MULTIPHASE CONDUCTORS REALIZING ALEKSANDROV'S MEAN*

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Abstract. It is shown that Aleksandrov's mean for the effective linear conductivity of multiphase conductors is exact for a class of material systems with two-dimensional isotropic microstructures. The systems are constructed by successive use of iterated dilute homogenization. The result provides a more rigorous basis for the use of Aleksandrov's mean to estimate effective properties.

Key words. conductivity, heterogeneity, homogenization

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1. Introduction. The overall electrical conductivity of a multiphase material system is to a major extent dictated by the individual conductivities of its constituent phases and its microstructural morphology. The following developments are restricted to two-dimensional material systems with isotropic phases and isotropic microstructural morphologies.

The simplest theories to estimate the effective conductivity in terms of the local properties are those of Voigt [14] and Reuss [13]. These estimates are respectively given by the arithmetic and harmonic means of the local conductivities $\sigma^{(r)}$ weighted by the volume fractions $c^{(r)}$ of each constituent phase $r = 1, \ldots, N$:

(1)
$$\widetilde{\sigma}_V = \sum_{r=1}^N c^{(r)} \sigma^{(r)}$$
 and $(\widetilde{\sigma}_R)^{-1} = \sum_{r=1}^N c^{(r)} (\sigma^{(r)})^{-1}$

The Voigt and Reuss estimates actually bound from above and below the effective conductivity of all *N*-phase conductors with prescribed volume fractions. However, they both lie outside the optimal upper and lower bounds of Hashin and Shtrikman [6] and are therefore not realizable by any multiphase conductor with isotropic microstructure.

An equally simple estimate was later proposed by Aleksandrov and Aizenberg [2] on the purely mathematical requirement of commutation between inversion and average operations previously advocated by Aleksandrov [1] (see also [12]), which is given by the so-called Aleksandrov's mean:

(2)
$$\ln \widetilde{\sigma}_A = \sum_{r=1}^N c^{(r)} \ln \sigma^{(r)}.$$

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As already observed by Aleksandrov and Aizenberg [2], this estimate lies within the Hashin–Shtrikman bounds and is therefore potentially realizable. We prove here that this is indeed the case by first generating a sequence of two-phase conductors whose effective conductivities asymptote to the two-phase Aleksandrov's mean, and by then using the resulting two-phase microstructures to generate a sequence of *I*-phase conductors whose effective conductivities tend to the *N*-phase Aleksandrov's mean as $I \rightarrow N$. The estimate is realizable in the strong sense that the microstructures realizing it are independent of the values of the local conductivities $\sigma^{(r)}$.

2. Two-phase conductors. Two-phase conductors will be generated by successive use of the iterated dilute homogenization procedure of Bruggeman [4]. This incremental procedure allows the computation of the effective conductivity of two-phase conductors with arbitrary volume fractions of the constituent phases from knowledge of the effective conductivity of two-phase conductors with one of the phases present in dilute, i.e., infinitesimally small, volume fraction.

Consider a two-phase conductor with a dilute volume fraction $c_{dil}^{(2)}$ of phase r = 2, whose effective conductivity satisfies

(3)
$$\widetilde{\sigma}_{\rm dil} = \sigma^{(1)} + \mathcal{G}\left(\sigma^{(1)}, \sigma^{(2)}\right) \ c^{(2)}_{\rm dil} + o\left(c^{(2)}_{\rm dil}\right).$$

The function \mathcal{G} depends on the morphology of the two-phase dilute microstructure. The incremental procedure consists in considering a sequence of two-phase conductors with the same dilute microstructures where the nondilute phase is always made up of material r = 1 but the dilute phase is made up of the preceeding two-phase conductor in the sequence. The resulting material system is a two-phase conductor with an infinite number of microstructural length scales but a finite volume fraction $c^{(2)}$ of phase r = 2, whose effective conductivity is given by

(4)
$$\widetilde{\sigma} = \sigma|_{t=-\ln(1-c^{(2)})},$$

where the function $\sigma(t)$ is solution to the differential equation

(5)
$$\dot{\sigma}(t) = \mathcal{G}\left(\sigma(t), \sigma^{(2)}\right), \qquad \sigma(0) = \sigma^{(1)}$$

Here, the overdot denotes differentiation with respect to t. A rigorous basis for this procedure has been given by Avellaneda [3] along with a fairly general expression for \mathcal{G} in the context of elasticity—see Proposition 7 and Theorem 9 in that work. A simplified version of that expression to the present context is given in the appendix. In what follows we construct a class of two-phase conductors starting out of a matrix containing a dilute suspension of circular inclusions and making iterative use of expression (4). This iterative process amounts to considering a function \mathcal{G} with an explicit dependence on t.

