Exact relations for effective moduli of polycrystals.

1 Hilbert Space formalism.

Composite materials play an increasingly important role in our everyday life and technology from skis and golf clubs to sensors and actuators in high tech components. By "composite" we mean a perfectly bonded homogeneous mixture of two or more materials on a length scale much smaller than human size and much larger than interatomic distances. The physical properties of a composite (conducting, elastic, etc.) are described by a tensor—the *effective tensor* of a composite. In order to create a composite with desired properties two basic problems become important: prediction of the effective tensors of composite materials and determination of the properties of a given composite by as few measurements as possible.

The principal difficulty in prediction is the universally recognized fact that the effective tensors of composites in general depend on the microstructure (spatial arrangement of component materials). Therefore, the object of importance is the set of all possible effective properties of a composite made with given materials taken in prescribed volume fractions (a so called G-closure set). Unfortunately, aside from a few particular cases the G-closures are extremely difficult to compute analytically.

Usually a G-closure has a non-empty interior in the space of all tensors of appropriate type and may be described by a set of inequalities. On rare occasions researchers have found that a G-closure has empty interior, i.e. becomes part of a hyper-surface. The equations describing such a hyper-surface are called *exact relations* for effective moduli of a composite. We will also use the same term for the hyper-surface itself.

When an exact relation is present the variability of an effective tensor with the microstructure is affected drastically: at or near an exact relation the efforts of achieving certain properties by varying the microgeometry may be futile. At the same time the number of expensive measurements needed to determine material moduli may be significantly reduced. For such important materials as piezoelectrics the number of constants needed for its description is 45 in general but it is only 9 on a 9-dimensional exact relation.

There is a large plethora of known exact relations in various contexts (too large to give a fair list of references). The two most famous ones are Keller-Dykhne-Mendelson relation for 2-D conductivity: $\det\sigma^* = const$, [4, 16, 18], and Hill's result for elasticity that a mixture of isotropic materials with the fixed shear modulus is isotropic with the same shear modulus [13, 14]. All previously known exact relations have been discovered by methods specific to particular applications. These methods were not easily adaptable to every physical context and they by no means guaranteed that there would be no more exact relations. Amazingly, there is a universally applicable method that can produce a complete list of exact relations in virtually every setting.

Let us begin by reviewing the Hilber space formalism for equilibrium equations in non-homogeneous media [21] (see also [3, 9, 15, 17, 25, 30] for similar formal approaches). Assume for simplicity that we are dealing with spatially periodic composite with the period cell $Q = [0, 1]^3$. The intensity fields and fluxes in the body take their values in a certain finite dimensional tensor space $\mathcal{T}$. For example, electric fields and currents are vectors, so $\mathcal{T} = \mathbb{R}^3$ for conductivity. All intensity fields and fluxes will be assumed to belong to the ambient Hilbert space $\mathcal{H} = L^2(Q) \otimes \mathcal{T}$ in addition to satisfying appropriate differential constraints, which place them in a corresponding closed subspace of $\mathcal{H}$. For example, electric fields belong to the subspace of curl-free fields, while currents belong to the subspace of divergence-free fields, according to Maxwell's equations. In general the Hilbert space is split in the orthogonal sum $\mathcal{H} = \mathcal{E} \oplus \mathcal{J} \oplus \mathcal{U}$, where $\mathcal{E}$ and $\mathcal{J}$ are the subspaces of mean zero intensity fields and fluxes respectively, and $\mathcal{U} \cong \mathcal{T}$ is the subspace of uniform fields. The orthogonal projection operator Γ onto $\mathcal{E}$ is a pseudo-differential operator of degree zero and symbol $\Gamma(\vec{k})$, that is an orthogonal projection operator onto a subspace $\hat{\mathcal{E}}_{\vec{k}}$ of $\mathcal{T}$. Local properties of the composite will be described by an L^∞ mapping $C(x)$ of Q into $\mathrm{Sym}(\mathcal{T})$, the space of symmetric operators on $\mathcal{T}$. The function $C(x)$ can also be viewed as an operator C mapping $L^2(Q) \otimes \mathcal{T}$ into $L^2(Q) \otimes \mathcal{T}$: for any $f \in L^2(Q) \otimes \mathcal{T}$

$$(Cf)(x) = C(x)f(x).$$

This formalism allows one to define effective properties of a composite with local positive definite tensor $C(x)$. Given $e_0 \in \mathcal{U}$, find unique $e \in \mathcal{E} \oplus \mathcal{U}$ and $j \in \mathcal{J} \oplus \mathcal{U}$ such that $\langle e \rangle = e_0$ and $j = Ce$, where $\langle e \rangle$ denotes the mean value of e over the period cell Q. Once this cell problem is solved we define

$$\langle j \rangle = C^* e_0.$$

If one knows the solution for $n = \dim\mathcal{T}$ linearly independent tensors $\{e_1, \ldots, e_n\} \subset \mathcal{U}$, then C^* will be completely determined.

