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1998 J. Phys.: Condens. Matter 10 9729

(http://iopscience.iop.org/0953-8984/10/43/016)

J. Phys.: Condens. Matter **10** (1998) 9729–9735. Printed in the UK PII: S0953-8984(98)93541-X

Conductivity of realizable effective medium intergranularly random and completely random polycrystals against the bounds for isotropic and symmetrically random aggregates

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Received 22 April 1998, in final form 12 June 1998

Abstract. The envelopes of the overall conductivities of effective medium intergranularly random and completely random polycrystalline aggregates are compared with the available bounds on the polycrystals' properties. The geometrically realizable models cover the major parts of the property ranges permitted by the bounds, hence the estimates represent the behaviour of realistic random aggregates well, given the uncertainty in the shapes of constituent crystals.

1. Introduction

Most practical heterogeneous media are of random nature. Take a single-phase polycrystal, which is a random aggregate of crystalline grains. *Randomness* in the sense here is much more definite than the uncertainty of *arbitrariness*. For example, an arbitrary aggregate of anisotropic crystals can have a wide range of overall properties, including the anisotropic ones, while the randomness should imply, among other characteristics, the macroscopically isotropic appearance of an aggregate [1]. The random cubic resistor network of Kirkpatrick [2], in which the bonds are assigned different conductivities randomly, has quite definite overall behaviour independent of the particular samples taken, provided that the samples are sufficiently large in comparison with the inhomogeneities (here the bonds). The properties of different bonds in the network are completely uncorrelated, or in other words, the correlation is restricted to within a single bond. The crystalline and shape orientations, respectively, of different grains in a random aggregate are uncorrelated, or in other words, the correlation is restricted to within a single grain. We will refer to such a realistic aggregate as an *intergranularly random polycrystal*. In certain circumstances, the relative crystalline and shape orientations within a single grain are also uncorrelated. We will refer to that smaller class of random aggregates as *completely random polycrystals*. However, as distinct from the random cubic network, which is based on a definite idealistic *skeleton* (the cubic network geometry), a realistic random polycrystal is formed from grains having all possible irregular shapes. That uncertainty in the base skeleton (the grains' configuration) should result in some uncertainty in the observed overall properties of random polycrystalline aggregates. Evaluation of those *scatter intervals* for the effective conductivity of the realistic intergranularly random and completely random polycrystals should be of theoretical and practical interest.

There are certain upper and lower bounds constructed theoretically somewhere, the effective property of any aggregate should fall strictly between them. Here we would

0953-8984/98/439729+07$19.50

like to specify the validity of those bounds with respect to the two realistic classes of integranularly random and completely random polycrystals stated above. Another question is how good are the bounds for a class of aggregates, i.e. if the effective properties of the aggregates within the class may scatter on the whole interval between the bounds or they may cover only a much smaller interval, or they might even converge to unique values like those of the random cubic network of Kirkpatrick [2]? Here we are interested in the specific geometries, the effective properties of which can be determined exactly. The envelopes of the properties (formed from the greatest and smallest properties) should give us additional information about the possible scatter interval for the property, besides that of the bounds: while the interval between the upper and lower bounds is larger and contains the scatter interval, the interval between the upper and lower envelopes of realizable geometries is smaller and belongs to that scatter interval.

2. Realizable EM models versus the bounds

It is known [3–6] that the effective conductivity σ_e of any macroscopically isotropic aggregate of crystals should be bounded by certain estimates expressed exclusively through the principal conductivities $\sigma_1, \sigma_2, \sigma_3$ of the basic crystal. The upper arithmetic average bound is very simple

$$\sigma_e \leqslant \sigma_{av}^u = (\sigma_1 + \sigma_2 + \sigma_3)/3 \tag{1}$$

while the lower bound of Schulgasser [4] and Avallaneda *et al* [5], in the case of a uniaxial basic crystal ($\sigma_2 = \sigma_3$), can also be given explicitly

$$\sigma_e \geqslant \sigma_{SACLM}^l = \tfrac{1}{2}[(\sigma_1^2 + 8\sigma_1\sigma_2)^{1/2} - \sigma_1]. \tag{2}$$

Those *bounds for isotropic polycrystals* (i.e. any aggregate, including the non-random one, that has a macroscopically isotropic property) are attained by certain locally-ordered configurations and therefore are verified to be the optimal ones. The class of realistic intergranularly random polycrystals, which also have isotropic properties, should be smaller than that of all the isotropic ones, so its uncertainty interval for the effective conductivity should be smaller. How small it would be? We will construct envelopes of certain realizable intergranularly random aggregates, which provides an *inside estimate* for the interval, and compare it with the *outside estimate* (1) and (2).