Iteration k = 1. The effective conductivity of a dilute suspension of circular inclusions of material r = 2 embedded in a matrix of material r = 1 is given by (see, for instance, expression (3) in [4])

(6)
$$\widetilde{\sigma}_0 = \sigma^{(1)} + 2\sigma^{(1)} \frac{\sigma^{(2)} - \sigma^{(1)}}{\sigma^{(2)} + \sigma^{(1)}} c_0^{(2)} + o\left(c_0^{(2)}\right).$$

This two-phase conductor clearly belongs to the class (3). Thus, we can use it as the dilute system of the incremental procedure to generate a set of two-phase conductors with finite volume fractions. In view of expressions (4) and (5), the effective conductivity of the resulting two-phase conductor is given by

(7)
$$\tilde{\sigma}_1 = \sigma|_{t=-\ln(1-c_1^{(2)})}, \quad \dot{\sigma}(t) = 2\sigma(t)\frac{\sigma^{(2)} - \sigma(t)}{\sigma^{(2)} + \sigma(t)}, \quad \sigma(0) = \sigma^{(1)},$$

and, upon integration of this differential equation,

(8)
$$\widetilde{\sigma}_{1} = \left[\frac{c_{1}^{(1)}}{2}\frac{\sigma^{(1)} - \sigma^{(2)}}{(\sigma^{(1)})^{1/2}} + \sqrt{\left(\frac{c_{1}^{(1)}}{2}\frac{\sigma^{(1)} - \sigma^{(2)}}{(\sigma^{(1)})^{1/2}}\right)^{2} + \sigma^{(2)}}\right]^{2},$$

where $c_1^{(1)} = 1 - c_1^{(2)}$ is the finite volume fraction of phase r = 1 in the two-phase systems k = 1. This result is known as Bruggeman's differential scheme [4, 11].

Iteration k = 2. As $c_1^{(1)} \to 0$, the effective conductivity $\tilde{\sigma}_1$ is given by

(9)
$$\widetilde{\sigma}_1 = \sigma^{(2)} + \sigma^{(2)} \frac{\sigma^{(1)} - \sigma^{(2)}}{\left(\sigma^{(1)}\sigma^{(2)}\right)^{1/2}} c_1^{(1)} + o\left(c_1^{(1)}\right).$$

Clearly, the two-phase conductor constructed in the first iteration belongs to the class (3) with phases r = 1 and r = 2 interchanged. Thus, we can use it as the dilute system of an incremental process to construct a new set of two-phase conductors with finite volume fractions. In view of expressions (4) and (5), the effective conductivity of such two-phase conductors is given by

(10)
$$\tilde{\sigma}_2 = \sigma|_{t=-\ln(1-c_2^{(1)})}, \quad \dot{\sigma}(t) = \sigma(t) \frac{\sigma^{(1)} - \sigma(t)}{(\sigma^{(1)}\sigma(t))^{1/2}}, \quad \sigma(0) = \sigma^{(2)},$$

and, upon integration of this differential equation,

(11)
$$\widetilde{\sigma}_2 = \sigma^{(1)} \left(\frac{\left[(\sigma^{(1)})^{1/2} + (\sigma^{(2)})^{1/2} \right] - c_2^{(2)} \left[(\sigma^{(1)})^{1/2} - (\sigma^{(2)})^{1/2} \right]}{\left[(\sigma^{(1)})^{1/2} + (\sigma^{(2)})^{1/2} \right] + c_2^{(2)} \left[(\sigma^{(1)})^{1/2} - (\sigma^{(2)})^{1/2} \right]} \right)^2,$$

where $c_2^{(2)}$ is the finite volume fraction of phase r = 2 in the two-phase systems k = 2. Note that this result no longer corresponds to Bruggeman's differential scheme, even though it is closely related.