One of the key tools is the Milton's W-transformation [21] (independently derived by Zhikov [31]). Let C_0 be a reference medium and let $\Gamma'(\vec{n})$ denote the orthogonal projection onto $C_0^{1/2}\hat{\mathcal{E}}_{\vec{n}}$. Define

$$W_{\vec{n}}(C) = \left[(I - C_0^{-1/2} C C_0^{-1/2})^{-1} - \Gamma'(\vec{n})\right]^{-1}.$$

This transformation maps lamination formula [6, 28] into a convex combination. Namely, if C^* is an effective tensor of a laminate made with materials C_1 and C_2 taken in volume fractions θ_1 and θ_2 with lamination normal $\vec{n}$ then

$$W_{\vec{n}}(C^*) = \theta_1 W_{\vec{n}}(C_1) + \theta_2 W_{\vec{n}}(C_2) \tag{1.1}$$

(see also [1, 27, 29] for other functions mapping lamination formula into convex combinations). A corollary is that for any direction $\vec{n}$ a $W_{\vec{n}}$-image of any set stable under lamination must be a convex set. The idea to use this property to study geometry of sets stable under lamination is due to Francfort and Milton [5].

2 Main Ideas.

Now we are ready to describe our method. Suppose that we have a hyper-surface in $\mathrm{Sym}(\mathcal{T})$ that is stable under homogenization. Then it must also be stable under lamination and its $W_{\vec{n}}$-image must be convex. In addition the W-map is a diffeomorphism, so it maps k-dimensional hyper-surfaces (or simply k-surfaces) into k-surfaces. But a convex k-surface must be a part of a k-plane. Thus we need to identify all those k-surfaces that are mapped into planes by the Milton's W-transformation. In fact it will be easier to identify the corresponding k-planes. Once this is done the exact relation is obtained by taking inverse of the W map. To make life a little easier we may choose the reference medium C_0 to lie on the exact relations manifold. Then the k-planes will in fact be k-dimensional subspaces of $\mathrm{Sym}(\mathcal{T})$. Let us denote such a subspace by Π.

We may also make an additional very natural assumption that we do not restrict the spatial orientation of the constituent materials. A composite with this property is called a polycrystal. Then the sought after surfaces must possess an additional property of rotational invariance. In most physical contexts the subspaces $\mathcal{E}$ and $\mathcal{J}$ possess a rotational invariance property as well:

$$\Gamma(\boldsymbol{R}\cdot\vec{n}) = \boldsymbol{R}\cdot\Gamma(\vec{n}), \tag{2.2}$$

where $\boldsymbol{R}\cdot$ denotes natural action of the rotation group $SO(3)$ on the appropriate tensor space. This natural assumption facilitate the analysis by making methods from representation theory of the rotation group $SO(3)$ relevant. The transformed tensor Γ' will also satisfy (2.2) if we choose C_0 to be isotropic.

In order to formulate our first result we introduce a rotationally invariant subspace

$$\mathcal{A} = \mathrm{Span}\{R\cdot\Gamma' - Q\cdot\Gamma' :\ R, Q \in SO(3)\}, \tag{2.3}$$

where $\Gamma' = \Gamma'(\vec{e}_1)$, for example. The following theorem is proved in [10] in a slightly different form.

Theorem 1 *A subspace Π corresponds to an exact relation stable under lamination if and only if it is rotationally invariant and satisfies*

$$(\Pi\mathcal{A}\Pi)_{\mathrm{sym}} \subset \Pi, \tag{2.4}$$

where the product of subspaces is understood as a linear span of all possible products of elements from the respective subspaces and X_{sym} is a subspace of all symmetric matrices in X.

This theorem can be used to identify all possible exact relations in a given physical context. The rotational of Π allows to use methods from the representation theory of the rotation group $SO(3)$, [12], and significantly reduce the dimensionality of equations (2.4).

