tag{D.7}$$

is a mirror image of the tensor $\boldsymbol{S}^{1+}$, and therefore

$$\begin{array}{lll}
S_{11}^{1-} = S_{11}^{1+}, & S_{12}^{1-} = S_{12}^{1+}, & S_{13}^{1-} = -S_{13}^{1+}, \\
S_{22}^{1-} = S_{22}^{1+}, & S_{23}^{1-} = -S_{23}^{1+}, & S_{33}^{1-} = S_{33}^{1+}.
\end{array} \tag{D.8}$$

The next step is to laminate these two materials $\boldsymbol{S}^{1+}$ and $\boldsymbol{S}^{1-}$ together in equal proportions with the unit vector $\boldsymbol{i}$ being tangential to the layers. By the symmetry of the construction procedure the resulting effective tensor $\boldsymbol{S}_0$ is orthotropic (i.e. $\boldsymbol{a}_1 : \boldsymbol{S}_0 : \boldsymbol{a}_3 = 0$, $\boldsymbol{a}_2 : \boldsymbol{S}_0 : \boldsymbol{a}_3 = 0$), and using the lamination formula (4.4) we see that it has the same bulk part $\boldsymbol{a}_1 : \boldsymbol{S}_0 : \boldsymbol{a}_1 = s_{11}$ as the original crystal. We now choose the parameters f_1 and ψ of the composite so that

$$S_{12}^{1+} = S_{12}^{1-} = s_{12} c_* c + (f_1 - f_2) s_{12} s_* s = 0, \tag{D.9}$$

where we substitute (D.1) into (D.6) to get the value S_{12}^{1+}. As then follows from the lamination formula (4.4), $\boldsymbol{a}_1 : \boldsymbol{S}_0 : \boldsymbol{a}_2 = 0$ in this case. Therefore, the condition

$$f_2 - f_1 = \frac{c_* c}{s_* s} \tag{D.10}$$

(that is equivalent to (D.9) and leads to (4.32)) ensures that $\boldsymbol{S}_0$ has square symmetry. We can characterize it by the bulk modulus K_l (that coincides with the lower bound (1.5)) and by two shear moduli $\mu^{(1)}$ and $\mu^{(2)}$ given by

$$1/(2\mu^{(1)}) = S_{22}^{1+}, \quad 1/(2\mu^{(2)}) = S_{33}^{1+} - \frac{(S_{13}^{1+} - S_{23}^{1+})^2}{S_{11}^{1+} + S_{22}^{1+}}, \tag{D.11}$$

according to the lamination formula (4.4). Substituting (D.1), (D.6) into (D.11) we arrive at the expressions

$$1/(2\mu^{(1)}) = s_{33} + (s_{33} - s_{22})(c^2 c_*^2 - s^2 s_*^2) - s^2 x, \tag{D.12}$$

$$1/(2\mu^{(2)}) = s_{22} - (s_{33} - s_{22})(c^2 c_*^2 - s^2 s_*^2) - c^2 x - x', \tag{D.13}$$

where

$$\begin{aligned} x &= \frac{(s_{12} + (s_{22} - s_{33})c_*)^2 (s^2 s_*^2 - c^2 c_*^2)}{s^2 (s_{11} + s_{22} c_*^2 + s_{33} s_*^2 + 2 s_{12} c_*)}, \\ x' &= \frac{(s_{12} c_* + (s_{22} - s_{33})(c^2 c_*^2 - s^2 s_*^2) c + c s^2 x)^2}{s^2 (s_{11} + s_{33} + (s_{33} - s_{22})(c^2 c_*^2 - s^2 s_*^2) - s^2 x)}, \end{aligned} \tag{D.14}$$

in which c_* and s_* are determined by (4.23) and (D.2).
It remains to choose the parameter c and $s^2 = 1 - c^2$ so that

$$\frac{1}{2\mu^{(1)}} = -s_{11} - s_{12} + \sqrt{(s_{11} + s_{33})(s_{11} + s_{22} + 2 s_{12})} = \frac{1}{2\mu^+}. \tag{D.15}$$

and to check that the other shear modulus has the same value. After tedious calculations, too lengthy to reproduce here, we find that to insure equality in (D.15) we should put $c = c_\#$ where $c_\#$ is given by the expression (4.31).
We used Maple to check the isotropy of resulting material, i.e. to check that $\mu^{(2)} = \mu^+$ where $\mu^{(2)}$ is given by (D.13). As an additional test, we numerically verified that the two shear moduli are equal for a variety of specific choices of crystal moduli.