For the subclass of realistic completely random polycrystals, tighter bounds have been constructed. Based on the hypothesis that certain tensors, which reflect the relative shape and the crystalline orientations of constituent crystals, should be isotropic [7], one can derive the estimates known as Hashin–Shtrikman bounds [8, 9] which are much tighter than the bounds for isotropic polycrystals. By exploring the equiaxity of the polycrystals and the interpolation between the Hashin–Shtrikman and Avelaneda *et al* bounds [5], Helsing [10, 11] further improved the bounds. Using, in addition to the isotropy hypothesis, the assumption that an interchange of the places between any two sets of crystals having different crystalline orientations in a random aggregate should not alter the effective conductivity of the polycrystal, we derived estimates [7], which appear the best available shape-independent bounds for completely-random polycrystals over most ranges of parameters $\sigma_1, \sigma_2, \sigma_3$. Those estimates, referred to also as the *bounds for symmetrically random* (or *disordered*, or *perfectly random) polycrystals*, in the case of a uniaxial basic crystal ($\sigma_2 = \sigma_3$), reduce to

$$P(2\sigma_0^u) = \sigma_P^u \geqslant \sigma_c \geqslant \sigma_P^l = P(2\sigma_0^l) \tag{3}$$

where

$$P(2\sigma_0) = 3\left(\frac{1}{\sigma_1 + 2\sigma_0} + \frac{2}{\sigma_2 + 2\sigma_0}\right)^{-1} - 2\sigma_0$$

$$\sigma_0^u = \begin{cases} (\sigma_1 + 2\sigma_2)/3 & \text{if } \sigma_1 \leqslant \sigma_2 \\ (7\sigma_1 + 13\sigma_2)/20 & \text{if } \sigma_1 \geqslant \sigma_2 \end{cases}$$

$$\sigma_0^l = \begin{cases} 3/(1/\sigma_1 + 2/\sigma_2) & \text{if } \sigma_1 \geqslant \sigma_2 \\ 20/(7/\sigma_1 + 13/\sigma_2) & \text{if } \sigma_1 \leqslant \sigma_2. \end{cases}$$

Comparisons of different available bounds are given in [7]. Though the bounds (3) tell us that an effective property should lie between σ_p^u and σ_p^l, they would not confirm that the effective property of a completely random polycrystal can be as high as σ_p^u, or as low as σ_p^l, in other words, the bounds may be not the best possible ones for completely random polycrystals and can still be made narrower. Here we will construct an inside estimate of the uncertainty interval for completely random polycrystals as envelopes of certain realizable models, and compare it with the outside estimate (3).

Among various approximation schemes for the effective properties of heterogeneous media, the most successful one is the effective medium approximation (referred to also as the coherent potential approximation in the theory of electronic states in disordered alloys) [12–19]. For the approximation, each inhomogeneity is considered equally as a spherical (ellipsoidal, or generally, of any shape) particle embedded in a homogeneous medium with unknown effective property, so the fields inside it can be evaluated from the respective dilute suspension problem. Then a summation of the fields in the inhomogeneities should yield the self-consistent equation determining an effective property of the aggregate. The scheme agreed with many observations and is especially suitable for a description of the behaviour of random polycrystals. Moreover, the effective medium approximation is a realizable scheme, becomes exact for certain hierachical geometries, and, by construction, can represent certain intergranularly random and completely random aggregates. Presume that a dilute suspension of a basic crystal having principal conductivities $\sigma_1, \sigma_2, \sigma_3$ and certain shape with volume fraction $\mathrm{d}v$ in an isotropic matrix of conductivity $\boldsymbol{\sigma} = \mathrm{diag}\{\sigma, \sigma, \sigma\}$ has the effective conductivity $\boldsymbol{\sigma} + \mathrm{d}\boldsymbol{\sigma}$

$$\boldsymbol{\sigma} + \mathrm{d}\boldsymbol{\sigma} = \boldsymbol{\sigma} + \mathrm{d}v\boldsymbol{D}(\boldsymbol{\sigma}, \boldsymbol{\sigma}_R) \tag{4}$$

where $\boldsymbol{\sigma}_R = \boldsymbol{R}^t\boldsymbol{\sigma}_b\boldsymbol{R}$, $\boldsymbol{\sigma}_b = \mathrm{diag}\{\sigma_1, \sigma_2, \sigma_3\}$, $\boldsymbol{R}$ is an orthogonal matrix determining the relative orientation of the principal conductivity directions with respect to a local system of coordinates fixed with the shape of the basic crystal. According to the procedure of the effective medium approximation, the effective conductivity σ_e of the aggregate is determined from the self-consistent equation

