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Transport in Macroscopically Inhomogeneous Materials[1]

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We consider electrical and thermal transport in macroscopically inhomogeneous materials, when the two components forming a mixture have very different conductance properties. Because of their complexity, such systems are sometimes modeled by resistor networks. It is shown that the most natural models violate the Hashin–Shtrikman bounds to the effective conductivity of continuous composite materials. The distribution of the Joule heat between the phases, in the case of electrical conductance, is also largely erroneous. Thus, better estimates of conductance properties are obtained by disregarding detailed information about the phase geometry and instead using general methods for continuous materials, valid for a wide class of geometries.

KEY WORDS: composite materials; electrical conduction; resistor networks; thermal conduction.

1. INTRODUCTION

The effective conductivity of a macroscopically inhomogeneous two-phase material depends on the phase conductivities and on the concentration and geometrical distribution of the phases. However, it is usually difficult to evaluate the effective conductivity exactly, and approximate methods are required. Discrete models, in which few resistors in special configurations represent each grain, provide a drastic simplification.

The conductance properties of resistor networks, in which two resistors, R_1 and R_2, are distributed according to certain prescriptions, have been thoroughly studied from the point of view of critical behavior

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and percolation properties when R_1/R_2 is zero or infinite [1, 2]. Yet little is known about the applicability of discrete resistor models to real composite materials. It is the purpose of this paper to discuss that question. In a previous paper [3], dealing only with two dimensions, it was shown that natural discretizations violate certain absolute bounds to the overall conductivity of two-phase materials. The same behavior, illustrated by a specific example, is shown here to arise in three dimensions. Our presentation is in terms of the electrical conductivity, but it is relevant also for thermal conduction.

2. DISCRETIZATION MODELS

Consider a grain of a two-dimensional two-phase material. A reasonable description of the grain requires at least as many resistors as the number of neighboring grains (neglecting point-contacts). Figure 1 shows four such discretizations of a square grain. We call the resistor models in Figs. 1a, b, c, and d the cross, side, corner, and mid models, respectively. They are easily generalized to three dimensions. For instance, Fig. 2 shows the cross model of a cubic grain. The side model has 12 equal resistors, along the sides of a cube. The corner model has eight resistors, each one connecting a cube corner with the cube center. The mid model has 12 resistors, connecting the midpoints of adjacent cube sides. The number of resistors in each grain does not depend on the grain size. The magnitude of

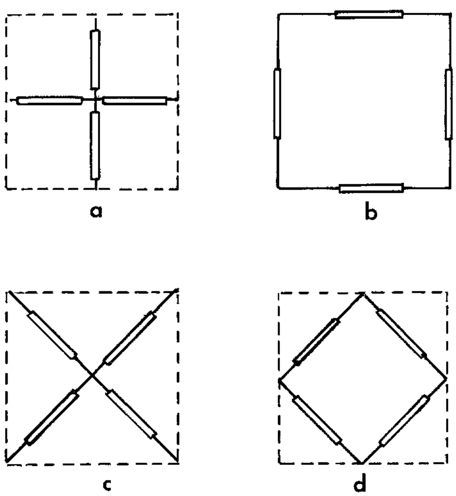

Fig. 1. The cross (a), side (b), corner (c), and mid (d) resistor models of a square grain.

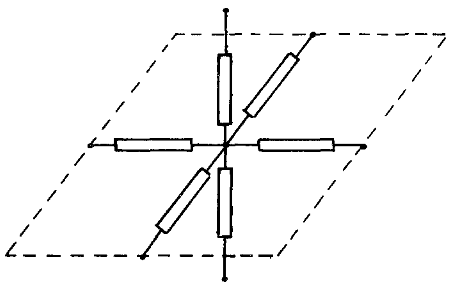

Fig. 2. The three-dimensional analogue of the two-dimensional cross model (Fig. 1a).

the resistors in a grain all have the value R_1 or R_2, depending on which phase they represent. The surface (or volume) fractions of the two phases are c_1 and c_2 $(=1-c_1)$. Hence, c_1 measures the total number of resistors R_1. A model like this keeps the number of parameters at a minimum, but it still contains information about the geometrical arrangement of the grains and their conductivities. Further details on discretization models are found in our treatment of the two-dimensional case [3, 4].

3. GENERAL RESULTS FOR TWO-PHASE MATERIALS

We now recapitulate some known results for three-dimensional two-phase materials. We have in mind materials which are isotropic and homogeneous, on a length scale larger than that characteristic of the grains. Hashin and Shtrikman [5] used a variational method to show that the overall effective conductivity, σ_e, is bounded as follows $(\sigma_2 > \sigma_1)$:

$$\sigma_1 + c_2[1/(\sigma_2 - \sigma_1) + c_1/(3\sigma_1)]^{-1} < \sigma_e < \sigma_2 + c_1[1/(\sigma_1 - \sigma_2) + c_2/(3\sigma_2)]^{-1} \tag{1}$$

Bruggeman [6], Landauer [7], and others derived an effective medium theory for σ_e. The result is

$$c_1(\sigma_1 - \sigma_e)/(\sigma_1 + 2\sigma_e) = c_2(\sigma_e - \sigma_2)/(\sigma_2 + 2\sigma_e) \tag{2}$$