Iteration k = 3. As $c_2^{(2)} \to 0$, the effective conductivity $\tilde{\sigma}_2$ is given by

(12)
$$\widetilde{\sigma}_2 = \sigma^{(1)} + 4\sigma^{(1)} \frac{(\sigma^{(2)})^{1/2} - (\sigma^{(1)})^{1/2}}{(\sigma^{(1)})^{1/2} + (\sigma^{(2)})^{1/2}} c_2^{(2)} + o\left(c_2^{(2)}\right)$$

Clearly, the two-phase conductor constructed in the second iteration also belongs to the class (3). Thus, we can use it as the dilute system of an incremental process to construct yet another set of two-phase conductors with finite volume fractions. In view of expressions (4) and (5), the effective conductivity of such two-phase conductors is given by

(13)
$$\tilde{\sigma}_3 = \sigma|_{t=-\ln(1-c_3^{(2)})}, \qquad \dot{\sigma}(t) = 4\sigma(t)\frac{(\sigma^{(2)})^{1/2} - (\sigma(t))^{1/2}}{(\sigma(t))^{1/2} + (\sigma^{(2)})^{1/2}}, \qquad \sigma(0) = \sigma^{(1)},$$

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 $\widetilde{\sigma}_{3} = \left[\frac{c_{3}^{(1)}}{2} \frac{(\sigma^{(1)})^{1/2} - (\sigma^{(2)})^{1/2}}{(\sigma^{(1)})^{1/4}} + \sqrt{\left(\frac{c_{3}^{(1)}}{2} \frac{(\sigma^{(1)})^{1/2} - (\sigma^{(2)})^{1/2}}{(\sigma^{(1)})^{1/4}}\right)^{2} + (\sigma^{(2)})^{1/2}}\right]^{4},$

where $c_3^{(1)} = 1 - c_3^{(2)}$ is the finite volume fraction of phase r = 1 in the two-phase systems k = 3.

Iteration k = 4. As $c_3^{(2)} \to 0$, the effective conductivity (14) is given by

(15)
$$\widetilde{\sigma}_3 = \sigma^{(2)} + 2\sigma^{(2)} \frac{(\sigma^{(1)})^{1/2} - (\sigma^{(2)})^{1/2}}{(\sigma^{(1)}\sigma^{(2)})^{1/4}} c_3^{(1)} + o\left(c_3^{(1)}\right),$$

and so, once again, the two-phase conductor constructed in the previous iteration belongs to the class (3). Making use of it as the dilute system of an incremental process we construct a new set of two-phase conductors whose effective conductivity is given by

(16)
$$\tilde{\sigma}_4 = \sigma|_{t=-\ln(1-c_4^{(1)})}, \quad \dot{\sigma}(t) = 2\sigma(t) \frac{(\sigma^{(1)})^{1/2} - (\sigma(t))^{1/2}}{(\sigma^{(1)}\sigma(t))^{1/4}}, \quad \sigma(0) = \sigma^{(2)},$$

and, upon integration of this differential equation,

(17)
$$\widetilde{\sigma}_4 = \sigma^{(1)} \left(\frac{\left[(\sigma^{(1)})^{1/4} + (\sigma^{(2)})^{1/4} \right] - c_4^{(2)} \left[(\sigma^{(1)})^{1/4} - (\sigma^{(2)})^{1/4} \right]}{\left[(\sigma^{(1)})^{1/4} + (\sigma^{(2)})^{1/4} \right] + c_4^{(2)} \left[(\sigma^{(1)})^{1/4} - (\sigma^{(2)})^{1/4} \right]} \right)^4,$$

where $c_4^{(2)}$ is the finite volume fraction of phase r = 2 in the two-phase systems k = 4. Iteration k. Following this iterative process k times it is easy to verify that the

effective conductivities of those two-phase conductors with odd k are given by

(18)

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$$\widetilde{\sigma}_{k} = \left[\frac{c_{k}^{(1)}}{2} \frac{(\sigma^{(1)})^{\alpha_{k}} - (\sigma^{(2)})^{\alpha_{k}}}{(\sigma^{(1)})^{\alpha_{k}/2}} + \sqrt{\left(\frac{c_{k}^{(1)}}{2} \frac{(\sigma^{(1)})^{\alpha_{k}} - (\sigma^{(2)})^{\alpha_{k}}}{(\sigma^{(1)})^{\alpha_{k}/2}}\right)^{2} + (\sigma^{(2)})^{\alpha_{k}}}\right]^{2/\alpha_{k}}$$

with $\alpha_k = (1/2)^{\frac{k-1}{2}}$, while the effective conductivities of those with even k are given by

(19)
$$\widetilde{\sigma}_{k} = \sigma^{(1)} \left[\frac{\left[(\sigma^{(1)})^{\beta_{k}} + (\sigma^{(2)})^{\beta_{k}} \right] - c_{k}^{(2)} \left[(\sigma^{(1)})^{\beta_{k}} - (\sigma^{(2)})^{\beta_{k}} \right]}{\left[(\sigma^{(1)})^{\beta_{k}} + (\sigma^{(2)})^{\beta_{k}} \right] + c_{k}^{(2)} \left[(\sigma^{(1)})^{\beta_{k}} - (\sigma^{(2)})^{\beta_{k}} \right]} \right]^{1/\beta_{k}}$$

with $\beta_k = (1/2)^{\frac{k}{2}}$.