$$\langle \mathrm{trace}\{\boldsymbol{O}^t\boldsymbol{D}(\boldsymbol{\sigma}_e, \boldsymbol{\sigma}_R)\boldsymbol{O}\}\rangle_{\boldsymbol{O}} = 0 \tag{5}$$

where $\langle\cdot\rangle_{\boldsymbol{O}}$ denotes the average over the rotations $\boldsymbol{O}$ (distributed equally over all directions in the space) of the basic crystal. In case the crystalline and shape orientations within a basic crystal are uncorrelated, we get the equation (the average is taken over all uncorrelated orientations $\boldsymbol{O}$ and $\boldsymbol{R}$)

$$\langle \mathrm{trace}\{\boldsymbol{O}^t\boldsymbol{D}(\boldsymbol{\sigma}_e, \boldsymbol{R}^t\boldsymbol{\sigma}_b\boldsymbol{R})\boldsymbol{O}\} >_{\boldsymbol{O},\boldsymbol{R}} = 0. \tag{6}$$

The aggregates with the effective conductivity determined by (5) are called *EM (effective medium) intergranularly random polycrystals*, while those by (6) will be named *EM completely random polycrystals*. Equations (5) and (6) can be made explicit for aggregates

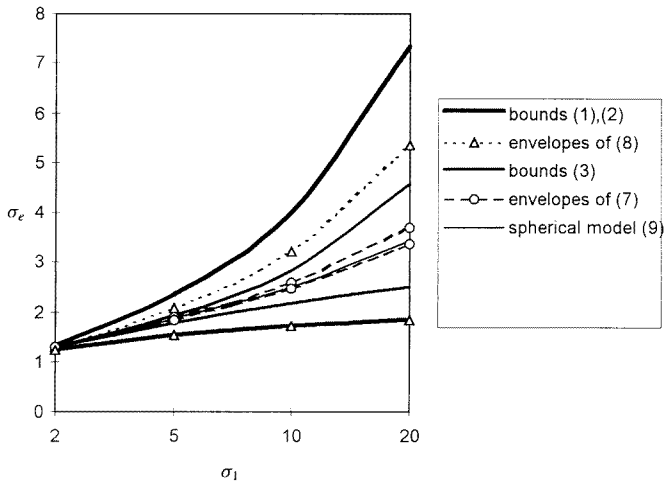

Figure 1. Bounds and envelopes of realizable models for aggregates of uniaxial crystals: $\sigma_2 = \sigma_3$ is normalized to be 1, $\sigma_1 = 2 \to 20$; the upper (1) and lower (2) bounds compared with the upper and lower envelopes of (8) for integranularly random polycrystals; the upper and lower bounds (3) compared with the upper and lower envelopes of (7) for completely random polycrystals.

of ellipsoidal grains [9, 20] In particular, the self-consistent equation (6) for *EM completely random ellipsoidal polycrystals* can be given in the form

$$\langle \text{trace}\{\boldsymbol{R}^t \delta\boldsymbol{\sigma} \boldsymbol{R}(\boldsymbol{\sigma}_e + \boldsymbol{O}^t \boldsymbol{L} \boldsymbol{O} \boldsymbol{R}^t \delta\boldsymbol{\sigma} \boldsymbol{R})^{-1}\}\rangle_{O,R} = 0 \tag{7}$$

where $\delta\boldsymbol{\sigma} = \text{diag}\{\sigma_1 - \sigma_e, \sigma_2 - \sigma_e, \sigma_3 - \sigma_e\}$, $\boldsymbol{L} = \text{diag}\{L_1, L_2, L_3\}$; the depolarization factors $L_i, 1 \leqslant i \leqslant 3$ satisfy $0 \leqslant L_i \leqslant 1, \sum_{i=1}^{3} L_i = 1$; the orientations $\boldsymbol{O}$ and $\boldsymbol{R}$ here are uncorrelated. The equation (5) for *EM intergranularly random ellipsoidal polycrystals* having principal conductivity directions which coincide with the principal axes of the ellipsoidal shape of the basic crystal becomes

$$\sum_{i=1}^{3} \frac{\sigma_i - \sigma_e}{\sigma_e + L_i(\sigma_i - \sigma_e)} = 0. \tag{8}$$

At $L_1 = L_2 = L_3 = 1/3$, (7) and (8) reduce to a unique equation for EM spherical polycrystals, which is resolved explicitly in the case of a uniaxial basic crystal ($\sigma_2 = \sigma_3$)

$$\sigma_e = \sigma_s = \tfrac{1}{4}[(\sigma_2^2 + 8\sigma_1\sigma_2)^{1/2} + \sigma_2]. \tag{9}$$