Schulgasser [8] proved that for cell materials (which includes our example in Section 4) with $c_1 = c_2 = 0.5$,

$$\sigma_e > (\sigma_1 \sigma_2)^{\frac{1}{2}} \tag{3}$$

Table I. The Effective Conductivity of Stacked Cubes, with $\sigma_2/\sigma_1 \gg 1$

Exact result	$2(\sigma_1\sigma_2)^{\frac{1}{2}}$
Lower Hashin–Shtrikman bound	$4\sigma_1$
Upper Hashin–Shtrikman bound	$(2/5)\,\sigma_2$
Effective medium theory	$\sigma_2/4$
Lower Schulgasser bound	$(\sigma_1\sigma_2)^{\frac{1}{2}}$
Discretized model (side, corner)	$\sigma_2/2$
Discretized model (cross, mid)	$2\sigma_1$

4. AN EXAMPLE: STACKED CUBES

A specific example is now used to illustrate certain features of discretized models. Consider the three-dimensional analogue of a regular checkerboard, i.e., a material consisting of cubic grains of two phases, stacked in alternating sequencies. This system is one of the very few for which the conductivity is known exactly. When $\sigma_1/\sigma_2 \gg 1$ (or $\ll 1$), one has $\sigma_e = 2(\sigma_1\sigma_2)^{\frac{1}{2}}$ [9]. Table I compares this result with the Hashin–Shtrikman bounds, the Bruggeman–Landauer effective medium theory, and Schulgasser's inequality, all for $\sigma_2/\sigma_1 \gg 1$. One should note that a system of cubic symmetry, such as our example of stacked cubes, has an isotropic overall conductivity, and the results in Section 3, for isotropic systems, are applicable [10].

It is not very difficult to calculate the effective conductivity in the four discretization models discussed above. The result is a series or parallel coupling of R_1 and R_2, which can be written

$$\sigma_e = (\sigma_1 + \sigma_2)/2 \tag{4}$$

and

$$\sigma_e = 2\sigma_1\sigma_2/(\sigma_1 + \sigma_2) \tag{5}$$

for the side and corner [Eq. (4)] and the cross and mid [Eq. (5)] models. Here we have normalized R_1 and R_2 so that all four discretizations give the

Table II. The Effective Conductivity of Stacked Cubes, with $\sigma_2/\sigma_1 = 10$

Lower Hashin–Shtrikman bound	$2.8\,\sigma_1$
Upper Hashin–Shtrikman bound	$4.706\,\sigma_1$
Lower Schulgasser bound	$3.161\,\sigma_1$
Discretized model (side, corner)	$5.5\,\sigma_1$
Discretized model (cross, mid)	$1.818\,\sigma_1$

same conductivity when $\sigma_1 = \sigma_2$ (i.e., when $R_1 = R_2$). Table II gives the effective conductivities as obtained from Eqs. (1), (4), and (5), when $\sigma_2/\sigma_1 = 10$. We note that σ_e in all four discretization models falls outside the Hashin–Shtrikman bounds.

Let $r = Q_1/Q_2$ be the ratio of the total Joule heat in phase 1 and phase 2, when an electric current flows through a composite material. In the case of stacked cubes with $\sigma_2/\sigma_1 \gg 1$, $r = 1$ [8]. Our discretization models of the same system are easily found to yield $r = \sigma_1/\sigma_2$ for the corner and side models and $r = \sigma_2/\sigma_1$ for the cross and mid models.

5. DISCUSSION AND CONCLUSIONS

The effective conductivity of a composite material which, in a statistical sense, is isotropic and homogeneous, always falls within the Hashin–Shtrikman bounds, irrespective of details in the geometrical distribution of the phases. If the phase distribution is known, one might try to include that information through a resistor network model. Our example shows that such a procedure can yield results which violate the Hashin–Shtrikman bounds. Then the attempt to include the phase geometry leads to an estimate of σ_e which is worse than an estimate based only on the amounts c_i of the phases, without regard to the detailed phase geometry. We have also seen that the ratio of the total Joule heat in the two phases is largely erroneous, if estimated by a discretized model.

REFERENCES

1. S. Kirkpatrick, *Rev. Mod. Phys.* **45**:574 (1973).
2. D. Stauffer, *Phys. Rep.* **54**(1):1 (1979).
3. M. Söderberg and G. Grimvall, *J. Appl. Phys.* (1986, in press).
4. M. Söderberg, P.-O. Jansson, and G. Grimvall, *J. Phys. A* **18**:L633 (1985).
5. Z. Hashin and S. Shtrikman, *J. Appl. Phys.* **33**:3125 (1962).
6. D. A. G. Bruggeman, *Ann. Phys.* **24**:636 (1935).
7. R. Landauer, *J. Appl. Phys.* **23**:779 (1952).
8. K. Schulgasser, *J. Math. Phys.* **17**:378 (1976).
9. M. Söderberg and G. Grimvall, *J. Phys. C* **16**:1085 (1983).
10. G. W. Milton, *J. Appl. Phys.* **52**:5286 (1981).