Iteration $k \to \infty$. As $k \to \infty$ the exponents α_k and β_k in the previous expressions tend to zero. Therefore, if $c_k^{(r)} \to c^{(r)}$, the sequence of $\tilde{\sigma}_k$ given by (18)–(19) is such that

(20)
$$\widetilde{\sigma}_k \to \widetilde{\sigma}_A,$$

where $\tilde{\sigma}_A$ is Aleksandrov's mean (2) with N = 2. These limiting material systems will be referred to as two-phase Aleksandrov conductors.

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3. *N*-phase conductors. By means of a second iterative process, the above class of two-phase Aleksandrov conductors can be used to generate *N*-phase conductors whose effective conductivities are given by Aleksandrov's mean (2).

Iteration i = 1. Consider a two-phase Aleksandrov conductor whose constituent phases are made up of materials r = 1 and r = 2; the effective conductivity of this material system is then given by

(21)
$$\ln \tilde{\sigma}_1 = c_1^{(1)} \ln \sigma^{(1)} + c_1^{(2)} \ln \sigma^{(2)},$$

where $c_1^{(r)}$ denotes the volume fraction of each phase r in the two-phase conductor i = 1.

Iteration i = 2. Now consider a two-phase Aleksandrov conductor made up of material r = 3 and of the two-phase Aleksandrov conductor of the previous iteration. The effective conductivity $\tilde{\sigma}_2$ of this three-phase conductor is then given by

(22)
$$\ln \tilde{\sigma}_{2} = (1 - c_{2}^{(3)}) \ln \tilde{\sigma}_{1} + c_{2}^{(3)} \ln \sigma^{(3)} \\ = (1 - c_{2}^{(3)})c_{1}^{(1)} \ln \sigma^{(1)} + (1 - c_{2}^{(3)})c_{1}^{(2)} \ln \sigma^{(2)} + c_{2}^{(3)} \ln \sigma^{(3)} \\ = c_{2}^{(1)} \ln \sigma^{(1)} + c_{2}^{(2)} \ln \sigma^{(2)} + c_{2}^{(3)} \ln \sigma^{(3)},$$

where $c_2^{(r)}$ refers to the volume fraction of material r in the three-phase conductor i = 2.

Iteration N. Performing this iterative process N times leads to an N-phase conductor with effective conductivity given by

(23)
$$\ln \widetilde{\sigma}_N = \sum_{r=1}^N c_N^{(r)} \ln \sigma^{(r)},$$

where $c_N^{(r)}$ refers to the volume fraction of material r in the N-phase system. Evidently, if $c_N^{(r)} = c^{(r)}$, the effective conductivity $\tilde{\sigma}_N$ is such that

(24)
$$\widetilde{\sigma}_N = \widetilde{\sigma}_A$$

where $\tilde{\sigma}_A$ is Aleksandrov's mean (2) for an N-phase conductor.

4. Discussion. The microstructures produced by the above contruction process are of granular character in the sense that there is no continuous phase playing the role of a matrix. Alexandrov estimates are therefore expected to be accurate for granular systems. These estimates are compared with various bounds and estimates for twophase conductors in Figure 1. Part (a) shows plots of the effective conductivity as a function of volume fraction $c^{(2)}$ for the choice $\sigma^{(1)} = 5\sigma^{(2)}$. It is observed that Aleksandrov estimates (AL) lie within the bounds of Hashin and Shtrikman (HS) as expected in view of their realizability. In addition, it is seen that Aleksandrov estimates lie close to the effective medium approximation (EMA) of Bruggeman [4] and agree exactly with this approximation when both phases are present at equal volume fractions ($c^{(1)} = c^{(2)} = 1/2$). Recall that the EMA estimate is also realizable by a class of granular systems [10]. That Aleksandrov and EMA estimates agree exactly when both volume fractions are equal is a consequence of the fact that the two-phase conductors realizing these estimates have symmetric microstructures in that case and their effective conductivities must therefore be given by the Keller–Dykhne formula