The bounds for the polycrystals, and the envelopes of the realizable EM ellipsoidal aggregates over all possible values of depolarization factors L_1, L_2, L_3 for a number of uniaxial basic crystals are collected in table 1 and plotted in figures 1 and 2. Here σ_{int}^u denotes the upper envelope of those effective properties σ_e determined by (8), i.e. the greatest effective conductivity from all those of EM integranularly random ellipsoidal models with all possible values of L_1, L_2, L_3; σ_{int}^l is the lower envelope of the effective properties determined by (8) (the smallest one); σ_{com}^u and σ_{com}^l are the upper and lower envelopes of the effective conductivities σ_e of EM completely random ellipsoidal geometries determined by (7).

We observe that the uncertainty intervals (through comparisons of both the bounds and the envelopes) for intergranularly random polycrystals are substantially larger than those for

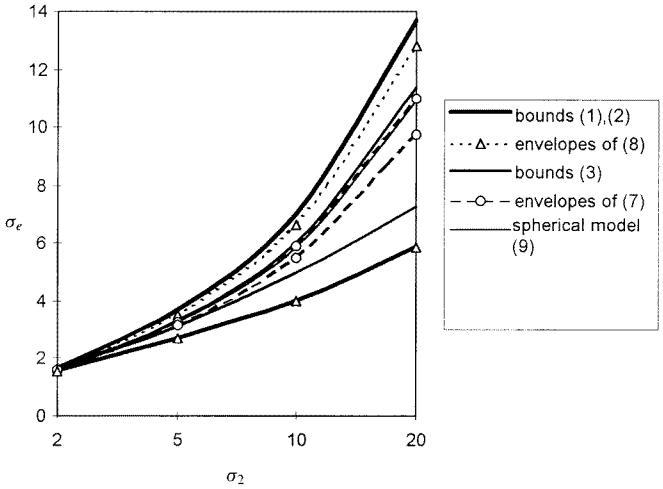

Figure 2. Bounds and envelopes of realizable models for aggregates of uniaxial crystals: σ_1 is normalized to be 1, $\sigma_2 = \sigma_3 = 2 \to 20$; The upper (1) and lower (2) bounds compared with the upper and lower envelopes of (8) for integranularly random polycrystals; the upper and lower bounds (3) compared with the upper and lower envelopes of (7) for completely random polycrystals.

Table 1. Bounds and envelopes of realizable models for random aggregates of uniaxial crystals at the ranges of normalized principal conductivities $\sigma_1 = 1 \to 20$, $\sigma_2 = \sigma_3 = 1 \to 20$; σ^u_{av} and σ^l_{SACLM} are the upper and lower bounds for isotropic aggregates; σ^u_{int}, σ^l_{int} are the upper and lower envelopes of EM intergranularly random ellipsoidal aggregates; σ^u_P, σ^l_P are the upper and lower bounds for symmetrically random polycrystals; σ^u_{com}, σ^l_{com} are the upper and lower envelopes of EM completely random ellipsoidal aggregates; σ_s is the conductivity of the EM spherical polycrystal.

σ_1	$\sigma_2 = \sigma_3$	σ^u_{av}	σ^u_{int}	σ^u_P	σ^u_{com}	σ_s	σ^l_{com}	σ^l_P	$\sigma^l_{int} = \sigma^l_{SACLM}$
2	1	1.333	1.306	1.282	1.282	1.281	1.279	1.279	1.236
5	1	2.33	2.08	1.91	1.88	1.85	1.84	1.78	1.53
10	1	4	3.22	2.82	2.6	2.5	2.47	2.17	1.71
20	1	7.33	5.35	4.56	3.71	3.42	3.38	2.5	1.83
1	2	1.667	1.646	1.619	1.619	1.618	1.615	1.615	1.56
1	5	3.67	3.52	3.3	3.28	3.27	3.18	3.12	2.7
1	10	7	6.62	6	5.89	5.85	5.5	4.96	4
1	20	13.67	12.8	11.35	11.01	10.92	9.73	7.28	5.84

completely random aggregates. It is interesting to see that the bound (2) coincides with the conductivity σ_e of the EM intergranularly random platelet ($L_1 = L_2 = 0$, $L_3 = 1$) aggregate (8), for a uniaxial basic crystal. Thus, in that case, both the lower estimates for isotropic polycrystals and the smaller class of intergranularly random aggregates are identical and optimal (the bound is attained by a specific model). Note that the respective isotropic optimal model of Schulgasser [4] is locally-ordered, while the platelet model (8) belongs to the smaller but realistic class of intergranularly random polycrystals. Comparisons of numerical results indicate that the bounds and the envelopes of realizable EM models for intergranularly random aggregates (as well as those for the completely random ones) do not differ much (the interval between the upper and lower bounds compared with the