All physically interesting spaces correspond to problems that couple n_0 scalar fields, n_1 electric fields and n_2 elastic fields:

$$\mathcal{T} = \mathbb{R}^{n_0} \otimes 1 \oplus \mathbb{R}^{n_1} \otimes \mathbb{R}^3 \oplus \mathbb{R}^{n_2} \otimes \mathrm{Sym}(\mathbb{R}^3). \tag{2.5}$$

Unfortunately, in general the number of rotationally invariant subspaces is infinite and has quite a few parameters. So, the simpleminded checking of all possible cases is not possible in problems larger than 3-D elasticity. Therefore, we need to understand the algebraic structure of (2.4) in order to proceed. One useful observation is that if the subspaces X and Y are rotationally invariant then so is XY. Then one can try to identify the resulting subspace in the complete classification of rotationally invariant subspaces. This approach allows us to handle many parameters in a systematic algebraic fashion. We hope to create a Maple program that produces a list of exact relations for any given problem in finite time.

Another important question is about stability under homogenization. We have proved the following theorem [11]:

Theorem 2 *Let Π' be a rotationally invariant subspace in End($\mathcal{T}$), the set of all linear maps of $\mathcal{T}$ into $\mathcal{T}$, such that*

$$\Pi' \mathcal{A} \Pi' \subset \Pi'. \tag{2.6}$$

Then the subspace Π of symmetric operators in Π' corresponds to an exact relation stable under homogenization.

It is known [23] that there are sets closed under lamination yet not closed under the homogenization. It is not yet known if there are exact relations that are stable under lamination but not under homogenization. The practical utility of (2.6) is similarly limited by the infinite number of invariant subspaces. Again, a theoretical understanding of the nature of (2.6) becomes necessary. For example (2.6) implies that $\Pi'\mathcal{A}$ is an algebra and a rotationally invariant subspace at the same time.

We have applied the above theory to two and three dimensional elasticity obtaining a complete list of exact relations [12] and confirming their stability under homogenization [11]. We have also looked at two and three dimensional piezoelectricity and obtained a partial list of exact relations [12] and showed their stability under homogenization [11]. At present we don't know if there are any more piezoelectric exact relations. It is important to be able to treat situations with coupled fields, the richest source of numerous exact relations [2, 7, 8, 19, 20, 24, 26], because they often can be thought of as correspondences between different physical problems.

Another possible direction of investigation is using rotational invariance together with methods of Francfort and Milton [5] to improve on their result for sets stable under lamination and having non-empty interior. The problem can be formulated as looking for a convex function $H(W)$ such that $h(C) = H(W(C))$ is rotationally invariant. Then all sets $L_c = \{C : h(C) < c\}$ will be stable under lamination. For the case of conductivity, the rotational invariance can be shown to be equivalent to

$$h(C) = F(\mathbf{Tr}(C), \ldots, \mathbf{Tr}(C^n)),$$

where F is a smooth function of n variables. Convexity can be ensured locally by requiring $\nabla\nabla H(W_0) > 0$ at a fixed point $W_0 = W(C_0)$. This results in a set of inequalities for some *constants* involving F and its derivatives at a point. It is still not clear how to investigate stability under homogenization of such sets.

There is a natural connection between sets stable under lamination and quasi-convex translations [22]. We would like to explore this connection in our particular case of exact relations (that don't quite fit in the framework of [22]) as well as in the case of conducting polycrystals. The rotational invariance of the set entails rotational invariance properties of the translations, cutting the number of free parameters (a major nuisance for the translation method) significantly. It is still not clear whether these translation can be used to prove stability under homogenization.

References

[1] G. E. Backus. Long-wave elastic anisotropy produced by horizontal layering. *J. Geophys. Res.*, 67:4427–4440, 1962.

[2] Y. Benveniste. Correspondence relations among equivalent classes of heterogeneous piezoelectric solids under anti-plane mechanical and in-plane electrical fields. *J. Mech. Phys. Solids*, 43(4):553–571, 1995.

[3] G. F. Dell'Antonio, R. Figari, and E. Orlandi. An approach through orthogonal projections to the study of inhomogeneous random media with linear response. *Ann. Inst. Henri Poincaré*, 44:1–28, 1986.

[4] A. M. Dykhne. Conductivity of a two-dimensional two-phase system. *Sov. Phys. JETP*, 32:63–65, 1971.

[5] G. A. Francfort and G. W. Milton. Sets of conductivity and elasticity tensors stable under lamination. *Comm. Pure Appl. Math.*, 47:257–279, 1994.

[6] G. A. Francfort and F. Murat. Homogenization and optimal bounds in linear elasticity. *Arch. Rat. Mech. Anal.*, 94:307–334, 1986.

[7] L. V. Gibiansky and S. Torquato. Link between the conductivity and elastic moduli of composite materials. *Phys. Rev. Lett.*, 71(18):2927–2930, 1993.