FIG. 1. Various bounds and estimates for the effective conductivity of two-phase conductors: (a) as a function of volume fraction $c^{(2)}$ for the choice $\sigma^{(1)} = 5\sigma^{(2)}$, and (b) as a function of local conductivity contrast for the choice $c^{(2)} = 0.3$.

for symmetric materials [5, 7] ($\tilde{\sigma} = \sqrt{\sigma^{(1)}\sigma^{(2)}}$). Part (b) shows plots of the effective conductivity as a function of local conductivity contrast for the choice $c^{(2)} = 0.3$. It is observed that Aleksandrov and EMA estimates give similar predictions for low to moderate contrasts but diverge for large contrasts. This is a consequence of the fact that these estimates exhibit different percolation thresholds for infinitely contrasted systems: the EMA estimates percolate at $c^{(2)} = 1/2$, while Aleksandrov estimates percolate at $c^{(2)} = 0$. In any case, the fact that Aleksandrov estimates are realized by two-dimensional systems provides a more rigorous basis for its use to estimate effective properties, at least for model systems of interest to theoretical studies in linear as well as nonlinear homogenization [8, 9, 11].

We conclude by recalling that the Voigt, Reuss, and Aleksandrov estimates are all particular instances of the more general family of geometric estimates given by

(25)
$$(\widetilde{\sigma}_G)^{\gamma} = \sum_{r=1}^N c^{(r)} (\sigma^{(r)})^{\gamma},$$

with the Voigt and Reuss estimates corresponding to $\gamma = 1$ and $\gamma = -1$, respectively, and the Aleksandrov estimate corresponding to $\gamma \to 0$. Letting $\sigma^{(r)} = \sigma_0 + t \,\delta\sigma^{(r)}$ and expanding to second order in t the geometric estimate (25) along with the Hashin– Shtrikman upper and lower bounds we obtain

(26)
$$\widetilde{\sigma}_G = \langle \sigma \rangle - \frac{1}{2} (1 - \gamma) \frac{\langle \sigma^2 \rangle - \langle \sigma \rangle^2}{\sigma_0} + O(t^3),$$

(27)
$$\widetilde{\sigma}_{HS}^{+} = \widetilde{\sigma}_{HS}^{-} = \langle \sigma \rangle - \frac{1}{2} \frac{\langle \sigma^2 \rangle - \langle \sigma \rangle^2}{\sigma_0} + O(t^3),$$

where angular brackets denote volume average over the composite. Since the Hashin–Shtrikman upper and lower bounds agree exactly to second order in t, any realizable estimate must also agree with these bounds to second order in t. Thus, the geometric estimate (25) can be realizable *only* when the exponent $\gamma \to 0$, i.e., when it reduces to Aleksandrov's mean.

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Appendix. Avellaneda [3] derived an expression for \mathcal{G} in the context of linear elasticity under the assumption that the domain $\Omega^{(2)}$ occupied by the dilute phase is a bounded subset of \mathbb{R}^n with Lipschitz boundary. In the case of two-dimensional (n = 2) conductors with isotropic phases and isotropic microstructural morphologies, that expression can be written as

(28)
$$\mathcal{G}\left(\sigma^{(1)},\sigma^{(2)}\right) = \left(\sigma^{(2)} - \sigma^{(1)}\right) \left[1 + \overline{\mathbf{e}} \cdot \langle \nabla \phi \rangle^{(2)}\right].$$

where $\overline{\mathbf{e}}$ is a unit vector, $\langle \cdot \rangle^{(2)}$ denotes averaging over $\Omega^{(2)}$, and $\phi(\mathbf{x})$ satisfies the field equations

(29)
$$\nabla \cdot [\sigma(\mathbf{x})(\overline{\mathbf{e}} + \nabla \phi)] = 0 \quad \text{in} \quad \mathbb{R}^2, \quad \int_{\mathbb{R}^2} |\nabla \phi|^2 < \infty,$$

with

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(30)
$$\sigma(\mathbf{x}) = \chi^{(1)}(\mathbf{x})\sigma^{(1)} + \chi^{(2)}(\mathbf{x})\sigma^{(2)}.$$

Here, $\chi^{(2)}(\mathbf{x}) = 1 - \chi^{(1)}(\mathbf{x})$ denotes the characteristic function of $\Omega^{(2)}$. The function \mathcal{G} is independent of $\overline{\mathbf{e}}$ in view of the assumed overall isotropy.

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