interval between the upper and lower envelopes) when the differences between the principal conductivities $\sigma_1, \sigma_2, \sigma_3$ are not large (in other words, the anisotropy of the base crystal is not strong). Hence both the bounds and the envelopes can well approximate the uncertainty limits for the effective behaviour of the polycrystals at those ranges of the parameters. Similar observations have been made concerning the estimations for the overall behaviour of quasisymmetric randomly inhomogeneous composites [19].

3. Closure

The different EM hierarchical aggregates, the effective property of which can be determined exactly, reveal that the macroscopic conductivity of an intergranularly random (and completely random) polycrystal should scatter over some interval unlike the random cubic network of Kirkpatrick [2], the effective conductivity of which can be calculated and appears unique. The theoretical results indicate that the measured values of the macroscopic property of a random polycrystal should vary from sample to sample. Still, the scatter interval for a property of a polycrystal may be very small due to the weak anisotropy of most practical constituent crystals, so one can tabulate a sufficiently accurate approximate value of the aggregate's property for practical use. However small it was, this scatter interval is a reality of the physical world and deserves our attention. The available bounds predict that the scatter intervals should lie between them, but they cannot tell how small the scatter intervals really are. The realizable geometries considered perhaps partly give the answer; the scatter intervals are expected to be larger than those given by the envelopes of the models. The bounds together with the envelopes should give us estimations for the scatter intervals from both sides. It should be very desirable, but unfortunately we do not have the available experimental data on the observed scatter intervals for polycrystals' conductivity to compare with the theoretical results. Some available experimental data on the elastic constants of a number of cubic crystal aggregates collected in [21, 22] appears to fit well the respective bounds [23]. Though the bounds and the envelopes do not differ much when the base crystal is weakly anisotropic, they would differ much for strongly anisotropic base crystals. In such cases it is worth looking for more refined bounds and possibly new geometric models to narrow the bounds and to broaden the envelopes theoretically toward each other until they converge to optimal estimates. Here we have established the optimal lower estimate for the conductivity of intergranularly random aggregates of uniaxial crystals, where the lower bound (2) and the lower envelope of the constructed models (8) coincide.

References

[1] Hill R 1951 *Proc. Phys. Soc.* A **65** 349
[2] Kikrpatrick S 1971 *Phys. Rev. Lett.* **27** 1722
[3] Schulgasser K 1977 *J. Phys. C: Solid State Phys.* **10** 407
[4] Schulgasser K 1982 *Appl. Phys.* **54** 1380
[5] Avellaneda M, Cherkaev A V, Lurie K A and Milton G W 1988 *J. Appl. Phys.* **63** 4989
[6] Nesi V and Milton G W 1991 *J. Mech. Phys. Solids* **39** 525
[7] Pham D C 1996 *J. Appl. Phys.* **80** 2253
[8] Hashin Z and Shtrikman S 1963 *Phys. Rev.* **130** 129
[9] Avellaneda M and Bruno J 1990 *J. Math. Phys.* **31** 2047
[10] Helsing J 1993 *Proc. R. Soc. A* **443** 451
[11] Helsing J 1994 *Proc. R. Soc. A* **444** 363
[12] Bruggeman D A G 1935 *Ann. Phys.* **24** 636
[13] Kröner E 1958 *Z. Phys.* **151** 504
[14] Hill R 1965 *J. Mech. Phys.* **13** 213

[15] Elliot R J, Krumhansl J A and Leath P L 1974 *Rev. Mod. Phys.* **46** 465
[16] Landauer R 1978 *Electrical Transport and Optical Properties of Inhomogeneous Media* ed J C Garland and D B Tanner (New York: AIP)
[17] Milton G W 1985 *Commun. Math. Phys.* **99** 463
[18] Bergman D J and Stroud D 1992 *Solid State Phys.* **46** 147
[19] Pham D C 1997 *Phys. Rev.* E **56** 652
[20] Frank V 1963 *Electromagnetic Theory and Antenas* ed E C Jordan (New York: Macmillan)
[21] Warra H H, Koch H and Kröner E 1977 *J. Mater. Sci. Soc. Japan* **14**(2) 131
[22] Kröner E 1980 *J. Engng. Mech. Div.* **106** 889
[23] Pham D C 1997 *Philos. Mag.* A **76** 31