[8] L. V. Gibiansky and Milton G. W. On the effective viscoelastic moduli of two-phase media: I. Rigorous bounds on the complex bulk modulus. *Proc. Roy. Soc. Lond., Series A*, 440:163–188, 1993.

[9] Kenneth M. Golden and G. Papanicolaou. Bounds for effective parameters of heterogeneous media by analytic continuation. *Comm. Math. Phys.*, 90:473–491, 1983.

[10] Y. Grabovsky. Exact relations for effective tensors of polycrystals. I: Necessary conditions. *Arch. Rat. Mech. Anal.*, 143(4):309–330, 1998.

[11] Y. Grabovsky, G. W. Milton, and D. S. Sage. Exact relations for effective tensors of polycrystals: Necessary conditions and sufficient conditions. *Comm. Pure. Appl. Math.*, 53(3):300–353, 2000.

[12] Y. Grabovsky and D. S. Sage. Exact relations for effective tensors of polycrystals. II: Applications to elasticity and piezoelectricity. *Arch. Rat. Mech. Anal.*, 143(4):331–356, 1998.

[13] R. Hill. Elastic properties of reinforced solids: Some theoretical principles. *J. Mech. Phys. Solids*, 11:357–372, 1963.

[14] R. Hill. Theory of mechanical properties of fibre-strengthened materials: I. Elastic behaviour. *J. Mech. Phys. Solids*, 12:199–212, 1964.

[15] Y. Kantor and D. J. Bergman. improved rigorous bounds on the effective elastic moduli of a composite material. *J. Mech. Phys. Solids*, 32:41–62, 1984.

[16] J. B. Keller. A theorem on the conductivity of a composite medium. *J. Math. Phys.*, 5:548–549, 1964.

[17] W. Kohler and G. C. Papanicolaou. Bounds for effective conductivity of random media. In R. Burridge, S. Childress, and G. Papanicolaou, editors, *Macroscopic properties of disordered media*, pages 111–130, Berlin, 1982. Springer.

[18] K. S. Mendelson. A theorem on the conductivity of two-dimensional heterogeneous medium. *J. Appl. Phys.*, 46:4740–4741, 1975.

[19] M. Milgrom and S. Shtrikman. Linear responce of polycrystals to coupled fields: Exact relations among the coefficients. *Phys. Rev. B*, 40(9):5991–5994, 1989.

[20] M. Milgrom and S. Shtrikman. Linear responce of two-phase composites with cross moduli: Exact universal relations. *Phys. Rev. A*, 40(3):1568–1575, 1989.

[21] G. W. Milton. On characterizing the set of possible effective tensors of composites: the variational method and the translation method. *Comm. Pure Appl. Math.*, 43:63–125, 1990.

[22] G. W. Milton. A link between sets of tensors stable under lamination and quasiconvexity. *Comm. Pure Appl. Math.*, 47:959–1003, 1994.

[23] G. W. Milton. Personal communication. 1997.

[24] G. W. Milton and A. B. Movchan. A correspondence between plane elasticity and the two-dimensional real and complex dielectric equations in anisotropic media. *Proc. Roy. Soc. Lond., Series A*, 450:293–317, 1995.

[25] G. Papanicolaou and S. Varadhan. Boundary value problems with rapidly oscillating coefficients. In *Colloquia Mathematica Societatis János Bolyai 27, Random Fields (Esztergom, Hungary 1979)*, pages 835–873. North-Holland, 1982.

[26] J. P. Straley. Thermoelectric properties of inhomogeneous materials. *J. Phys. D*, 14:2101–2105, 1981.

[27] L. Tartar. Estimation de coefficients homogénéisés. In *Computing methods in applied sciences and engineering (Proc. Third Internat. Sympos., Versailles, 1977), I,*, volume 704 of *Lecture Notes in Mathematics*, pages 364–373. Springer-Verlag, Berlin, 1979.

[28] L. Tartar. Estimation fines des coefficients homogénéisés. In P. Kree, editor, *Ennio de Giorgi's Colloquium*, pages 168–187, London, 1985. Pitman.

[29] L. Tartar. Estimation of homogenized coefficients. In A. V. Cherkaev and R. V. Kohn, editors, *Topics in the mathematical modeling of composite materials*, volume 31 of *Progress in nonlinear differential equations and their applications*, pages 9–20. Birkhäuser, Boston, 1997.

[30] J. R. Willis. Elasticity theory of composites. In H. G. Hopkins and M. J. Sewell, editors, *Mechanics of solids*, pages 653–686. Pergamon Press, Oxford, New York, 1982.

[31] V. V. Zhikov. Estimates for the homogenized matrix and the homogenized tensor. *Russian Math Surveys*, 46(3):65–136, 1991.